Materials Science & Engineering R 140 (2020) 100543

Contents lists available at ScienceDirect

Materials Science & Engineering R

journal homepage: www.elsevier.com/locate/mser

3D printing of hydrogels: Rational design strategies and emerging biomedical applications

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ARTICLE INFO

Keywords:
Additive manufacturing
3D printing
3D bioprinting
Biofabrication
Biomaterials
Hydrogels

ABSTRACT

3D printing alias additive manufacturing can transform 3D virtual models created by computer-aided design (CAD) into physical 3D objects in a layer-by-layer manner dispensing with conventional molding or machining. Since the incipiency, significant advancements have been achieved in understanding the process of 3D printing and the relationship of component, structure, property and application of the created objects. Because hydrogels are one of the most feasible classes of ink materials for 3D printing and this field has been rapidly advancing, this Review focuses on hydrogel designs and development of advanced hydrogel-based biomaterial inks and bioinks for 3D printing. It covers 3D printing techniques including laser printing (stereolithography, two-photon polymerization), extrusion printing (3D plotting, direct ink writing), inkjet printing, 3D bioprinting, 4D printing and 4D bioprinting. It provides a comprehensive overview and discussion of the tailorability of material, mechanical, physical, chemical and biological properties of hydrogels to enable advanced hydrogel designs for 3D printing. The range of hydrogel-forming polymers covered encompasses biopolymers, synthetic polymers, polymer blends, nanocomposites, functional polymers, and cell-laden systems. The representative biomedical applications selected demonstrate how hydrogel-based 3D printing is being exploited in tissue engineering, regenerative medicine, cancer research, in vitro disease modeling, high-throughput drug screening, surgical preparation, soft robotics and flexible wearable electronics. Incomparable by thermoplastics, thermosets, ceramics and metals, hydrogel-based 3D printing is playing a pivotal role in the design and creation of advanced functional (bio) systems in a customizable way. An outlook on future directions of hydrogel-based 3D printing is presented.

Abbreviations: APS, Ammonium persulfate; ASAP, Acrylic sodium salt polymer; ASCs, Adipose derived stem cells; BMP-2, Bone morphogenetic protein 2; CA, Citric acid; CEC, Carboxyethylcellulose; CMC, Carboxymethylcellulose; CPC, Calcium phosphate cement; DOPA, 3,4-Dihydroxyphenylalanine; DVS, Divinylsulfone; EC, Ethylcellulose; ECH, Epichlorohydrin; ECM, Extracellular matrix; EDC, 1-ethyl-3-(3-dimethylaminopropyl) carbodiimide; EGDE, Ethylene glycol diglycidyl ether; FDA, Food and Drug Administration; GelMA, Gelatin methacryloyl; GMHA, Glycidyl methacrylate-hyaluronic acid; GP, β-glycerol phosphate disodium salt; HEC, Hydroxyethylcellulose; HPMC, Hydroxypropyl methylcellulose; IL-4, Interleukin-4; IPN, Interpenetrating polymer networks; iPSCs, Induced pluripotent stem cells; KRSR, Lysine–Arginine–Serine–Arginine; LDHs, Layered double hydroxides; MC, Methylcellulose; MeHA, Methacrylated hyaluronic acid; Me_4N^+, Tetramethylammonium; NHS, N-hydroxysuccinimide; PAA, Poly(acrylic acid); PAAm, Poly(acrylamide); PDMAAm, Poly(*N,N*-dimethylacrylamide); PEDOT:PSS, Poly(3,4-ethylenedioxythiophene) polystyrene sulfonate; PEG, Poly(ethylene glycol); PEGA, Poly(ethylene glycol) monoacrylate; PEGDA, Poly(ethylene glycol) diacrylate; PEGMA, Poly(ethylene glycol) methacrylate; PEGDMA, Poly(ethylene glycol) dimethacrylate; PEO, Poly(ethylene oxide); PHEMA, Poly(2-hydroxyethyl methacrylate); PLA, Poly(lactic acid); PMO, Periodic mesoporous organosilica; PNaAMPS, Poly(2-acrylamido-2-methyl-1-propanesulfonic sodium); PNIPAAm, Poly (N-isopropylacrylamide); PPO, Poly(propylene oxide); PVA, Poly(vinyl alcohol); PVME, Poly(vinyl methyl ether); PVP, Polyvinylpyrrolidone; RGDS, Arginine–Glycine–Aspartic Acid–Serine; SWCNT, Single-wall carbon nanotubes; sulfo-SANPAH, Sulfosuccinimidyl 6-(4′-azido-2′-nitrophenylamino) hexanoate; TEMED, *N,N,N′,N′*-tetramethylethylenediamine; TEMPO, 2,2,6,6-tetramethylpiperidine-1-oxyl radical; TPP, Sodium tripolyphosphate; VEGF, Vascular endothelial growth factor

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https://doi.org/10.1016/j.mser.2020.100543
Received 18 November 2019; Received in revised form 30 December 2019; Accepted 2 January 2020

1. Introduction

Three-dimensional (3D) printing was first introduced in 1986 as sterolithography, in which laminae of a fluid material can respond to ultraviolet light and be successively printed in layers to generate solid 3D objects [1]. Conceptually, 3D printing, alias additive manufacturing or rapid prototyping, is a versatile layer-by-layer fabrication technology of 3D objects through progressive adding of materials (inks) directly guided by predefined digital models [2–4]. Since its first introduction, 3D printing technology has steadily increasing impact on society and is progressively transforming the science and engineering of advanced materials [5–9]. One good example is 3D bioprinting as a relatively younger field has rapidly developed over the past ten years due to the advances in 3D printing, cell biology, and materials science [10–16]. 3D printing allows for the direct production of customized complex components from metals [17–28], ceramics [29–34], glass [35–40], polymers [41–47], and multimaterials [48–50] through computer-aided design/computer-aided manufacturing (CAD/CAM). In comparison with conventional formative and subtractive manufacturing technologies that typically require molds, tooling and machining, 3D printing is much more flexible, less wasteful in materials and processes, and thus economically favorable in time cost and financial expenditure [7,51–53]. By virtue of CAD/CAM, 3D printing can enable the inclusion of more complex elaborate substructures [54]. Design mistakes can be identified at an earlier validation stage of product development through more efficient creating and amending of 3D models, thereby avoiding expensive and cumbersome amendments at a later stage [55,56].

For these reasons, 3D printing is considered as the next global industrial and manufacturing revolution [57–61] and finding wide applications in a range of industrial sectors such as aerospace [62–64], automotive [65], robotics [66–68], energy [69–73], education [74–77], food [78–80], chemical [6,81–84], pharmaceutical [85–91], biomedical [92–99], etc. For instance, 3D printing can be put into use for modeling complex protein interactions and customizing laboratory reactionware for chemical synthesis and analysis [100–105]. Multiscale electronics can be printed to order, just as printed circuit boards, thereby leading to a new era of printable robotics or printable electronics [106–110]. Currently, polymers are the most widely used materials for 3D printing to manufacture custom-made components [41]. To fully unleash the huge potential of 3D printing, new printable inks made of different materials are needed, and this requires on-demand design and development of the composition, structure, function, and dynamics of the ink materials [111–118].

With the accelerating population ageing tendency and prolonged life expectancy, there is an increasingly high demand for tissue and organ grafts and synthetic biomaterials that hold great potential to replace, repair or regenerate lost, injured or diseased tissues and organs [119–131]. New-generation biomaterials for tissue and organ regeneration should be bioresorbable, bioactive and mechanically robust [132–138]. Scaffold-based strategies hold great promise for tissue engineering and regenerative medicine due to the existent 3D porous architectures for cell ingrowth and matter transport, which will facilitate new tissue formation and material biodegradation [139–147]. For this reason, increasing efforts have been focused on the design and development of novel 3D porous scaffolds for tissue and organ regeneration. In the recent decades, 3D printing technology has made considerable progress in tissue engineering and is opening up the possibility of fabricating patient-specific scaffolds/constructs with defined properties [10,148–152]. 3D printing technology has been transforming the field of tissue engineering and regenerative medicine by providing a tool that enables unprecedented control, flexibility, speed, and precision over conventional manufacturing techniques. A vital yet limiting aspect of the design and application of 3D printing is the selection of suitable materials to be used as biomaterial inks [153–155]. Polymer hydrogels, i.e. highly hydrated 3D polymer networks, are one of the most feasible classes of ink materials for creating 3D porous scaffolds [156–158], as they can mimic extracellular matrix (ECM) and modulate cell fate [159–163]. Meanwhile, hydrogel networks can also facilitate matrix remodeling, cell migration, and cell adhesion in 3D environment that are required for the normal development of functional tissues [164–173]. Furthermore, the behaviors of cells on/in these printed scaffolds can be regulated by the material, physical, and chemical properties of the hydrogel networks [174–182]. Although significant successes have been achieved in designing single component hydrogels e.g. alginate- and gelatin-based ink formulations for 3D printing applications [183,184], the lack of a larger variety of printable hydrogel systems has been identified as one major drawback that limits the rapid advancement of this field [111,185,186]. In addition, for 3D printing of cell-laden hydrogels, i.e. 3D bioprinting, the cell-laden bioink formulations will be more stringent than biomaterial inks [185,187–191]. At the same time, 4D printing and 4D bioprinting that are based on the 3D printing of stimuli-responsive materials particularly hydrogels, have been recently introduced and rapidly developed as a new generation of technology [192–199].

This review aims to be a comprehensive, authoritative, critical, and accessible review of general interest to the chemistry and materials science communities because 3D printing of hydrogels can combine the advantages of both additive manufacturing and hydrogel materials: (i) Additive manufacturing offers the unique advantages of precision, customization, and the usage of different materials across different scales in predefined 3D organization with high degree of automation and reproducibility, allowing for fabricating patient-specific constructs with customized properties; (ii) Hydrogels have tailorable material, physical, chemical, and biological properties, hold great promise for various biomedical applications since they can mimic/recapitulate the complex ECM, and allow for directing cell fate in 3D scaffold environment as well as 3D printing with cells together. The choice of hydrogels or hydrogel-forming polymers as 3D printable inks requires the unprecedented control over their material, physical, chemical, and biological properties e.g. degradability, processability, biocompatibility, mechanical strength, gelation mechanism, ease of chemical modification, composition and architecture, structure and dynamics, as well as other functional properties e.g. electrical, optical, thermal or stimuli-responsive properties. Furthermore, 3D bioprinting involves additional complexities e.g. the choice of bioinks, cell types, bioactive molecules, as well as technical challenges relevant to the sensitivity of living cells and the complex construction of tissues and organs. The gap between rapidly advancing 3D printing technology and limited printable hydrogel systems has made the design and development of printable hydrogels emerge as an active research direction in this field. The scope of this comprehensive review ranges from an overview of the most important 3D printing techniques for hydrogels, the design and printability of hydrogels or hydrogel-forming polymers with a focus on materials, to the selection and development of advanced biomaterial inks and bioinks together with the summary of representative achievements in 3D printing/bioprinting for wide biomedical applications. Our goal is to deliver a comprehensive overview on the hydrogel-based 3D printing and 3D bioprinting that can be used as a toolbox for a variety of researchers (polymer chemists, material scientists, etc.), engineers, students, and other end-users with a general background information of basic design principles of printable hydrogel-based ink materials for 3D printing/bioprinting from a materials science perspective in a way like Materials Genome Initiative (MGI): prediction/design first, validation experiments next. It is hoped that this review work will be helpful to understand the current status of the 3D printing of hydrogel materials, direct the rational design of advanced biomaterial inks and bioinks, and accelerate the advancement of the field of hydrogel-based 3D printing and 3D bioprinting.

Although there are already a great number of studies that have been dedicated to the investigation of hydrogel-based 3D printing and 3D bioprinting and some excellent reviews that have been focused on relevant specific topics [45,111,156–158,186–191,200–202], we noted

that a comprehensive review in this field is still absent. Therefore we will summarize the recent advancements and future perspectives for the design and applications of hydrogel-based 3D printing/bioprinting in biomedical fields. In this review, we will first indroduce the most important and best established techniques for 3D printing of hydrogels, including laser-based, extrusion-based and inkjet printing. Then we will summarize in detail the most representative hydrogel systems from a materials science perspective, aiming to elucidate the tailorability of material, physical, chemical, and biological properties of hydrogels or hydrogel-forming polymers and guide the rational design of advanced hydrogel materials for 3D printing. Thereafter, we will highlight the state-of-the-art design strategies for developing advanced biomaterial inks/bioinks and the recent advancements of fabricating complex 3D functional hydrogel scaffolds or tissue-like constructs with the versatile 3D printing/bioprinting techniques. Following this, 4D printing/bioprinting, which combine stimuli-responsive hydrogel materials and advanced 3D printing/bioprinting techniques, will be introduced in detail. Afterwards, we will discuss the emerging representative biomedical applications regarding the 3D printing of hydrogels, including tissue regeneration, cancer research, in vitro tissue modeling, surgical preparation in clinic, soft robotics, and soft wearable/printable electronics. Finally, a prospect on the future research directions of 3D printing of hydrogel materials is provided.

2. 3D printing techniques for hydrogels

2.1. Overview

There is a primary classification of various 3D printing techniques for biomedical applications based on their working principles: (i) Laser-based systems by virtue of photopolymerization pathway, (ii) Nozzle-based systems through extrusion of (pre)polymers, and (iii) Printer-based systems by material and binder jetting. For more detailed information of the classification, we refer the readers to some excellent reviews [6,12,41,150,157,186]. Nevertheless, not all the 3D printing techniques are applicable to process hydrogel materials since the processing of hydrogel materials needs mild conditions rather than harsh conditions and therefore, it is not the objective of this review to discuss all of these 3D printing techniques. Since this review will focus on probing into the material design strategies that have been and/or can be developed for 3D printing of hydrogels, here we focus on the most important and best established techniques for 3D printing of hydrogels, including laser-based, extrusion-based, and inkjet printing. In the following sections, these 3D printing techniques will be discussed with comparison in detail.

2.2. Laser printing

Laser-based techniuqes are generally applicable to 3D print hydrogels [186,203] with the exception of selective laser sintering/melting that is suitable for processing polymer, metal, or ceramic powders [204,205]. They work by sequential deposition of light energy in predefined patterns; in such a way only photo-crosslinkable prepolymers can be applied to print crosslinked hydrogels.

2.2.1. Stereolithography

Stereolithography is the first commercialized 3D printing technique that was developed by Chuck Hull in 1986 [1]. A stereolithography setup is made of a container that holds photocurable liquid resin, a laser source (usually UV light) that induces the polymerization and crosslinking of liquid resin, a system that permits the horizontal plane (X- and Y- directions) movement of laser beam, and a system that controls the vertical plane (Z-direction) movement of fabrication platform. Fig. 1 depicts the main parts of a stereolithography setup and how they assemble together to allow for the additive layer-by-layer fabrication process. Irradiating the surface of photocurable liquid resin in a 2D pattern makes the resin solidify thru absorbing single photon to a predefined depth that is typically greater than the step height of fabrication platform. As result of this, unreacted functional groups on the first layer after photopolymerization will polymerize with the irradiated liquid resin in subsequent layer. The fabrication platform moves layer by layer in the Z-direction after curing each layer of resin to create the solidified 3D construct with a resolution of 30 μm [206,207]. Due to the incomplete conversion of reactive groups, post treatments of washing-off residual resin and curing with UV light are usually needed to promote mechanical property. For more detailed information of commonly used photoinitiators in stereolithography, we refer the readers to some excellent reviews [207,208].

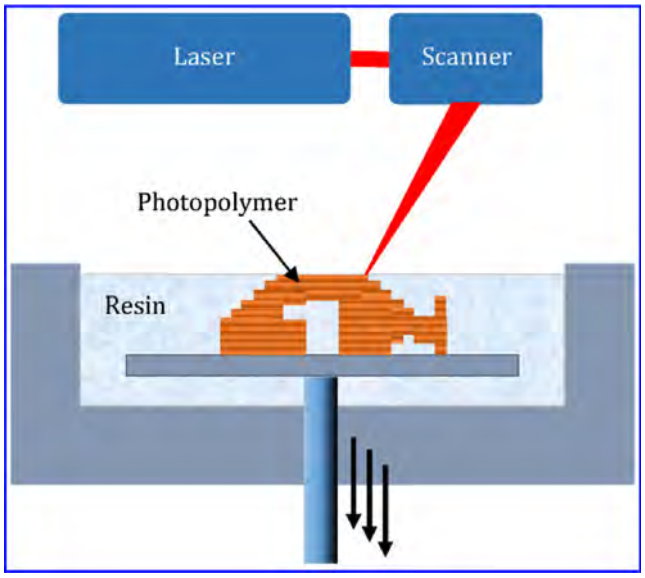

Fig. 1. Illustrative scheme to introduce the configuration of stereolithography. Reproduced with permission [43]. Copyright 2015, Academy of Dental Materials.

2.2.2. Two-photon polymerization

This technique uses near-infrared femtosecond laser pulses in the focal volume particularly titanium:sapphire laser at 800 nm wavelength to initiate the polymerization of a photosensitive material [158,209]. In the focal point of the laser beam, a suitable photoinitiator resin can simultaneously absorb two photons of 800 nm wavelength within a small volume in the photosensitive resin and let them serve as one photon of 400 nm wavelength (UV light region) to trigger the chemical reactions between photoinitiator molecules and monomers [210]. Some photoinitiators for the fabrication of hydrogels through two-photon polymerization have been summarized in previous work [211]. For more detailed principles of two-photon absorption, we refer the readers to some excellent reviews [212–214]. The nature of nonlinear excitation can make sure the photopolymerization reaction will be triggered only in the focal point of near-infrared laser beam thus without affecting other areas [213,215]. Fig. 2 illustrates the working principle of two-photon polymerization (2 P P) technique. As a result of the movement of laser focus, a 3D construct can be directly fabricated in the volume of photosensitive material with a resolution lower than the diffraction limit of the applied light.

2.2.3. Laser-induced forward transfer

The term laser-induced forward transfer was first coined in 1986 by Bohandy et al. [216]. The laser-induced forward transfer system mainly consists of three components, as depicted in Fig. 3: a pulsed laser that is focused onto a thin layer of metal or other laser-absorbing materials (e.g. hydrogels), a donor substrate (laser-transparent quartz or glass print ribbon) from which laser-absorbing materials are propelled, and

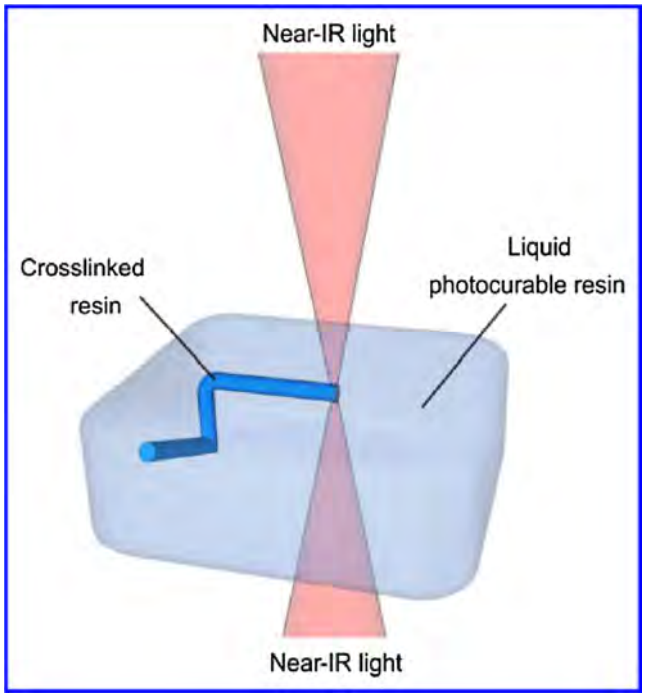

Fig. 2. Schematic diagram to elucidate the working principle of two-photon photopolymerization.
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an acceptor/receiving substrate for the inks. The laser-transparent donor is coated with a solid, liquid, or paste layer to supply the source of transfer material when laser pulses propagate thru the donor slide and get absorbed by the coating layer. Upon an incident energy above a specific threshold, the ink materials will be ejected from the coating and propelled toward the acceptor substrate. The movement of computer-controlled translation stages or galvanometric scanning mirrors can form complex high-resolution 2D and 3D patterns (10 ~ 100 μm resolution) from material droplets during the laser transfer process [217,218]. This laser transfer technique can be very applicabe for cell printing [203,219–221].

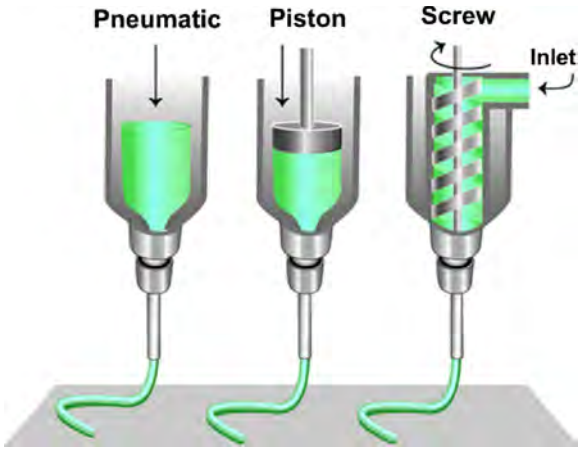

Fig. 4. Schematic diagram to illustrate the working principles of extrusion-based 3D printing technique.
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2.3. Extrusion printing

Extrusion-based 3D printing is the computer-controlled layer-by-layer deposition of molten/semimolten polymers, polymer solutions,

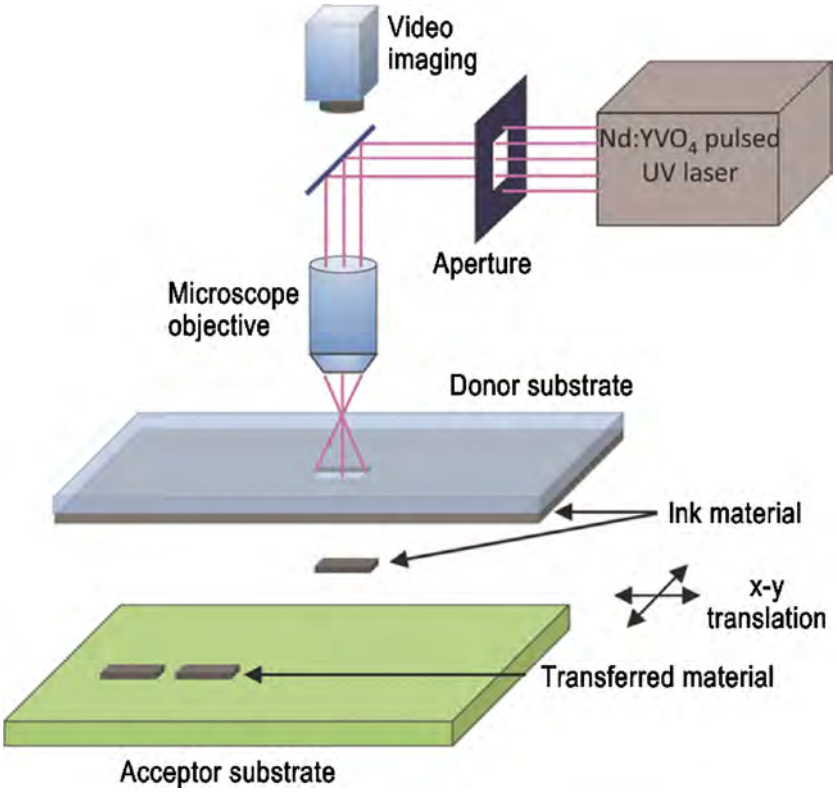

Fig. 3. Schematic image to illustrate the the main parts and process of laser-induced forward transfer technique.
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pastes, or dispersions thru a movable nozzle acting as the extrusion print head in a direct ink writing mode [41,222]. Fig. 4 reveals the different mechanisms of ejection of ink materials. Extrusion printing can be divided into two classes of melting-based process e.g. fused deposition modelling [223] and melt electrospinning writing [224] and dissolution-based process e.g. 3D plotting. Based on 3D dispensing that was first developed as AM method in 2000 [225], 3D plotting provides a versatile extrusion printing technique. In this 3D plotting process, the extrusion print head consists of a nozzle and a cartridge, which can horizontally (X- and Y- directions) and vertically (Z direction) move through a computer-controlled manner. 3D dispensing is pneumatically controlled by altering air pressure. This technique is very situable and attractive for scaffold fabrication and hydrogel processing [41,45,98,226]. The 3D objects are constructed in a laminar fashion thru computer-controlled deposition of ink materials on a stationary surface of fabrication platform with a resolution in μm to cm range. Generally, some stabilization strategies are necessary for the 3D plotting technique during/following printing procedure [227].

2.4. Inkjet printing

Inkjet printing is a non-contact reprographic technique that can acquire digital data from the computer and reproduce them onto a substrate through ink droplets [228]. Inkjet printers can be classified into the drop-on-demand jetting system and continuous inkjet system. In the continuous ejection system, continuous ink comes out of a print head (nozzle) under pressure to generate a jet. Then the jet can break up into droplets with electrical signals controlling the direction of movement [229]. In the drop-on-demand jetting system, an actuator can create pulses and lead to the ejection of an individual droplet with predefined ink volume [230] through piezoelectric or thermal heads [205], as illustrated in Fig. 5. For more detailed principles of inkjet printing, we refer the readers to some excellent reviews [231–234]. Inkjet printing is seen as a key technique in the field of customized polymer deposition [228,235]. This technique is well suitable for manufacturing complex scaffolds because of its capability to print multiple materials into constructs with high fidelity and resolution of 50 ~ 500 μm [151].

3. Polymer hydrogels

3.1. Overview

A gel refers to a solid jelly-like material that can have properties ranging from soft and weak to hard and tough. A polymer hydrogel is a 3D cross-linked network of flexible polymer chains that contains a large amount of water but retains the properties of solids [172]. The 3D network enables the liquid retention to form a swollen gel phase and the liquid inside can prevent the polymer network from collapsing, which imparts characteristics similar to those of soft tissues. At the gel point, a polymer sol or solution can abruptly change from viscous liquid state to solid phase hydrogel (sol–gel transition) [236]. Fig. 6 shows the classification of polymer hydrogels according to the synthetic methods, physical properties, polymer source, ionic charge, degradation rate and cross-linking types. A variety of polymers have been used to synthesize hydrogels, which include natural polymer hydrogels, synthetic polymer hydrogels and derivative hydrogels [168,172,237]. The general methods to synthesize polymer hydrogels are summarized in Table 1. The potential applications of hydrogels reported include tissue engineering [168,175], regenerative medicine [164,165,179], delivery systems (drugs, proteins, genes, cells) [166,238–241], synthetic extracellular matrix [177,180,242], implantable devices [243,244], biosensors [245–247], chemical sensors [248], diagnostic devices [169], separation systems [249], microfluidics [250], soft electronics [167], actuators [251], and so on.

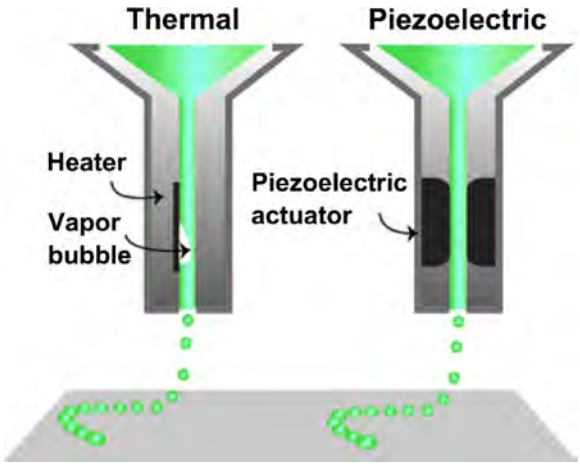

Fig. 5. Schematic diagram to show the working principles of inkjet printing technique.
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3.2. Natural polymer hydrogels

As shown in Fig. 6, according to the polymer source, hydrogel-forming polymers can be classified into natural polymers and synthetic polymers. Natural polymers are also referred to as biopolymers because they are derived from all organisms including human, animals, plants and bacteria [45,267,268]. Table 2 summarizes the biopolymers used for hydrogel formation including polysaccharides (sugars linked by O-glycosidic bonds), glycosaminoglycans (polysaccharides with amine functionality) and polypeptides/proteins. Glycosaminoglycans can naturally cover the surface of all eukaryotic cells and combine with various proteins to form natural extracellular matrix, thereby resulting in excellent biocompatibility and cell affinity [269].

3.2.1. Polysaccharide hydrogels

3.2.1.1. Agarose hydrogel. Agarose, present in red algae, is a linear polysaccharide with some ionized sulfate groups and one of the main components of agar [270,311]. It is water-soluble at temperatures above 65 °C and can gel in a range of temperatures from 17 °C to 40 °C below the gel-melting temperature 90 °C, depending upon the molecular weight and chemical modification of side groups. Once agarose gels, it is stable and does not swell at constant temperature or re-liquefy until heated to 65 °C [312]. The gelling mechanism of agarose lies in the formation of intermolecular hydrogen bonds upon cooling, thereby leading to the aggregation of double helices via the physical entanglement of anhydro bridges on individual molecules [313]. The viscoelastic properties of agarose hydrogel depend upon the molecular weight and solution concentration. The elastic moduli of physical gels are tunable from ~1 kPa to a few thousand kPa and well in the stiffness range of natural tissues [314]. Table 3 summarizes the features of agarose hydrogel.

3.2.1.2. Alginate hydrogel. Alginate is a water-soluble linear polysaccharide extracted from brown algae or bacteria [346]. It is generally regarded as safe and biocompatible by the U. S. Food and Drug Administration (FDA) [347] and can gel under benign conditions, making it attractive for cell encapsulation. The gelation can proceed through different mechanisms. At pH below 3, alginate self-assembles into acidic gels by forming intermolecular hydrogen bonds [348]. Alternatively, alginate forms a physical hydrogel by cooperative binding with divalent or trivalent cations such as Ca^{2+}, Mg^{2+}, Sr^{2+}, Ba^{2+} or Al^{3+} which can interact with carboxylic acid groups in the sugars [349–351]. As seen in Fig. 7, An alginate chain consists of mannuronic acid (M unit) and guluronic acid (G unit), and arranges in

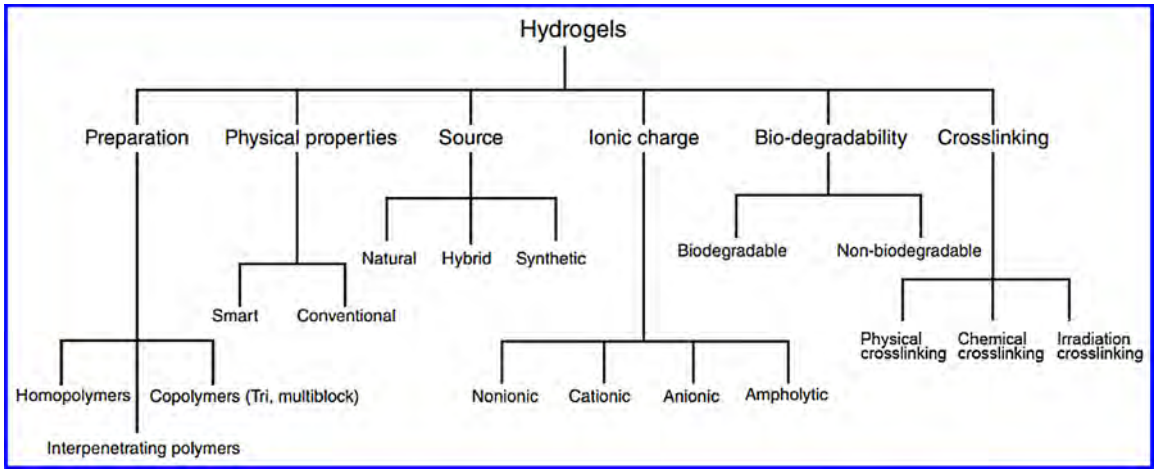

Fig. 6. Classification of polymer hydrogels.
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Table 1
Synthesis methods for polymer hydrogels.

	Cross-linking types	Methods	Examples	References
Physical hydrogels	Physical cross-linking	Heating polymer solution	PEG–PLA, PEO–PPO, PNIPAAm	[172,253,254]
		Cooling polymer solution	agarose, carrageenan, gelatin	[255,256,257]
		Ionic interaction	Na^+alginate$^-$ + Ca^{2+} + $2Cl^-$, Na^+alginate$^-$–polylysine, chitosan–polylysine, chitosan–TPP	[258,259,260,261]
		Freeze-thaw cycle	PVA, xanthan	[262,263]
		Complex coacervation	xanthan–chitosan	[264]
		Hydrogen bonding	CMC	[265]
Chemical hydrogels	Chemical cross-linking	Functional groups (OH, COOH, NH_2, etc.) react with chemical cross-linkers (aldehyde, bis-epoxide, etc.)	collagen–glutaraldehyde	[254]
	Radiation cross-linking	Exposure to high energy source (gamma ray, X-ray, electron beam, etc.)	PEG, PVME	[236,266]

blocks that are rich in G units, blocks that are rich in M units, and blocks that comprise alternating G and M units. The crosslinking density of alginate hydrogels is a function of the G blocks and molecular weight. The viscosity of alginate solution and its overall stiffness once gelled depend upon the polymer concentration and its molecular weight distribution [352,353]. Alginate hydrogels degrade slowly via a process involving loss of divalent ions into surrounding media and subsequent dissolution, in which their mechanical properties change with time. Table 4 summarizes the features of alginate hydrogel.

3.2.1.3. Carrageenan hydrogel. Carrageenan is a linear water-soluble polysaccharide extracted from red algae [267]. The isomers include κ-, ι- and λ-carrageenan, which differ in the position and number of the ester sulfate groups on the repeating galactose units [414]. The κ- and ι-carrageenans are composed of sulphated D-galactose and 3,6-anhydro-D-galactose, and λ-carrageenan is composed of sulphated D-galactose. The 3,6-anhydrogalactose is essential for the gelling properties of κ- and ι-carrageenans [274]. They can form thermoreversible hydrogels upon cooling of hot aqueous solutions containing various salts, which may take several hours at the gelation temperature. Nonionic solutes and colloids can affect carrageenan solubility by interacting with carrageenan or influencing water binding (Table 5) [415]. The strength of carrageenan hydrogels greatly depends on the carrageenan concentration [416,417] and monovalent cation type and concentration [418–421]. Nevertheless, cation concentration beyond a threshold (~0.2 M) can weaken the hydrogels [421,422]. For κ-carrageenan, it gels most strongly with the order of $K^+ > Ca^{2+} \gg Na^+$. K^+ forms rigid and elastic hydrogel while Ca^{2+} gives stiff and brittle hydrogel [423]. For ι-carrageenan, it gels most strongly with the order of $Ca^{2+} > K^+ > Na^+$. Ca^{2+} forms soft and resilient hydrogel [424]. Among all carrageenans, κ-carrageenan forms the strongest hydrogel. Table 6 summarizes the features of carrageenan hydrogel.

3.2.1.4. Cellulose hydrogel. Cellulose is the most abundant polysaccharide in the nature extracted from plants, natural fibers or bacteria [452,453]. Cellulose hydrogels can be prepared from native cellulose solution via hydrogen bonding [454]. However, cellulose is insoluble in water and other common solvents, so searching for appropriate solvents is the main issue for preparing cellulose hydrogels [455–457]. Water-soluble cellulose derivatives are synthesized through etherification of hydroxyl groups with methyl or ethyl units. Examples of these cellulose ethers include methylcellulose (MC), carboxymethylcellulose (CMC), hydroxypropyl methylcellulose (HPMC), ethylcellulose (EC), carboxyethylcellulose (CEC) and hydroxyethylcellulose (HEC), which have been used to prepare hydrogels via physical or chemical cross-linking [456,457]. MC is a semi-flexible linear-chain polysaccharide and has the most straightforward chemical composition with methoxy moieties partially replacing hydroxyl groups [275,458,459]. It is recognized as an acceptable food additive by the U. S. FDA [460]. In the context of this review, we will primarily focus on MC hydrogel. MC can gel from aqueous solution upon heating or salt addition [461–466], which enables it as a promising functional hydrogel for various biomedical applications [467–469]. Table 7 summarizes the features of cellulose hydrogel.

Table 2
Hydrogel-forming natural polymers.

	Biopolymers	Descriptions	Functionalities	References
Polysaccharides	Agarose	Neutrally charged, composed of D-galactose and 3,6-anhydro-L-galactose	ether and hydroxyl	[270]
	Alginate	Anionic polyelectrolyte, composed of D-mannuronate and L-guluronate	carboxylate and hydroxyl	[271,272]
	Carrageenan	Anionic polyelectrolyte, composed of D-galactose and 3,6-anhydro-D-galactose	hydroxyl and sulfate	[273,274]
	Methylcellulose	Neutrally charged, composed of D-glucose	hydroxyl	[275,276]
	Chitosan	Cationic polyelectrolyte, composed of D-glucosamine and N-acetyl-D-glucosamine	amine and hydroxyl	[277,278]
	Dextran	Neutrally charged, composed of D-glucopyranose	hydroxyl	[279]
	Gellan gum	Anionic polyelectrolyte, composed of D-glucose, D-glucuronic acid and L-rhamnose	carboxylate and hydroxyl	[280,281]
	Pectin	Anionic polyelectrolyte, composed of homogalacturonan, rhamnogalacturonan-I and rhamnogalacturonan-II	carboxylate and hydroxyl	[282,283]
	Pullulan	Neutrally charged, composed of maltotriose	hydroxyl	[284,285]
Glycosaminoglycans	Chondroitin sulfate	Anionic polyelectrolyte, composed of D-glucuronic acid and N-acetyl-D-galactosamine	amide, carboxylate, hydroxyl and sulfate	[286,287]
	Dermatan sulfate	Anionic polyelectrolyte, composed of D-glucuronic acid and N-acetyl-D-galactosamine	amide, carboxylate, hydroxyl and sulfate	[288,289]
	Heparin	Anionic polyelectrolyte, composed of D-glucuronic acid, D-glucosamine, L-iduronic acid and N-acetyl-D-glucosamine	amide, carboxylate, hydroxyl and sulfate	[290,291]
	Hyaluronic acid	Anionic polyelectrolyte, composed of D-glucuronic acid and N-acetyl-D-glucosamine	amide, carboxylate and hydroxyl	[292,293]
	Keratan sulfate	Anionic polyelectrolyte, composed of D-galactose and N-acetyl-D-glucosamine	amide, hydroxyl and sulfate	[294,295]
Polypeptides/proteins	Albumin	Amphoteric polyelectrolyte, composed of various amino acids	amine, carboxylate and hydroxyl	[296,297]
	Casein	Amphoteric polyelectrolyte, composed of various amino acids	amine, carboxylate and phosphate	[298,299]
	Collagen	Amphoteric polyelectrolyte, composed of various amino acids	amine, carboxylate and hydroxyl	[300,301]
	Elastin	Amphoteric polyelectrolyte, composed of various amino acids	amine and carboxylate	[302,303]
	Fibrin	Amphoteric polyelectrolyte, composed of various amino acids	amine, carboxylate and hydroxyl	[304,305]
	Gelatin	Amphoteric polyelectrolyte, composed of various amino acids	amine, carboxylate and hydroxyl	[306]
	Resilin	Amphoteric polyelectrolyte, composed of various amino acids	amine and carboxylate	[307,308]
	Silk	Amphoteric polyelectrolyte, composed of various amino acids	amine, carboxylate and hydroxyl	[309,310]

3.2.1.5. Chitosan hydrogel. Chitosan, structurally similar to glycosaminoglycans, is a linear polysaccharide prepared by N-deacetylation of chitin [278]. It is generally insoluble in neutral conditions but easily soluble in the presence of acid due to the protonation of free amino groups on glucosamine [492]. Meanwhile, many chitosan derivatives have been prepared to enhance the solubility and processibility [493–497]. Metal cations can be adsorbed on amine groups of chitosan through chelation in neutral solution [498,499]. Chitosan is approved nontoxic after oral administration in humans by the U. S. FDA [267], and has been widely explored for various biomedical applications [500–506]. Chitosan can form hydrogels by various ionic or chemical cross-linking methods [507,508]. Table 8 summarizes the features of chitosan hydrogel.

3.2.1.6. Dextran hydrogel. Dextran, produced by bacteria from sucrose or by chemical synthesis, is a highly water-soluble polysaccharide with some degree of branching via 1,3-linkage (branch point) [279,573,574]. It has been used in clinic for more than five decades for plasma volume expander, peripheral flow enhancer, and antithrombosis agent [267]. Dextran has excellent stability during prolonged storage under mild acidic and basic conditions [575]. The presence of large amount of hydroxyl groups enables it suitable for derivatization and subsequent physical or chemical cross-linking to form a hydrogel [576]. Table 9 summarizes the features of dextran hydrogel.

3.2.1.7. Gellan gum hydrogel. Gellan gum, produced by the aerobic submerged fermentation of bacteria, is a highly water-soluble linear extracellular polysaccharide (exopolysaccharide) [624–626]. It can undergo a thermally reversible coil to double helix transition and the double helical molecules aggregate to form the “junction zones”; this conformational transition is a prerequisite for the gelation of gellan gum [627–630]. The conformational transition temperature of gellan gum is around 30 °C and the temperature is strongly affected not only by the concentration of gellan gum but also by the presence of cations in aqueous solution [624,627,631–635]. Gellan gum can gel in water induced by cations (alkali metal, alkaline earth metal, tetramethylammonium (Me_4N^+)) or without added salts but with increasing polymer concentration. When adding cations, the gelation of gellan gum depends on both ionic strength and cation identity. For monovalent cations at ionic strength of 0.1 M, gel strength increases in the order: $Me_4N^+ < Li^+ < Na^+ < K^+ < Cs^+ < H^+$; For divalent cations the order is: Mg^{2+}, Ca^{2+}, Sr^{2+}, $Ba^{2+} < Zn^{2+} < Cu^{2+} < Pb^{2+}$ [628]. The lack of specificity among alkaline earth cations distinguishes gellan gum remarkably from other uronic acid containing polysaccharides such as alginate and pectin. For all the divalent

Table 3
Summaries of agarose hydrogel.

Chemical modifications	Cross-linking methods	Properties	Applications
Carboxylation [315,316] Methacrylation [317] EDC/NHS activation for coupling reactions [318,319,320,321] -CH_x- functionalization via sulfo-SANPAH [322,323] Modification by genipin [324]	Heat cross-linking [325]	Thermally reversible [326] Not biocompatible [327] Non-degradable [328]	Tissue regeneration: cartilage [323,329,330,331,332,333,334,335,336], cornea [337], enamel [255], nerve [312,318,321] Delivery system: drugs [338,339], growth factors [340], genes [341,342,343] Cancer therapy: ovarian cancer [344] Immunoengineering [345]

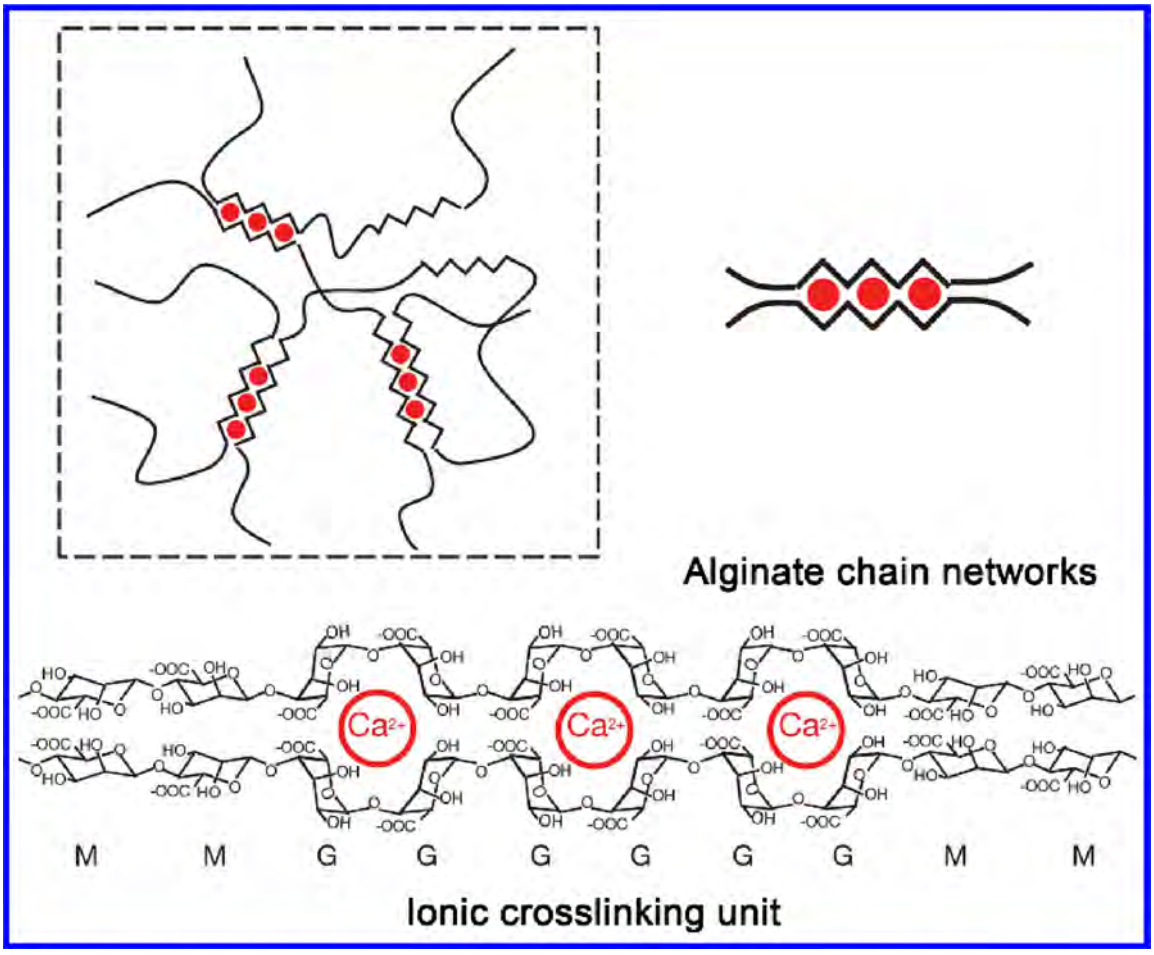

Fig. 7. Schematic of alginate hydrogel. In an aqueous solution, the G blocks in different alginate chains form ionic crosslinks through Ca^{2+} (red circles) and lead to the alginate hydrogel.
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Table 4
Summaries of alginate hydrogel.

Chemical modifications	Cross-linking methods	Properties	Applications
Carboxymethylation [355] Oxidation [356,357,358,359] EDC/NHS activation for coupling reactions [160,360,361,362,363,364,365] Tetrazine − norbornene functionalization via click chemistry [366] Modification by tetrazine [367] Sulfation [368]	Ionic cross-linking [369,370] Covalent cross-linking [371,372,373] Photo cross-linking [374,375] Thermal gelation [376] Cell cross-linking [377]	Biocompatible [271] Immunogenic relevant to L-guluronic acid [274] Degradable: hydrolysis, ion exchange/chelation [327]	Tissue regeneration: bone [378,379,380,381,382], cartilage [383,384], myocardial [385,386], nerve [387,388,389], cell encapsulation and transplantation [160,349,390,391,392,393,394] Wound healing [367,395] Delivery system: drugs [396,397,398], proteins [347,399,400,401], growth factors [402,403,404], peptides [405], genes [360], cells [406] Cancer therapy: ovarian cancer [407], tumor models [408,409] Immunoengineering [392,410,411,412,413]

cations, the ion-exchange affinity order is the same as the above one for increasing the gel strength. Generally, gellan gum hydrogel can be obtained with only 0.04 to 0.05 % (w/v) concentration in aqueous solution. Upon adding divalent cations, this content can be further decreased [625,636]. The hydrogel shows high clarity and even at a 15 % sugar content it is crystal clear. The melting point of hydrogel can be either below or above 100 °C, which allows for the design and preparation of both heat resistant hydrogel and hydrogel that should liquify during fabrication. Gellan gum has good compatibility with other gums/polymers and can be easily blended together [637]. In addition to native gellan gum, there are three types of modified gellan gum [630]: (i) High acetyl gellan gum (partially deacetylated) producing thermoreversible hydrogel, fairly soft, elastic, and non-brittle, (ii) Low acetyl gellan gum (highly deacetylated) generating firm and brittle hydrogel, and (iii) High clarity gellan gum (highly deacetylated and clarified). Table 10 summarizes the features of gellan gum hydrogel.

3.2.2. Glycosaminoglycan hydrogels

3.2.2.1. Chondroitin sulfate hydrogel. Chondroitin sulfate, the major component of aggrecan, is a water-soluble linear glycosaminoglycan that is covalently linked to proteins to generate proteoglycans, which is abundant in the articular cartilage [287,691]. It can stimulate the metabolic response of cartilage tissue [692]. It can play crucial roles in development process [693–696] and be involved in cell recognition, connection of ECM components to cell surface glycoproteins, and the

Table 5
Solubility of carrageenans [415].

Solvents	Types		
	κ-Carrageenan	ι-Carrageenan	λ-Carrageenan
Hot water	Soluble > 60 °C	soluble > 60 °C	soluble
Cold water	Na^+ salt soluble, K^+ and Ca^{2+} salts insoluble	Na^+ salt soluble, K^+ and Ca^{2+} salts insoluble	soluble
Hot milk	Soluble	Soluble	Soluble
Cold milk	Insoluble, swelling	Insoluble	Soluble
Concentrated sugar solution	Soluble, hot	Slightly soluble, hot	Soluble, hot
Concentrated salt solution	Insoluble	Soluble, hot	Soluble, hot
35 % ethanol solution	Insoluble	Insoluble	Na^+ salt soluble

intracellular signaling functions of varieties of growth factors and chemokines e.g. platelet-derived growth factors [697], fbroblast growth factors [698], epidermal growth factors [698], and transforming growth factor beta (TGF-β) [699]. Therefore, chondroitin sulfate has been widely investigated as a promising biomaterial for biomedical applications by physical or chemical cross-linking to develop hydrogels. In addition, a highly modified chondroitin sulfate chain at least at the uronic acid level is known as dermatan sulfate [287,696]. Table 11 summarizes the features of chondroitin sulfate hydrogel.

3.2.2.2. Heparin hydrogel. Heparin, only produced in mast cells, is a linear highly sulfated glycosaminoglycan that exists mainly in helical structure [290,742,743]. It has the highest density of negative charges among known biological macromolecules due to its high content of negatively charged sulfo and carboxyl groups. Heparin can bind to growth factors (e.g. bFGF), enzymes (e.g. antithrombin III), plasma proteins (platelet factor 4), and ECM proteins (e.g. fibronectin, laminin) [291,744–748]. Since its discovery, heparin has been widely used as an anticoagulant in clinic and modification of biomaterials with heparin has been extensively conducted for thrombogenicity reducing and drug delivery [749,750]. Heparin-based affinity delivery systems have demonstrated advantageous for the delivery of protein-based drugs e.g. growth factors [751]. In addition to the utilization for biomaterials modification, heparin has been recently used as a polymer matrix for hydrogel formation and its capability of binding to cationic polymers and basic peptides has been applied to promote the self assembly of hydrogels [752,753]. Table 12 summarizes the features of heparin hydrogel.

3.2.2.3. Hyaluronic acid hydrogel. Hyaluronic acid, alias hyaluronan, is a water-soluble linear non-sulfated glycosaminoglycan which can be found in almost every tissue in vertebrates [292,795–798]. In the body, hyaluronic acid can be degraded by free radicals or a family of enzymes called hyaluronidases, and then undergo endocytosis and lysosomal digestion [293,799]. It is actively involved in many important biological processes e.g. regulating cellular migration, proliferation, differentiation and angiogenesis and ECM organization and metabolism, and plays crucial roles in metastasis, inflammation, wound healing, and tissue repair [293,800–802]. Hyaluronic acid and its derivatives have been used for decades in clinic as medical products, and it has been increasingly recognized as important building block for producing functional biomaterials for tissue engineering and regenerative medicine [803–807]. Table 13 summarizes the features of hyaluronic acid hydrogel.

3.2.3. Polypeptide/protein hydrogels

3.2.3.1. Collagen hydrogel. Collagen is the most abundant protein in mammalian species and marine organisms [866]. It is widely available from bone, skin, tendon, cornea, ligament, intestine, and blood vessels, and makes up 1/3 of the total protein content in human body [300,867–869]. This fibrous structural protein consists of a right-handed triple helix of three parallel left-handed polypeptide strands (polyproline II-type helical conformation), exhibiting favorable mechanical properties [300,870–873]. Collagen is usually soluble in acidic aqueous solution and able to be processed into different forms e.g. powders, sheets, tubes, foams, sponges, hydrogels. Collagen is the most prevalent component of ECM and maintains its structural and biological integrity. Therefore, collagen biomaterial can create an ECM-mimetic microenvironment that is advantageous for promoting various cell functions (e.g. adhesion, migration, spreading, proliferation, differentiation, etc.) [126,868,874–876]. Various cross-linking methods have been developed to functionalize the molecular structure of collagen and optimise its properties for a wide range of biomedical applications [877]. Table 14 summarizes the features of collagen hydrogel.

3.2.3.2. Gelatin hydrogel. Gelatin is a class of proteinaceous substances that is derived from the parent protein collagen by heat denaturation and partial hydrolysis procedures [929], after which the backbone retains the bioactive sequences of collagen, thus able to promote cell functions e.g. adhesion, migration, proliferation, and differentiation [930,931]. In addition to the safe use for food processing (approved by

Table 6
Summaries of carrageenan hydrogel.

Chemical modifications	Cross-linking methods	Properties	Applications
Carboxylation [316] Methacrylation [424] Cyclization through alkali treatment [425,426,427] Acylation [428,429,430,431] Sulphation [428,429,432,433] Phosphorylation [428,429] Modification by genipin [434]	Heat cross-linking [435] photo cross-linking [424]	Thermally reversible [274,316,436,437] Apatite forming [436] Biocompatible [438] Inflammation inducing [439] Antitumor [274] Degradable: hydrolysis [440] κ-Carrageenan: syneresis-susceptible [415] ι-Carrageenan: thixotropic flow, good freezing-thawing stability, syneresis-free, less hysteresis (gelling/melting temperature interval) [415]	Tissue regeneration: bone [441], cartilage [442,443,444] Delivery system: drugs [445,446,447,448,449,450,451], growth factors [441,444]

Table 7
Summaries of cellulose hydrogel.

Chemical modifications	Cross-linking methods	Properties	Applications
TEMPO oxidation [470,471] Methacrylation [472]	Thermal gelation [473] Photo cross-linking [472] Chemical cross-linking: divinylsulfone (DVS) [474,475], carbodiimide [476], citric acid (CA) [477] Radiation cross-linking [478,479]	Thermally reversible [480,481] Biocompatible [456] Non-degradable [456]	Tissue regeneration: cartilage [482,483,484], brain [468], myocardial [466], nerve [485,486,487], liver [454], cell encapsulation and transplantation [488] Delivery system: drugs [489,490], enzymes [491]

Table 8
Summaries of chitosan hydrogel.

Chemical modifications	Cross-linking methods	Properties	Applications
Trimethylation [509,510] N-succinylation [511] Thiolation by EDC [512,513,514] EDC/NHS activation for coupling reactions [515] Azidation by EDC [516,517,518] Modification by sugar [519,520,521] Conjugation with catechol [522] Quaternized modification [523] Sulfation [524,525]	Ionic interaction: GP [526,527,528], TPP [259,529,530,531] Photo cross-linking [516,517,518] Chemical cross-linking: glutaraldehyde [532], sulfuric acid [533], APS/TMEDA [534,535], disulfide [536], genipin [537,538,539], ECH [540], EGDE [541]	Biocompatible, non-cytotoxic [542,543] Antibacterial [503,544,545,546] pH sensitive [547] Mucoadhesive [548] Degradable: hydrolysis [543], proteolysis (lysozyme, chitosanase) [549]	Tissue regeneration: bone [550,551], cartilage [534,552,553,554], vascular constructs [555,556], nerve [258,557,558], myocardial [559,560,561,562], liver [521], intervertebral discs [526], cell encapsulation and transplantation [563] Delivery system: drugs [537,564,565], proteins [259,530,536], growth factors [556], genes [566,567,568], antibiotics [531] Wound healing [516,517,569] Cancer therapy: lung cancer [570], liver cancer [571] Immunoengineering [572]

Table 9
Summaries of dextran hydrogel.

Chemical modifications	Cross-linking methods	Properties	Applications
Acrylation [577,578,579,580,581,582,583,584] Oxidation [585,586] Carboxymethylation [587] Tyramine functionalization [580,585,588,589,590] Thiol functionalization [591] Alkyne and azide modification [592]	Physical cross-linking: ionic interaction [593], stereocomplex formation [594,595,596] Chemical cross-linking: photo cross-linking [597], free radical polymerization [580,581,598], covalent cross-linking (isocyanate [599], glutaraldehyde [600])	Biocompatible [582,601] Non-cell-adhesive [602] Immunogenic [603] Degradable: hydrolysis [604], proteolysis (dextranase [605,606])	Tissue regeneration: cartilage [589,607], skin [608], vascular constructs [609,610], nerve [602], cell encapsulation [611] Delivery system: drugs [573,574,599,600,612,613,614], proteins [576,595,598,615,616,617], growth factors [609,618], liposomes [604], antibiotics [619,620,621], cells [622] Wound healing [618] Cancer therapy: breast carcinoma [623]

Table 10
Summaries of gellan gum hydrogel.

Chemical modifications	Cross-linking methods	Properties	Applications
Deacetylation [280,638] Methacrylation [639,640,641,642,643,644,645] EDC/NHS activation for coupling reactions [646,647] Oxidation [648,649] Carboxymethylation [650] Thiolation [651,652] Furan modification [653,654,655] Sulfation [433] Octadecylamine modification [656]	Physical cross-linking: ionotropic gelation with cations [627,657], ionic cross-linking with polyamines (spermine [658,659,660], spermidine [660,661]), addition of standard cell culture medium [662,663] Chemical cross-linking: covalent cross-linking (EDC [664,665])	Thermally reversible [657,666] Biocompatible [667] Non-cytotoxic [666,668] Poor-cell-adhesive [629] Degradable: hydrolysis [669], proteolysis (metalloproteinase [670])	Tissue regeneration: cartilage [641,648,657,666,671,672,673,674], bone [639,643,675], vascular constructs [670], adipose [676], nerve [653,654,677], intervertebral disc [642,644,678], spinal cord injury [655], cell encapsulation [679] Delivery system: drugs [672,680,681,682,683,684], proteins [685], peptides [686], antibiotics [687,688,689,690], enzymes [688]

U. S. FDA) [866], gelatin is widely applied to pharmaceutical industry [932] and tissue engineering and regenerative medicine field [184,933–935]. Gelatin can form physical hydrogel below 27 °C, but it is not stable at body temperature to be used as a biomaterial due to the reversible thermal gelation, and this can be overcome by various chemical modification and covalent cross-linking strategies [256,936,937]. Table 15 summarizes the features of gelatin hydrogel.

3.2.3.3. Silk hydrogel. Silk is an important class of protein materials that has been widely used by humans in ancient times. The sources of silk include silkworm silk and spider silk, which have been extensively studied to elucidate their processing mechanisms and thus exploit their properties for use as biomaterials [309,983–987]. Silk derived from silkworms mainly consists of sericin and fibroin proteins, which has shown huge potentials for wide applications in the fields of tissue engineering and regenerative medicine [310,988–990]. Silk fibroin is a

Table 11
Summaries of chondroitin sulfate hydrogel.

Chemical modifications	Cross-linking methods	Properties	Applications
Methacrylation [700,701,702,703,704,705,706,707,708,709,710,711] Oxidation [700,712,713,714,715] Thiolation [513,514,716,717] Sulfation [699,718] Azidation [719] EDC/NHS activation for coupling reactions [720,721] Furan modification [722] Fucosylation [723]	Physical cross-linking: complex coacervate gelation with polyelectrolyte cations [286] Chemical cross-linking: free radical polymerization [724]	Biocompatible, nonimmunogenic, pliable [327,720,725] Anti-inflammatory [692,726,727,728] Degradable: proteolysis (chondroitinase) [729]	Tissue regeneration: bone [706,730,731], nerve [732,733,734], cartilage [703,709,735,736], spinal cord injury [711] Delivery system: drugs [724,737], growth factors [738], antibiotics [739], genes [737], proteins [740] Wound healing [720,725,741] Arthritis treatment [728]

readily available protein from silkworms [991]. The spider web is made of the dragline silk, which mainly comprises two major ampullate spidroins (MaSp1 and MaSp2) [992]. The mechanical toughness of silk fibers is superior to any of the best synthetic fibers now available including Kevlar, and the spider dragline silk has the highest toughness among all types of silk [983,992–994]. Nevertheless, harvesting the mechanically strong silk spidroin is impractical and it lacks intrinsic bioactivity, which are the limitations of natural spider silk. To overcome this, researchers have focused on the recombinant spider silk proteins and silk-like polypeptides [995–1004]. Silk proteins can be processed to form hydrogels, sponges, foams, meshes, porous scaffolds, films/coatings, as well as fibers for diffferent biomedical applications [1005–1007]. Table 16 summarizes the features of silk hydrogel.

3.3. Synthetic polymer hydrogels

According to the types of synthesis method, synthetic polymer hydrogels are classified into: (i) Homopolymer hydrogels that consist of cross-linked network of one hydrophilic monomer unit (e.g. PAM, PNIPAM, PVA), (ii) Copolymer hydrogels that are generated by cross-linking of two co-monomer units (including one hydrophilic), (iii) Multipolymer hydrogels that are formed by reaction of three or more co-monomer units, and (iv) Interpenetrating polymer hydrogels that are prepared by forming a first network and then swollen in other monomer to generate a second intermeshing network [252,1060,1061].

3.3.1. Polyacrylate hydrogels

3.3.1.1. Polyacrylamide hydrogel. Polyacrylamide (PAAm) hydrogel can be synthesized using acrylamide monomer (AAm) in aqueous solution with adding *N,N′*-methylenebisacrylamide (MBAAm, cross-linking agent), ammonium persulphate (APS, photo/thermal initiator), *N,N*,N',N'-tetramethylethylenediamine (TEMED, crosslinking accelerator) [354,1062–1067]. PAAm hydrogel plays an important role in biomedical applications for studying cell mechanics [1068–1071] and plastic and aesthetic surgery e.g. as soft tissue fillers and augmentation materials [1072–1074]. Table 17 summarizes the features of polyacrylamide hydrogel.

3.3.1.2. Poly(N-isopropylacrylamide) hydrogel. Poly(N-isopropylacrylamide) (PNIPAAm), a chemical isomer of poly-leucine, has a polar peptide group in the side chain rather than in the backbone. PNIPAAm is the most studied synthetic responsive polymer, which undergoes a sharp coil–globule transition in aqueous solution at 32 °C, the Lower Critical Solution Temperature (LCST), and changes from a hydrophilic state below LCST to a hydrophobic state above LCST [253,1092]. When heating PNIPAAm aqueous solution to 32 °C, the polymer will precipitate from the clear solution and phase separation occurs to produce a two-phase system, in which the polymer-rich phase (globules) is insoluble in water. This temperature-induced sol–gel transition is a reversible process and thus forms the thermoreversible/thermoresponsive PNIPAAm hydrogel [1093], as well as leading to thermochromic, photochromic, and magnetochromic properties [1094–1097]. The LCST (32 °C) of PNIPAAm homopolymer is very close to human body temperature (37 °C) and can be tuned below or above the body temperature as desired by incorporating co-monomer units, which therefore make PNIPAAm-based materials particularly suitable for various biomedical applications [1098–1106]. For more detailed information of PNIPAAm, we refer the readers to some excellent reviews [253,1107–1113].

3.3.1.3. Sodium polyacrylate hydrogel. Sodium polyacrylate, alias acrylic sodium salt polymer (ASAP), is a sodium salt of polyacrylic acid (PAA), an anionic polyelectrolyte with negatively charged carboxylic groups in the main chain, which is well known as waterlock and has wide applications in consumer products e.g. cosmetics, infant diapers, and personal care products. This polymer can absorb as much as 100–1000 times its mass in water and therefore sodium acrylate is a monomer used to prepare superabsorbent polymers [1114,1115]. Using sodium acrylate and acrylamide co-monomers, along with MBAAm cross-linker, APS initiator, and TEMED accelerator, an expansion microscopy (ExM) has been developed to allow nanoscale resolution imaging of biological samples with conventional microscopes [1116–1124], and an implosion fabrication (ImpFab) process has been achieved to 3D print a range of materials with nanoscale feature sizes inside a hydrogel scaffold, thus developing a versatile 3D nanofabrication method [1125].

3.3.2. Poly(ethylene glycol) hydrogel

Poly(ethylene glycol) (PEG), alias poly(ethylene oxide) (PEO), is one of the best biocompatible and most widely used synthetic polymer hydrogels in medicine and biomedicine (approved by the U. S. FDA)

Table 12
Summaries of heparin hydrogel.

Chemical modifications	Cross-linking methods	Properties	Applications
Sulfation [754,755] Thiolation [756,757,758] EDC/NHS activation for coupling reactions [721,759,760,761,762] Methacrylation [763,764,765] Amidation [766,767] Tyramine functionalization [768] Oxidation [740]	Chemical cross-linking: photo cross-linking [763], PEG cross-linking [762]	Anticoagulant [290,769,770] Blood-compatible [768] Degradable: photodegradation [269,771], proteolysis (heparinase) [772]	Tissue regeneration: bone [773,774], cartilage [775,776], liver [777,778,779,780], vascular constructs [759,766,781,782], renal [783], cell encapsulation [784] Delivery system: drugs [754], growth factors [760,767,785,786], proteins [787,788,789] Wound healing [790,791,792] Cancer therapy: breast cancer [793], tumor models [794] Immunoengineering [759,760]

Table 13
Summaries of hyaluronic acid hydrogel.

Chemical modifications	Cross-linking methods	Properties	Applications
Esterification [808,809,810] Amidation [811,812] Methacrylation (MeHA) [374,813,814] EDC/NHS activation for coupling reactions [805,815,816,817] Oxidation [818] Sulfation [699,819] Thiolation [820,821] Pentenoate functionalization [822]	Physical cross-linking: ionotropic gelation with cations [823] Chemical cross-linking: photo cross-linking [374,801,811,815,824,825,826], click cross-linking [827], disulfide cross-linking [828], covalent cross-linking [829], carbodiimide cross-linking [830]	Angiogenic [831,832] Biocompatible [808,833] Cell-adhesive [834] Chondroprotective [293] Immunoprotective and immunomodulatory [835] Degradable: proteolysis (hyaluronidase, β-D-glucuronidase, β-N-acetylhexosaminidase) [293]	Tissue regeneration: bone [822,831,836,837], cartilage [819,838,839,840,841,842], intervertebral disc [843], liver [824], myocardial [824], nerve [844,845], retinal [846], spinal cord injury [847,848,849] Delivery system: cells [850], drugs [851,852,853,854,855], genes [851,856,857,858], growth factors [786,859,860], proteins [788,861,862] Arthritis treatment [795,796] Wound healing [720,863,864] Cancer therapy: thyroid cancer [865], tumor models [827]

[1126]. PEG itself is a very hydrophilic and hydrolytically non-degradable polymer of high solubility in water and many organic solvents [1127–1129]. It is well known for the excellent biocompatibility because the mobile, non-ionic, and highly hydrated features make PEG effectively repellent to both protein adsorption and cell adhesion [1130–1135]. Since PEG can only undergo limited metabolism in human body, the whole polymer chains are cleared through the kidney (< 30 kDa) or ultimately through the liver (> 30 kDa) [1136]. Therefore, typically, only PEGs with < 50 kDa molecular weight are considered for applications in tissue engineering and regenerative medicine to insure their complete clearance from human body [1137]. When drying, PEG can easily form crystals, which therefore can be readily measured with endothermic melting signals upon use for blends or copolymers [1138–1140]. The melting point depends on the chain molecular weight. For instance, at room temperature PEG 400 is a viscous liquid, whereas PEG 2000 and PEG 6000 are solid state materials of approximately 40 °C and 60 °C melting point, respectively. And the melting point and glass transition temperature of PEG blends or copolymers are tailorable. This kind of plasticizing effect of PEG can be used to modulate the mechanical properties of polymer biomaterials at human body temperature. For more detailed information of PEG, we refer the readers to some excellent reviews [1141,1142]. Other broadly used hydrogels based on PEG derivatives include poly(ethylene glycol) monoacrylate (PEGA) [1143,1144], poly(ethylene glycol) diacrylate (PEGDA) [1145–1150], poly(ethylene glycol) methacrylate (PEGMA) [1151,1152], and poly(ethylene glycol) dimethacrylate (PEGDMA) [1153–1155]. These PEG-based hydrogels play an important role for various biomedical applications. Table 18 summarizes the features of PEG-based hydrogels.

3.3.3. Poly(vinyl alcohol) hydrogel

Poly(vinyl alcohol) (PVA) is mainly synthesized by the polymerization of vinyl acetate. Its chemical structure is relatively simple with pendant hydroxyl groups [1209,1210]. PVA is a water-soluble linear polymer with chemical inertness in many organic solvents and optical transparency in UV–vis region. Pure or non-modified PVA can repel protein adsorption and cell adhesion, which thus contributes to its good biocompatibility for biomedical applications [1211,1212]. PVA must be cross-linked into hydrogels for a broad range of biomedical applications. Physical, chemical, and radiation cross-linking methods can be used to form PVA hydrogels. For example, PVA hydrogels can be produced by the alternating freezing and thawing cycles (freezing/thawing technique) due to crystallite formation, as first reported by Peppas in 1975 [262,1213], which is regarded as the most preferred approach for preparing PVA hydrogel without involving toxic chemical cross-linkers. The freezing/thawing treatment can make PVA crystallize rapidly from solution to form hydrogel network; increasing the number of freezing/thawing cycles (aging), additional crystallization can reinforce the original PVA hydrogel network [1214–1216]. Such physically cross-linked PVA hydrogels, of an elastic and rubbery nature [1217,1218], have higher mechanical strength than those formed by chemical and radiation cross-linking [1219]. For the more detailed information of PVA cryogels, we refer the readers to some excellent reviews [1220–1223]. PVA hydrogels are featured by their good mechanical property and water retention capability to provide a prolonged humid environment [1224]. Table 19 summarizes the features of poly (vinyl alcohol) hydrogel.

3.4. Composite hydrogels

The major drawback of single-component hydrogels from either natural or synthetic polymers is their lack of mechanical strength. They are mechanically weak, fragile, and brittle. Therefore, maintaining and improving the mechanical integrity is a key issue regarding hydrogel-based materials for biomedical applications. On the other hand, essentially, human tissues are composite materials possessing anisotropic

Table 14
Summaries of collagen hydrogel.

Chemical modifications	Cross-linking methods	Properties	Applications
Amination, acetylation, succinylation [878] Glutathione modification [879]	Physical cross-linking: self-assembling gelation [880,881], dehydrothermal treatment [882,883], ultraviolet irradiation [882,883] Chemical cross-linking: photo cross-linking [884], $NaHCO_3$ [885,886], carbodiimide [887], genipin [888], glutaraldehyde [889,890], tannic acid [891,892] Radiation cross-linking [893,894]	Biocompatible, cell-adhesive, hydrophilic, flexible, chemotactic, low antigenic [895,896] Immunomodulatory [897] Degradable: proteolysis (collagenase) [898,899]	Tissue regeneration: bone [884,900,901,902], brain [903,904], cartilage [905,906,907,908,909], liver [910], myocardial [911], nerve [912,913,914], renal [915,916], skin [886], tendon [917], vascular constructs [918] Delivery system: drugs [895,919], growth factors [920,921], peptides [922], proteins [923], antibiotics [924] Wound healing [925] Cancer therapy: tumor models [926,927,928]

Table 15
Summaries of gelatin hydrogel.

Chemical modifications	Cross-linking methods	Properties	Applications
Methacryloyl functionalization (GelMA) [933,938,939,940,941,942] Heparin functionalization [943]	Physical cross-linking: heat cross-linking, ultraviolet irradiation [944,945,946] Chemical cross-linking: photo cross-linking [947,948], EDC [934,949], formaldehyde [950], genipin [946,950,951], glutaraldehyde [937,952], PEG [953], pullulan dialdehyde [954], tannic acid [955,956,957], enzymatic cross-linking (microbial transglutaminase [958,959])	Biocompatible, limited antigenic [960], cell-adhesive [961] Water-soluble [962] Degradable: proteolysis (gelatinase [931], subtilisin [963])	Tissue regeneration: bone [947,964,965], cartilage [966,967,968,969,970], corneal [971], myocardial [972], retinal [973], skin [974] Delivery system: drugs [929,951,975,976], genes [975,977], growth factors [978], peptides [979], proteins [980] Cancer therapy: gastric cancer [981], tumor models [982]

Table 16
Summaries of silk hydrogel.

Chemical modifications	Cross-linking methods	Properties	Applications
Diazonium coupling [1008] Methacrylation [1009]	Physical cross-linking: hydrophobic interaction [1010], ultrasonication [1011,1012], vortex [1013] Chemical cross-linking: photo cross-linking [1009], enzymatic cross-linking (horseradish peroxidase [1014,1015,1016,1017])	Biocompatible [1018], cell-adhesive [1019], biodegradable [1020], low immunogenic [1021] Apatite-forming ability [996]	Tissue regeneration: bone [1022,1023,1024,1025,1026,1027,1028], cartilage [1014,1029,1030,1031,1032,1033], cervix [1034], intervertebral disc [1035], pancreatic islet [1036,1037], nerve [1038,1039], vascular constructs [1040] Delivery system: drugs [1041,1042,1043,1044,1045,1046], antibiotics [1047,1048], antibodies [1049,1050], genes [1051,1052], growth factors [1012,1053], proteins [1027,1050,1054] Wound healing [1055] Cancer therapy: breast cancer [1056], glioblastoma [1057], neuroblastoma [1058], tumor models [1059]

properties that rely upon the roles and structure arrangements of various components e.g. collagen, elastin, keratin, and hydroxyapatite of different tissues e.g. bone, cartilage, tendon, and skin. To replace/repair/regenerate the natural host tissues with hydrogels, hydrogel materials should simultaneously possess an appropriate elastic modulus and high mechanical strength [1267–1271]. This can be achieved using the strategy of composite hydrogels. Through tuning the proportion and arrangement of reinforcement phase, the properties of composite hydrogels can be tailored to match the mechanical and physiological requirements of different host tissues.

3.4.1. Polymer composite hydrogels

Here we mainly focus on composite hydrogels prepared from the double-component blends of natural and synthetic polymers. Brifely, such blends include: (i) Natural–natural polymer hydrogels, (ii) Natural–synthetic polymer hydrogels, and (iii) Synthetic–synthetic polymer hydrogels. Table 20 summarizes some examples of the double-component polymer composite hydrogels.

Interpenetrating polymer networks (IPN) are the unique “alloys” of cross-linked polymers [1380], among which at least one network is synthesized/cross-linked in the existence of the other network. They are held together only by permanent topological interlocking interactions without covalent bonds in between, thus not able to separate them from each other without breaking their own cross-linked networks [1380–1384]. From the point of preparation chemistry, IPN hydrogels are classified into: (i) Sequential IPN hydrogels, i.e. one hydrogel network is polymerized and formed in the presence of the other swollen single-network hydrogel, and (ii) Simultaneous IPN hydrogels, i.e. the monomers/prepolymers of both hydrogel networks are mixed and synthesized at the same time by two independent, non-interfering polymerization routes. When one network comprises an uncross-linked linear polymer, it is a semi-IPN hydrogel; otherwise with cross-linking, it is a full-IPN hydrogel. For the more detailed information of IPN hydrogels, we refer the readers to some excellent reviews [1385–1387]. Mechanically reinforced double-network (DN) hydrogels are a unique type of IPN hydrogel, as first described by Gong et al. in 2003 [1388]. They can exhibit an impressive nonlinear enhancement of the mechanical strength and have attracted increasing attention for use as biomaterials for tissue replacement particularly for load-bearing tissues [961,1388–1393]. In such DN hydrogels that are featured by high toughness, high fracture strength, high resistance to wear, and high water content (ca. 90 wt%), the first network is a tightly/abundantly ionically cross-linked hydrogel as the minor component (rigid and brittle skeleton), which is interpenetrated with the second network, a loosely/poorly crosslinked neutral hydrogel as the major component (soft and ductile polymer), and the preparation should be performed in that order to achieve their high mechanical strength [354,1064,1394–1398]. Recently, a universal strategy was developed to convert the polymer composite hydrogels into the DN hydrogels with prominent mechanical properties by a facile soaking treatment, which may aid in the fabrication of various DN hydrogels for a wide range of biomedical applications [1399].

3.4.2. Nanocomposite hydrogels

Organic–inorganic hybrid nanocomposites are omnipresent in the nature such as bone, tooth, and nacre, which comprise biomacromolecules

Table 17
Summaries of polyacrylamide hydrogel.

Chemical modifications	Cross-linking methods	Properties	Applications
2-Pyridinecarboxaldehyde functionalization [1075] Protein conjugation [1069] Mannich reaction [1076,1077] Hofmann reaction [1078,1079]	Chemical cross-linking: free radical polymerization [354,1080,1081]	Non-toxic, non-inflammatory [1061,1082] Non-degradable, stable in natural environment [1083] Tunable substrate mechanics [1084]	Delivery system: drugs [1085] Wound healing [1086] Facial corrections [1087] Ionic skin [1063] Stretchable and compressible electronics [1088,1089] Actuators and artificial muscles [1090,1091]

Table 18
Summaries of PEG-based hydrogels.

Chemical modifications	Cross-linking methods	Properties	Applications
Derivative synthesis [1127,1156] Bioactive modification [1157] Thiol − ene reaction [1158] Heparin functionalization [774,793] Azidation [1159] DOPA modification [1160]	Physical cross-linking: laponite [1161,1162] Chemical cross-linking: photo cross-linking [1163,1164,1165], click cross-linking [180,1166,1167,1168,1169]	Biocompatible, non-toxic [1156] Low inflammatory [1170] Degradability: (i) PEG: non-degradable [1141]; (ii) derivatives: photodegradable [1171,1172,1173], hydrolysis [1174], proteolysis [1175] Tunable substrate mechanics [826,1176,1177]	Tissue regeneration: bone [774,1178,1179], cartilage [703,821,1180,1181,1182,1183,1184], nerve [753,771], muscle [1175], corneal [1185], liver [1186], kidney [1187], adipose [1188], cell expansion [1189], vascular constructs [1190], cell encapsulation [1191,1192] Delivery system: drugs [1136,1193,1194,1195,1196,1197], cytokines [1198], proteins [1199,1200,1201,1202], growth factors [1203,1204] Wound healing [1205,1206] Cancer therapy: tumor models [539] Biosensors [1207,1208]

(proteins) and inorganic nanomaterials (minerals) with outstanding mechanical strength [1400–1402]. The insufficient mechanical strength of polymer hydrogels has limited their otherwise extensive applications for different biomedical purposes. To overcome this bottleneck, inspired by natural materials, nanocomposite hydrogels have been developed as mechanically reinforced materials, which thus enable a broader range of use in different fields e.g. electronics, optics, magnetics, sensors, and biomedicine [1403–1406]. Nanocomposite hydrogels are 3D organic–inorganic hybrid nanocomposites, which are formed by physical or chemical cross-linking of polymer networks in aqueous solution together with functional inorganic nanomaterials incorporated into the matrices [1407–1411]. In general, there are five major approaches to fabricate hydrogel–nanoparticle composites with the uniform distribution, as illustrated in Fig. 8. The nanocomposite hydrogels can not only combine the individual functions of polymer hydrogels and inorganic nanoparticles (NPs), but also exhibit new or reinforced mechanical, electrical, optical, magnetic, remote-control, self-healable, stimuli-responsive, controlled drug delivery and biological properties due to their synergistic effect [1115,1404,1411–1414]. The commonly used inorganic nanomaterials for developing nanocomposite hydrogels include metal NPs (Au, Ag, etc), mineral NPs (natural silicate clay, Laponite, layered double hydroxides, etc), silica NPs (mesoporous silica, etc), magnetic NPs (Fe_3O_4, etc), carbon NPs (graphene, graphene oxide, etc) and polymer NPs (nanocellulose, etc). For example, Au NPs are considered as the most widely used metal NPs in nanomedicine due to their excellent biocompatibility, tailorable shapes (spherical, nanorod, nanoshell, etc) and sizes and unique electrical and optical properties (surface plasmon resonance, photothermal conversion) [1415,1416]. To engineer nanocomposite hydrogels with unique properties to meet specific needs, Au NPs have been widely incorporated as reinforcing elements into the matrices of natural polymer hydrogels or synthetic polymer hydrogels. In addition, to endow nanocomposite hydrogels with enhanced electrical/electronic property, various electrically conductive nanomaterials e.g. Au, Ag, graphene, carbon nanotubes, polypyrrole, polyaniline, PEDOT:PSS can be combined with the polymer networks of hydrogels. Table 21 summarizes some examples of the nanocomposite hydrogels.

3.5. Supramolecular hydrogels

Supramolecular hydrogel networks are materials crosslinked by reversible non-covalent supramolecular interactions e.g. π–π stacking, hydrogen bonding, metal coordination, ionic interactions, and hydrophobic interactions. Supramolecular hydrogels can exhibit quite interesting and significant properties originating from their dynamic nature, such as bioadhesive [965,1456,1457], self-healing [1458,1459], stimuli-responsiveness [1460–1462], adaptability [1463], and molecular recognition [1464,1465]. One good example is the cyclodextrin-based host − guest supramolecular hydrogel systems [1466,1467], e.g. mixing the cyclodextrin- and azobenzene-modified hyaluronic acid [810] or PAA [1468]. For the more detailed information of supramolecular hydrogels, we refer the readers to some excellent reviews [1469–1474].

3.6. Stimuli-responsive hydrogels

Stimulus-responsive hydrogels are polymer networks that are able to actively respond to a wide range of environmental stimuli e.g. light [1145,1171,1173,1475], temperature [258,538,1476–1479], mechanics [801,826,1480], glucose [1151,1345,1481], enzyme [1184,1199,1482,1483], pH [571,1484,1485] and so forth. One representative examlpe is the thermosensitive hydrogels that can undergo reversible sol − gel transition as a response to temperature changes, such as agarose, gelatin, PNIPAAm, etc. For the more detailed information of stimuli-responsive hydrogels, we refer the readers to some excellent reviews [178,239,245,1486–1493].

4. 3D printing of hydrogels

4.1. Multifaceted considerations

4.1.1. Extracellular matrix mimics

Extracellular matrix is a complicated cell-centered microenvironment that comprises proteins (collagen, elastin, laminin, fibronectin),

Table 19
Summaries of poly(vinyl alcohol) hydrogel.

Chemical modifications	Cross-linking methods	Properties	Applications
Acrylation [1225,1226,1227,1228,1229] Norbornene functionalization [1230,1231] Iodine modification [1226] Tyramine functionalization [1232,1233,1234] Thiolation [1235] Acrylamide functionalization [1236] Heparinization [1237]	Physical cross-linking: freezing/thawing cycle [262], hydrogen bonding (tannic acid [1238]), PEG induction [1239,1240] Chemical cross-linking: photo cross-linking [1212,1241], free radical polymerization [1241], glutaraldehyde [1242], click cross-linking [1243,1244], borax [1245,1246] Radiation cross-linking: electron beam [1247,1248], gamma ray [1249,1250]	Biocompatible [1251,1252] Self-healable [1253] Non-degradable [168,1226] Transparent [1254]	Tissue regeneration: bone [1225], cartilage [1255,1256,1257], meniscus [1258], nerve [1259], vascular constructs [1260,1261], vocal cords [1262] Delivery system: drugs [1263,1264], genes [1265], growth factors [1239], proteins [1211] Wound healing [1266]

Table 20
Summaries of polymer composite hydrogels from the double-component blends of natural and synthetic polymers.

Polymer blends	Examples	
Natural–natural	Polysaccharide–polysaccharide	Gellan gum–carrageenan [1272]
	Polysaccharide–glycosaminoglycan	Dextran–hyaluronic acid [1273], dextran–heparin [1274], cellulose–hyaluronic acid [1275,1276,1277,1278], chitosan − chondroitin sulfate [1279], alginate–chondroitin sulfate [1280], alginate − heparin [1281,1282], chitosan–hyaluronic acid [1283,1284,1285,1286,1287,1288], pectin − hyaluronic acid [1289], alginate–hyaluronic acid [1290]
	Polysaccharide–polypeptide/protein	Alginate–silk [1291], gellan gum − silk [1292], chitosan–collagen [1293,1294], alginate–collagen [1295], alginate–gelatin [1296,1297,1298], cellulose–gelatin [1299], pectin–silk [1300]
	Glycosaminoglycan–glycosaminoglycan	Heparin − hyaluronic acid [1301,1302], hyaluronic acid–chondroitin sulfate [720]
	Glycosaminoglycan–polypeptide/protein	Chondroitin sulfate − collagen [718,1303,1304], hyaluronic acid − collagen [718,1305,1306,1307,1308,1309,1310,1311,1312,1313], hyaluronic acid − elastin [1314,1315], hyaluronic acid–fibrin [1316,1317], hyaluronic acid–silk [1318,1319], hyaluronic acid–gelatin [1320]
	Polypeptide/protein–polypeptide/protein	Silk − collagen [1321,1322,1323], collagen–gelatin [1324], gelatin − elastin [1325], gelatin–silk [1326,1327], silk–elastin [1328,1329,1330,1331,1332]
Natural–synthetic	Polysaccharide–synthetic	Alginate–PAAm [1333,1334,1335,1336,1337], gellan gum–PVA [1338], gellan gum − PEG [1339], dextran − PEG [1340], chitosan–PAAm [1341], cellulose–PVA [1251,1342], chitosan–PVA [1343,1344,1345], alginate–PVA [1346,1347,1348]
	Glycosaminoglycan–synthetic	Chondroitin sulfate–PEG [1349], heparin–PEG [757,760,761,771,792,1350,1351,1352,1353,1354,1355,1356,1357,1358,1359], heparin–PVA [764,1360,1361,1362,1363,1364], hyaluronic acid–PEG [1365,1366], hyaluronic acid–PNIPAAm [1367], hyaluronic acid–PNIPAAm [1368]
	Polypeptide/protein–synthetic	Silk–PEG [1369,1370,1371], silk–PVA [1372,1373,1374,1375], gelatin–PAAm [1376]
Synthetic–synthetic		PVA–PAA [1377,1378], PAAm–PVA [1379]

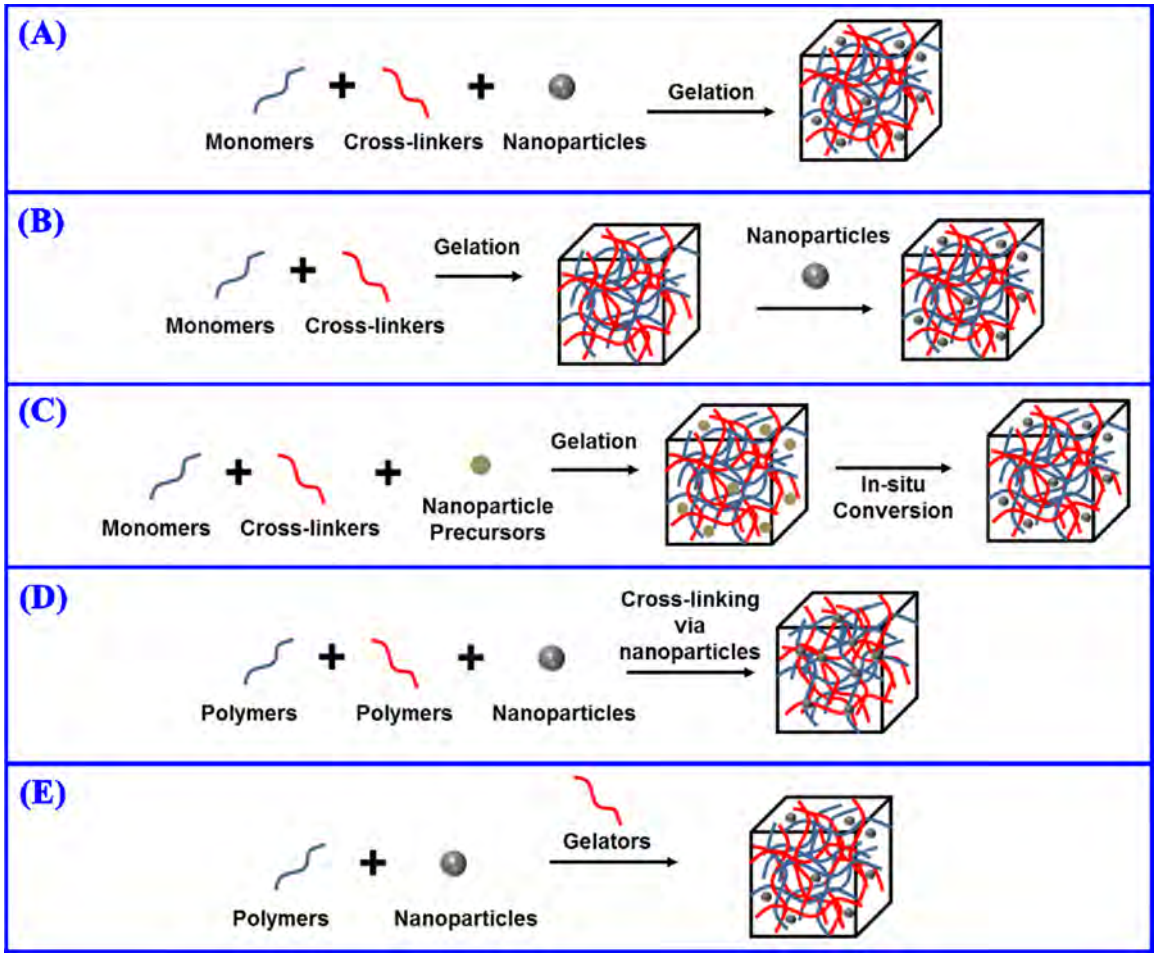

Fig. 8. Five major ways for the fabrication of hydrogel–nanoparticle composites with uniform distribution. (A) Synthesizing hydrogels in the suspension of nanoparticles. (B) Physically incorporating nanoparticles into the hydrogel matrices after gelation. (C) In situ synthesis of nanoparticles from precursors embedded in the hydrogel matrices after gelation. (D) Using the nanoparticles as cross-linker to generate hydrogels. (E) Fabricating the hydrogels using polymers, nanoparticles and gelators.
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Table 21
Summaries of nanocomposite hydrogels.

NPs	Polymers	Properties	Applications	Refs
Au	κ-Carrageenan	Thermoresponsive, mechanical reinforcement	Controlled drug release	[1418]
	Agarose	Electrically conductive	Catalysis	[1419]
	Hyaluronan	Extrudable for 3D bioprinting	Fibroblast remodeling	[1420]
	Gelatin	Biodegradable	Enhancing bone formation	[1421]
	Collagen	Injectable, mechanical enhancement, shear-thinning, self-healing	Combinatorial antitumor photothermal/photodynamic therapy	[1422]
	Silk	Laser-induced heating, injectable, biocompatible	Antibacterial treatment of focal infections	[1423]
	PAAm	Electrically conductive	Wearable pressure sensors	[1424]
	PNIPAAm	Thermoresponsive, rapid self-healing, excellent mechanical property	Biomedicine, biosensors, molecular electronics, engineering	[1425,1426]
Ag	PVA	Reusable robust carrier	Catalysis	[1427]
	Alginate/PNIPAAm	High stretchability, toughness, tissue adhesion, antimicrobial	Adhesive dressings, wound healing	[1428]
Montmorillonite	Chitosan	Electrostimulation-responsive	Controlled drug release	[1429]
	Alginate, hyaluronic acid	High osteointegration	Bone regeneration	[1430]
	Methacrylated glycol chitosan	Increased Young's modulus, controlled degradation, promoted calvarial healing	Bone tissue engineering	[1431]
Laponite	Collagen	Bioactive	Bone tissue engineering	[1432,1433]
	PNIPAAm	Excellent mechanical property, thermoresponsive bending property, transparent	Stimuli-responsive "smart" soft robots, sensors, artificial muscles, tissue engineering	[1434,1435]
Hectorite	PNIPAAm	Extraordinary mechanical, optical, and swelling/de-swelling properties	Tissue engineering, drug delivery, sensors, artificial muscles, microactuators	[1412,1436]
LDHs	PAAm	Ultrahigh tensibility, hierarchical porous structure	Artificial muscles, biomedical devices, drug delivery	[1437]
Silica	Alginate, κ-carrageenan, PVA	Tunable structural organization	Biomaterials	[1438]
PMO	Alginate	Controlled cell growth/migration	Tissue engineering, regenerative medicine	[1439,1440]
Fe_3O_4	PNIPAAm	Thermosensitive, magnetically responsive, remote-controlled	Active components of micro/nanoscale devices, biomaterials	[1441,1442]
	GelMA	Mechanically stiff	Tissue engineering, protein/cell delivery	[1443]
Graphene	PAAm	Improving biocompatibility, allowing neuronal adhesion	Neural interface of brain devices	[1444]
	Silk	Tunable stiffness, bioactive	Nerve regeneration	[1445]
	Elastin	Light-controlled	Actuators	[1446]
Graphene oxide	PNIPAAm	Photothermally sensitive, remote light-controlled	Microfluidic devices, liquid microvalves	[1447]
	PAAm	Tough, highly stretchable	Mechanically robust materials	[1448]
	GelMA	Tunable mechanical strength and electrical conductivity	3D scaffold materials for tissue engineering	[1449]
Nanocellulose	Hyaluronic acid	Injectable, mechanically stiff, cell supportive	Promoting ASC spread and proliferation, tissue engineering	[1450]
	PVA	Highly transparent, good UV-blocking property	Ophthalmic applications	[1451,1452]
	PAAm	Excellent mechanical and absorptive properties	Hygiene products, biomedical devices	[1453]
	Gellan gum	Mechanically reinforced, cell viability promotive	Annulus fibrosus tissue regeneration	[1454]
Polypyrrole	Agarose	Electronically conductive, self-healing, thermoplastic	Electronic skin (e-skin)	[1455]

glycosaminoglycans (sulfated heparin, chondroitin, keratin) and other soluble molecules. Most of the glycosaminoglycans are attached to the protein cores to generate proteoglycans. The glycosaminoglycans can maintain necessary compressive resistance and sequester soluble molecules for cells and tissues. ECM can provide dynamic biochemical and biophysical cues for the cell microenvironment, which is a critical determinant of cell physiology and fate [162,1494,1495].

As the paradigm for representative in vitro cell culture model, cells and tissues are quite often cultured on 2D substrates e.g. tissue culture plates or flasks. However, from the perspective of cell biology, it has been demonstrated that cells and tissues cultured on 2D substrates cannot mimic cell growth in vivo and express some tissue-specific genes or proteins that are comparable to those at levels in vivo [159,161,1496]. In vivo, cells grow in 3D with complex intercellular interactions, which has been encouraging a paradigm shift from the routine 2D cell culture model to mimetic 3D cell culture microenvironment [1497–1500]. Cells are able to behave more natively when they are cultured in 3D microenvironment. From the standpoint of tissue engineering, establishing a cell microenvironment that can mimic the native ECM is highly desired and meaningful to recapitulate in vivo milieu and replicate cell/tissue functions in vitro. By mimicking ECM, hydrogels are capable of not only supplying structural support for cell residence but also affording various predefined biochemical (cytokines, growth factors, cell adhesion peptides, etc) and biophysical (structures, stiffness, degradation, etc) cues for modulating cell fate [128,1501–1503].

4.1.2. Scaffold designs

Since Robert Langer and colleagues pioneered in 1993 the concept that combined therapeutic cells with polymeric matrices to regenerate/reconstruct damaged tissues and organs [1504], this research field has witnessed the explosive development of new materials and structures for tissue-engineered scaffolds. Ideally, scaffold designs should meet the

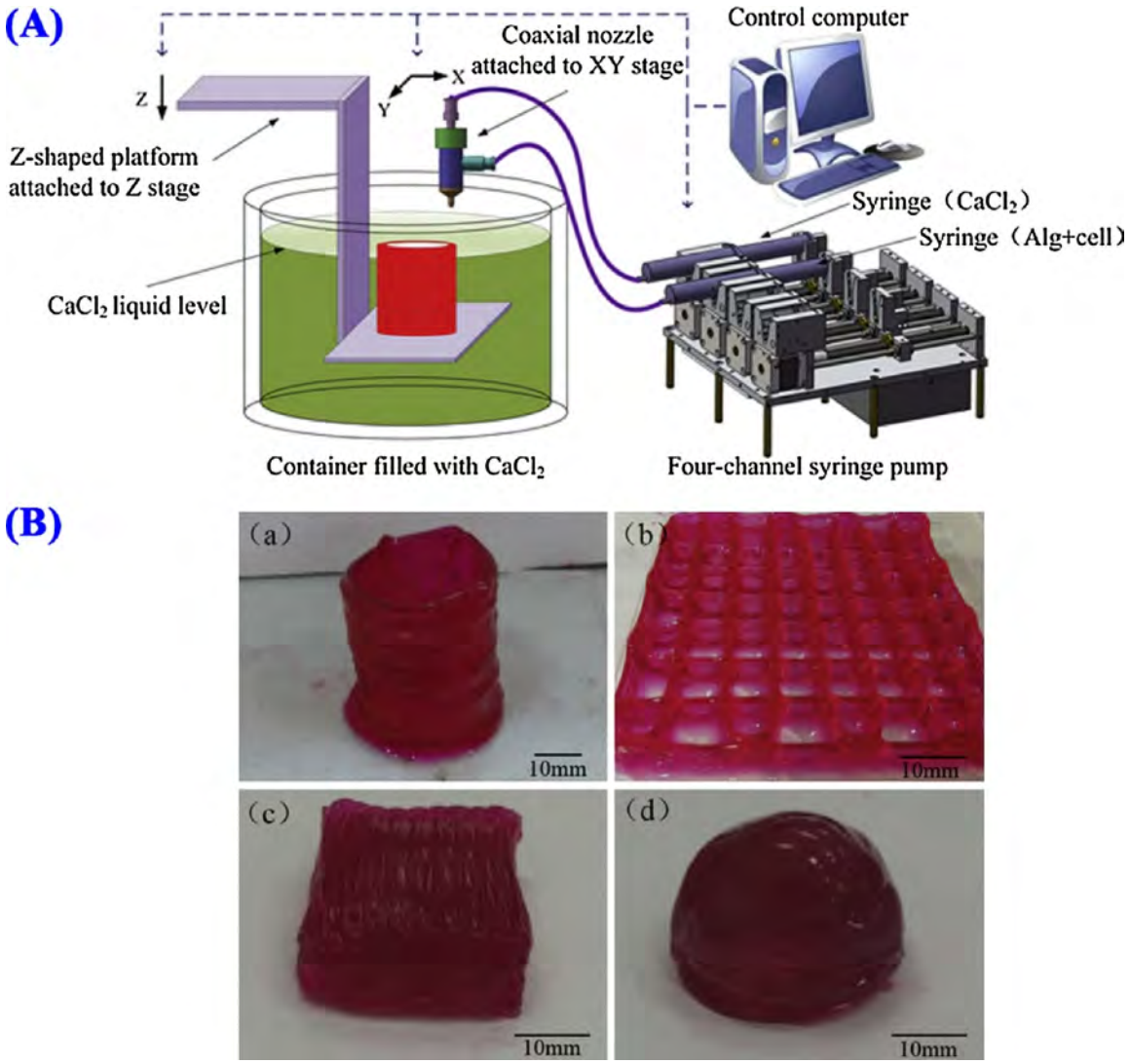

Fig. 9. (A) Schematic illustration of the coaxial nozzle-assisted 3D printing/bioprinting system. (B) Photographs of the printed 3D alginate constructs with built-in microchannels: (a) hollow cylinder, (b) grid, (c) cuboid and (d) hemispheroid.
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following requirements: (1) A scaffold should be highly porous 3D construct with interconnected pore network. The structure parameters (pore size and distribution, pore shape and orientation, specific surface area, pore interconnectivity, etc) should be optimized for migration and growth of cells and mass transfer of nutrients, oxygen molecules and metabolic waste [1505]; (2) The materials for scaffold designs should be biocompatible and biodegradable in a tailorable way so as to match the dynamic growth needs of different cells and tissues ex vivo and in vivo [1506]; (3) The scaffolds should have suitable surface physical and/or chemical cues for cell attachment, spread, proliferation and differentiation [127,130,1507]; and (4) A scaffold should be able to supply appropriate mechanical properties (strength, toughness, elastic stiffness, etc) that meet the needs of different cell types or match those of the surrounding tissues at implantation sites [139,145,175]. Such mechanical cues are preferentially dynamic and tunable at different phases of cell/tissue growth.

4.1.3. Cross-linking and printability

Whether a hydrogel material is suitable for a specific 3D printing technique is majorly dependent on its physicochemical properties when using the respective 3D printer. To think about the printability of a hydrogel material, two determinant factors are the rheological properties (viscosity, shear thinning, yield stress) and cross-linking mechanisms (physical cross-linking, chemical cross-linking). For physical hydrogels, the cross-linking process can be done during printing or after the printing. Here we take the alginate hydrogel system as an example. Pure alginate inks in the form of low concentration sols (<6 wt%) tend to diffuse after 3D printing through the nozzles and cause deformation of the whole 3D structure due to the low mechanical strength [1508]. To tackle this problem, He and colleagues printed alginate sols (2～5 wt%) into $CaCl_2$ solution to maintain the structural integrity of whole 3D scaffolds during the printing process [1509]. Using a coaxial nozzle, the team were able to fabricate high strength 3D hydrogel constructs with built-in microchannels, as illustrated in Fig. 9.

The viscosity of a polymeric solution or blended paste, such as a hydrogel precursor, is mainly dependent on the concentration and molecular weight of the polymer/precursor in solution. To enhance the printability of alginate sol, Schütz et al. [1510] developed an ink material composed of 3 % alginate and 9 % methylcellulose suitable for both 3D printing and bioprinting processes. The addition of methylcellulose lead to strongly increased viscosity of the alginate solution, which thereby allowed accurate and easy deposition and cross-linking with $CaCl_2$ after the printing process. Methylcellulose can be released from the printed scaffolds in solution, generating the constructs with

high elasticity and stability and increased microporosity. Similarly, the same group also formulated a blended paste of 16.7 % alginate and 6 % PVA, which significantly improved the viscosity of the paste for direct 3D printing of stable and regular scaffolds including vasculature-like constructs through a core/shell nozzle tip [1511].

In spite of the printability-favoring constant viscosity from reversible interactions and favorable compatibility with biologic systems, the major disadvantage of the physically cross-linked hydrogels is their lack of mechanical properties, probably causing the stability issues and handling difficulties of the printed scaffolds. In this context, increasing efforts have been focusing on an elaborate combination of the weak reversible interactions from physical cross-linking and the stable covalent bonds from chemical cross-linking during and/or after printing, as discussed in section 3.1, thereby capable of promoting the mechanical and handling properties of the printed hydrogel constructs. The chemical cross-linking is mainly used for stabilising the fabricated hydrogel constructs after 3D printing. For this purpose, the polymer precursors of hydrogels can be modified with photopolymerisable functional groups e.g. acrylate, diacrylate, methacrylate, methacrylamide, and in this case the fabricated constructs are irradiated by UV light immediately after printing. Such examples include single component ink materials e.g. PEGMA [1512], GelMA [1513], two-component ink materials e.g. hyaluronic acid/dextran methacrylate [1514], PEGDA/alginate [1515], etc. Burdick and colleagues [1516] introduced methacrylates into hyaluronic acid macromers, which were thereafter modified with either adamantane (Ad) or β-cyclodextrin (β-CD) moieties. These modifications produced the ink components Ad-MeHA and CD-MeHA. When mixed together, they can yield the HA-based hydrogel networks through both supramolecular bonding (i.e. intermolecular guest–host bonds between Ad and β-CD moieties) for dynamic bonding, shear-thinning, self-healing and supporting and covalent cross-linking for mechanical stabilising via UV light induced photopolymerization. Due to such a design, the ink materials can be directly printed into self-healable 3D hydrogel structures (Fig. 10), thereby holding potentials for 3D printing of multimaterials and complex constructs at high resolution even with open channels.

4.2. Biomaterial ink selection

4.2.1. Definition of biomaterial inks

According to a recent paper by Groll et al. [153], the term biomaterial inks are defined as "(bio-)materials that can be printed and subsequently seeded with cells after printing, but not directly formulated with cells". Representative examples of such inks cover a wide range of biomaterials including metals, ceramics, cements, glasses, non-degradable polymers (polyetheretherketone (PEEK) [1517], polytetrafluoroethylene (PTFE), silicone [1518], etc), biodegradable thermoplastics (polycaprolactone (PCL) [1519], polylactide-co-glycolide (PLGA) [1520], etc), hydrogel-forming natural polymers (alginate, silk, etc) and synthetic polymers (PAA, PEG, etc), as well as sacrificial materials (PVA [1511], MC [1510], etc), support materials (PCL [1521], calcium phosphate cement [1522], etc) and lithography-based resins

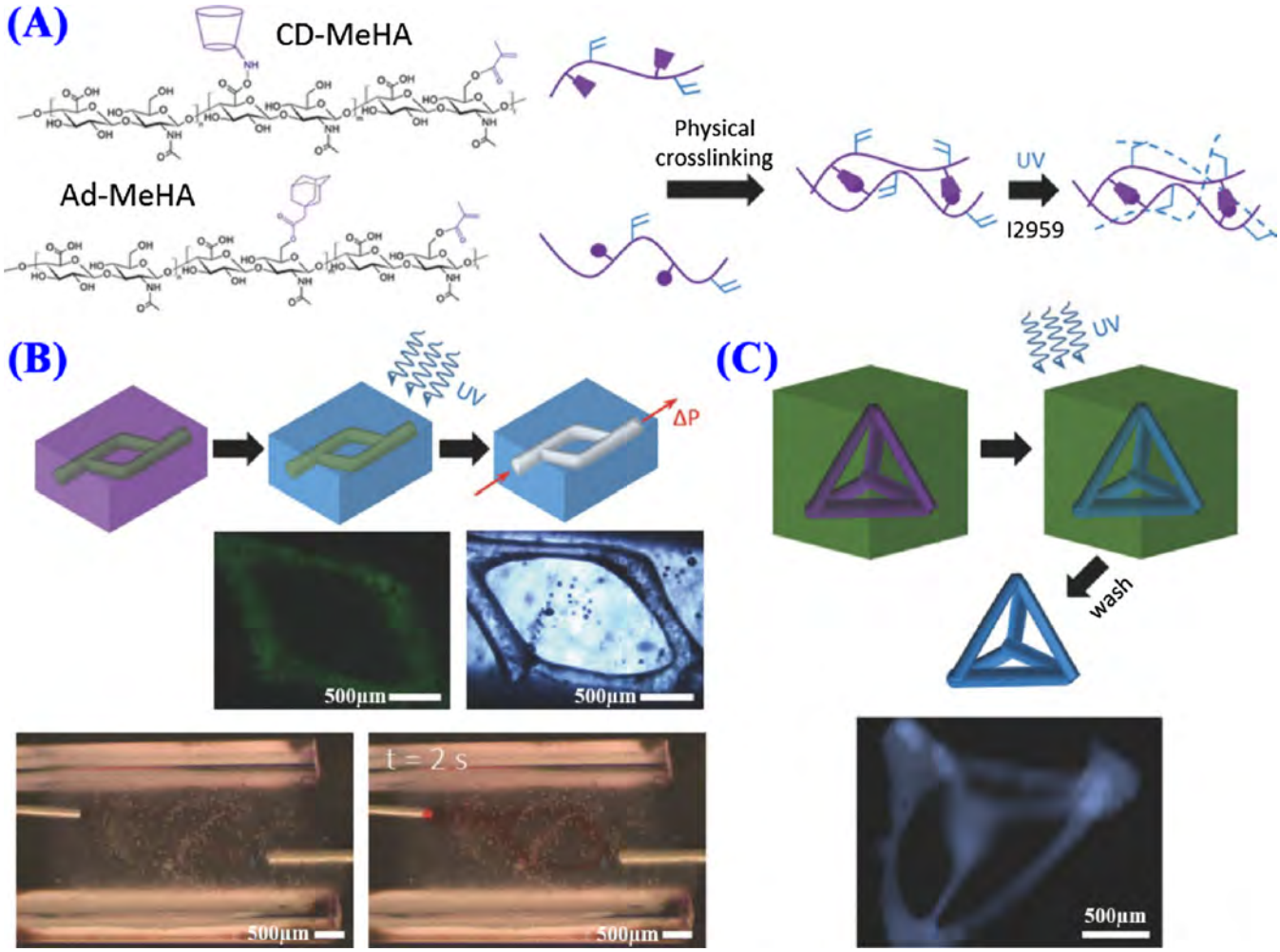


Fig. 10. 3D Printing of self-healing MeHA hydrogels with guest–host bonds. (A) Hyaluronic acid modification. (B) Printing of the channels. (C) Printing of the self-supporting architectures.
Reproduced with permission [1516]. Copyright 2015, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

Table 22
Summaries of reported single component hydrogel inks for 3D printing.

3D printing techniques	Biomaterial inks	Applications	Refs
Extrusion printing	Chitosan	Tissue engineering, drug delivery	[1524]
3D plotting	Nanocellulose	Wound healing, regeneration and tissue repair	[1525]
Extrusion printing	Chondroitin sulfate	Cartilage regeneration	[1526]
3D plotting	Collagen	Adipose and bone tissue engineering	[1527]
Stereolithography	PEGDA	Tissue engineering	[1528]
Laser jet printing	PEGDA	Tissue engineering, formation of immobilized cell and protein arrays	[1529]
Stereolithography	GelMA	Bone replacement materials	[1530]
2 P P	PEGDA	Tissue constructs, artificial ECM	[221]

[1523].

4.2.2. Design of biomaterial inks

In accordance with the above discussed in section 3, we here mainly focus on the following five types of biomaterial inks: (i) single component hydrogels, (ii) IPN hydrogels, (iii) supermolecular hydrogels, (iv) nanocomposite hydrogels, and (v) multimaterial inks. Besides, the components of hydrogels including polymers, prepolymers and nanoparticles can be chemically modified to meet specific needs for ink formulations and 3D printing.

4.2.2.1. Single component hydrogel inks. Just as its name implies, single component hydrogel inks refer to biomaterial inks made of only one component of hydrogel or hydrogel precursor, ranging from natural polymer hydrogels to synthetic polymer hydrogels as discussed in section 3.2 and 3.3, respectively. Table 22 lists the summaries of examples of reported single component hydrogel inks for 3D printing.

4.2.2.2. IPN hydrogel inks. The IPN hydrogel inks refer to biomaterial inks made of composite hydrogels or precursors with polymer networks thereof physically interweaved with each other (see section 3.4.1). compared with the single component hydrogel inks, IPN hydrogel inks can yield 3D scaffolds/constructs with improved toughness and fracture strength. Table 23 lists the summaries of examples of reported IPN hydrogel inks for 3D printing.

4.2.2.3. Supermolecular hydrogel inks. To meet practical application requirements in tissue engineering fields, hydrogels should be able to provide sufficient mechanical strength. Especially in the case of dynamic/cyclic loading and repeated deformation, hydrogel materials are expected to self-repair, self-heal and survive to maintain mechanical integrity. For this purpose, one strategy is to develop the supermolecular hydrogel inks. Table 24 lists the summaries of examples of reported supermolecular hydrogel inks for 3D printing. Recently, Wang and colleagues [1539] developed a novel host–guest supramolecule (HGSM) on the basis of inclusion reaction between cyclodextrin (CD) and adamantane (Ad), which can be utilised as multifunctional crosslinkers to cross-link GelMA during hydrogel formation. The produced hydrogel matrices are composed of physical noncovalent bonds incorporated into covalently cross-linked networks. Through this combination of weak noncovalent interactions and strong covalent bonds, the GelMA hydrogels can not only exhibit enhanced mechanical strength and maintain excellent biocompatibility, but also show self-healing and 3D printability as ink materials (Fig. 11).

4.2.2.4. Nanocomposite hydrogel inks. Nanocomposite hydrogel inks refer to engineered nanocomposite hydrogels or hydrogel precursors that are suitable for 3D printing. As discussed in section 3.4.2, dependent on the types of incorporated nanoparticles, the hydrogel matrices can obtain different physical, chemical and biological properties. Table 25 lists the summaries of examples of reported nanocomposite hydrogel inks for 3D printing. Hong and colleagues [1544] developed a nanocomposite hydrogel ink composed of biocompatible sodium alginate, PEGDA and nanoclay materials. The formulated hydrogel inks can allow direct 3D printing of highly stretchable and tough hydrogels into complex constructs, which first demonstrated the 3D printability of tough hydrogels and the suitability for long-term cell culture.

4.2.2.5. Multimaterial inks. Multimaterial inks have been widely explored to remedy the shortcomings of single component hydrogel inks. Having all the advantages of additive manufacturing, Multimaterial 3D printing can create complex constructs with defined geometries and desired functionalities [48]. In addition, different from IPN hydrogels in which polymer components are physically interentangled, the polymer networks of multimaterial hydrogels can be cross-linked together e.g. by amine–carboxylic acid coupling [1553]. Such printing examples include PEGDMA/PEGDA + PEG-RGDS ink through stereolithography [1554], agarose + SWCNT/PVP through extrusion printing [1555], alginate/methylcellulose + CPC through 3D plotting [1522], alginate/gellan gum + CPC through 3D plotting [1556].

4.3. Advances in 3D printing of hydrogels

4.3.1. New 3D printing techniques

3D printing techniques are increasingly applied to manufacture multicomponent, multimaterial and end-use products [54]. To overcome existing limitations and tackle new challenges, new additive manufacturing techniques are being developed to meet different user-specific requirements. When stereolithography is carried out above an oxygen-permeable build window, Continuous Liquid Interface

Table 23
Summaries of reported IPN hydrogel inks for 3D printing.

3D printing techniques	Biomaterial inks	Applications	Refs
Stereolithography	GelMA/PEGDA	Engineered nerve guidance conduits for repairing peripheral nerve injuries	[1531]
Extrusion printing	Gelatin/alginate	Tissue engineering	[1532]
3D plotting	Gelatin/alginate	Osteochondral tissue regeneration	[1533]
Extrusion printing	Gelatin/Hyaluronan	Traumatic brain injury repair	[1534]
3D plotting	Gelatin/agar	Tissue engineering	[1535]
3D plotting	Alginate/PAAm	Artificial tendons	[1536]
3D plotting	Alginate/gellan gum	Tissue engineering	[1537]
Extrusion printing	PVA/κ-carrageenan	Tissue engineering, drug delivery, bone regeneration	[1538]

Table 24
Summaries of reported supermolecular hydrogel inks for 3D printing.

3D printing techniques	Biomaterial inks	Applications	Refs
3D plotting	Ad- and CD-modified crosslinkers/GelMA	Tissue engineering, soft tissue repair	[1539]
Extrusion printing	HA-PNIPAAm/MeHA	Cartilage engineering	[1540]
Extrusion printing	Ad-HA/CD-HA, Ad-MeHA/CD-MeHA	Cartilage and cardiac tissue engineering	[808]
Extrusion printing	N-acryloyl 2-glycine/GelMA	Osteochondral regeneration	[1541]
Extrusion printing	Ad-HA/CD-HA, Ad-MeHA/CD-MeHA	Tissue engineering	[1516,1542]
Extrusion printing	N-acryloyl glycinamide/N-acryloyl glycinamide-co-N-[tris(hydroxymethyl) methyl] acrylamide	Osteochondral defect repair	[1543]

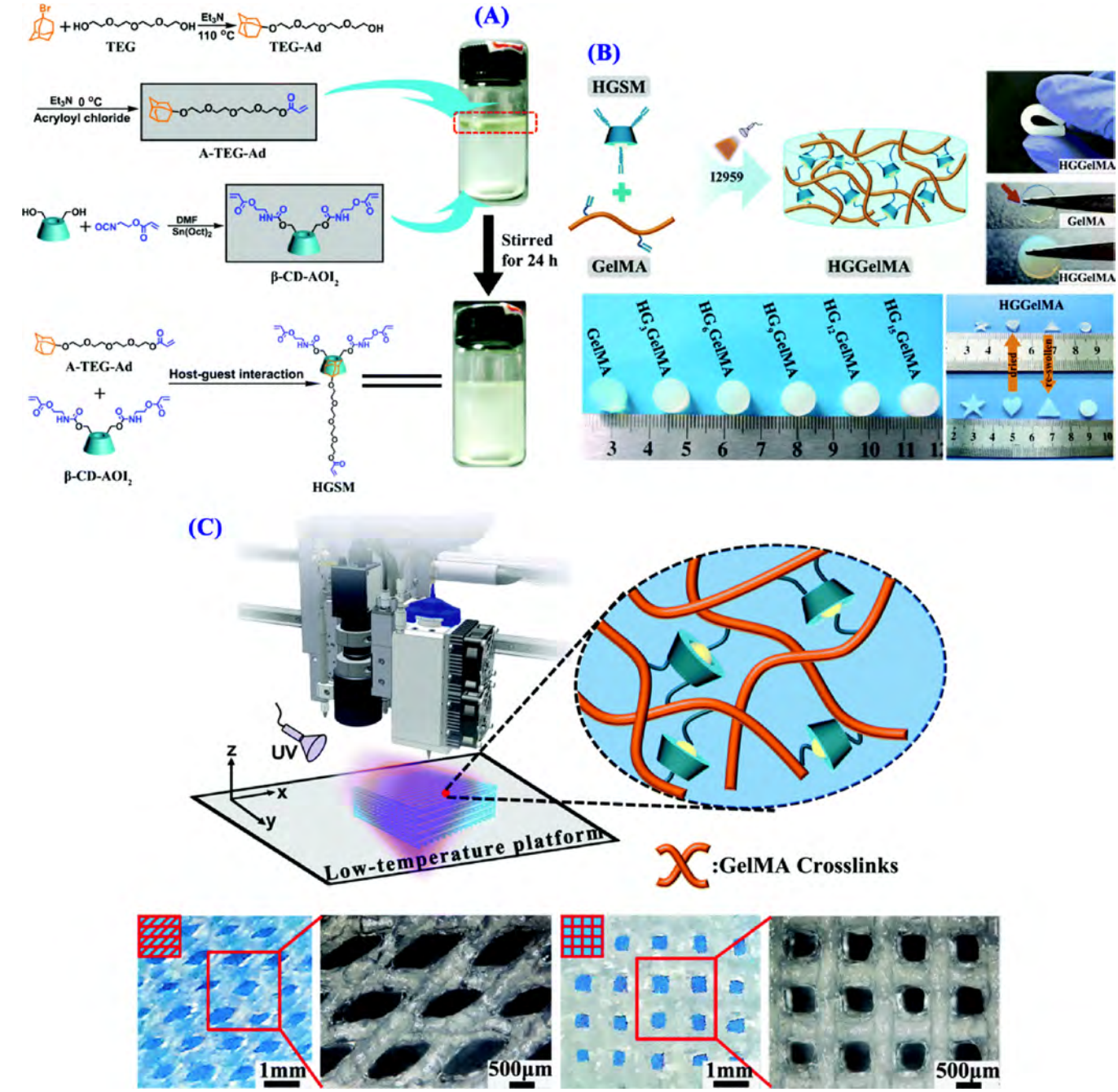


Fig. 11. 3D printing of self-healing GelMA hydrogels with host–guest interactions. (A) Schematic of HGSM synthesis. (B) Photos of HGGelMA and GelMA hydrogel samples. (C) 3D printing of HGGelMA hydrogel scaffolds.
Reproduced with permission [1539]. Copyright 2019, The Royal Society of Chemistry.

Table 25
Summaries of reported nanocomposite hydrogel inks for 3D printing.

3D printing techniques	Matrix materials	Nanoparticles	Applications	Refs
3D plotting	Alginate dialdehyde/gelatin	Bioactive glass	Bone tissue engineering	[1545]
Extrusion printing	PVA	Au	Optical filters	[1546]
3D plotting	N-acryloyl glycinamide	Laponite	Bone regeneration	[1547]
Extrusion printing	GelMA	Laponite	Bone tissue engineering	[1433]
3D plotting	Methacrylated chitosan	Graphene oxide	Tissue engineering	[1548]
3D plotting	Thiolated hyaluronan	Au	Vessel and duct tissue engineering	[1420]
Stereolithography	PEGDA	Hydroxyapatite	Breast cancer bone metastasis model, drug sensitivity assessment	[1549]
Stereolithography	PEGDA	Hydroxyapatite	Osteochondral regeneration	[1550]
Extrusion printing	PEGDA, alginate, gelatin	Laponite	Tissue engineering	[1551]
3D plotting	Alginate/methylcellulose	Laponite	Skeletal tissue engineering	[1552]
Extrusion printing	Alginate/PEGDA	Laponite	Tissue engineering	[1544]

Production (CLIP) can be achieved through generating an oxygen-containing "dead zone", a thin uncured liquid layer that can continuously provide raw ink materials to ensure the continuity of the whole printing process (Fig. 12**A-B**) [1557]. Taylor and colleagues [1558] recently demonstrated a rapid volumetric additive manufacturing method i.e. Computed Axial Lithography (CAL) by illuminating a rotating volume of photopolymer within dynamically evolving light field, which allowed the concurrent 3D printing of entire complex constructs and circumvented the need for layer-by-layer (Fig. 12**C-J**). This method enables printing components that encase pre-existing solid conctructs for multimaterial fabrication. By using the capacitor edge effect, a complete 3D printing system was established for patterning liquids with 100 μm resolution applicable to a wide range of hydrogels [1559]. Recently, Lewis and colleagues [1560] reported the design and creation of voxelated soft matter through multimaterial multinozzle 3D (MM3D) printing (Fig. 12**K-L**) that can program the material component, structure and function at voxel scale. On the basis of diode-like behavior, the MM3D printheads enable seamless high-frequency switching among multiple viscoelastic materials (up to 8 different materials) to generate voxels with the volume close to that of nozzle diameter cubed. This work demonstrates a powerful strategy to create voxelated soft matter with MM3D printing, which would exclude periodicity restraints from present printhead design and meanwhile improve feature resolution and reduce fabrication time.

4.3.2. 4D printing of hydrogels

4.3.2.1. Definition of 4D printing. The concept of 4D printing was first introduced in 2013 by Skylar Tibbits [1561,1562], in which the fourth dimension means time. Since then, 4D printing has developed into a novel and thrilling branch of the well-established 3D printing and has been increasingly attracting attentions from scientists and engineers from a wide range of research fields e.g. intelligent materials and biomedical engineering. The central feature of 4D printing is the printed objects are able to reshape over time rather than remain static in an active and programmable way, possibly coupled with function evolvement. By now, there is no standard definition given for 4D printing. Here, we give a definition by Miao and colleagues [192]: "3D printing of objects which can, immediately after printing, self-transform in form or function when exposed to a predetermined stimulus, including osmotic pressure, exposure to heat, current, ultraviolet light, or other energy sources". Active materials for 4D printing cover a broad range including shape memory materials (e.g. shape memory polymers, shape memory alloys), dielectric elastomers, photoactive polymers, liquid crystal elastomers and stimuli-responsive hydrogels as well as active composite materials, which have been attempted to manufacture wearable devices, biomedical devices, artificial muscles and other intelligent products [1563]. For the more detailed information of 4D printing, we refer the readers to some excellent review papers [192,193,1563,1564].

4.3.2.2. Hydrogel-based 4D printing. In this review paper, we will mainly focus on the hydrogel-based 4D printing. For 4D printing hydrogels, hydrogel materials are generally integrated with non-swelling polymer or filament components. When the printed constructs are soaked into certain solvent, the hydrogel parts will swell and produce mismatch strains between the two components, thereby resulting in the shape global deformation due to asymmetrical swelling with no need for further programming after the printing. Through elaborate composite strategies, these 4D printed architectures can acquire tunable mechanical strength and other functional properties. Table 26 lists the summaries of reported hydrogel-based materials for 4D printing.

Programmable shape-changing systems can be widely found in the nature. One representative example is the nastic plant motions, in which plant organs e.g. flowers and leaves can make response to various environmental stimuli (touch, light, humidity, etc) and alter interior turgor to yield anisotropic and asymmetric distribution of tissue components and microstructures of plant cell walls, thereby leading to the dynamic changes of morphology or conformation. Inspired by nature, Lewis and colleagues [1566] recently demonstrated the programmable 4D printing of plant-mimetic complex architectures capable of morphing shape and generating complicated 3D morphology when soaking in water (Fig. 13). The nanocomposite hydrogel inks were formulated from stiff cellulose nanofibrils and soft acrylamide matrix to imitate the compositions of plant cell walls. The added nanoclay particles served as the rheological aid and resulted in desired viscoelastic property for direct ink writing. Due to the predefined alignment of cellulose nanofibrils, the 4D printed hydrogel constructs were endowed with localised anisotropic swelling property.

The 4D printable inks can be formulated from single active material, multiple active materials or even the combination of active materials and non-active materials (termed active composite materials [1571,1572]) for the manufacturing of shape-morphing architectures. The multimaterial 4D printing can impart more versatile shape deformations to the fabricated structures. Fig. 14 shows some examples of using single-material or multimaterial 4D printing to fabricate fully soft robots or other shape-shifting objects. In regard to the non-active materials, with precise control of internal stress, it is also possible to achieve the intended shape changes [1573,1574]. An alternative design strategy is based on the self-folding origami mechanism, which enables the 4D printing for shape-deforming of the flat materials into complex 3D architectures with desired configurations [1575,1576].

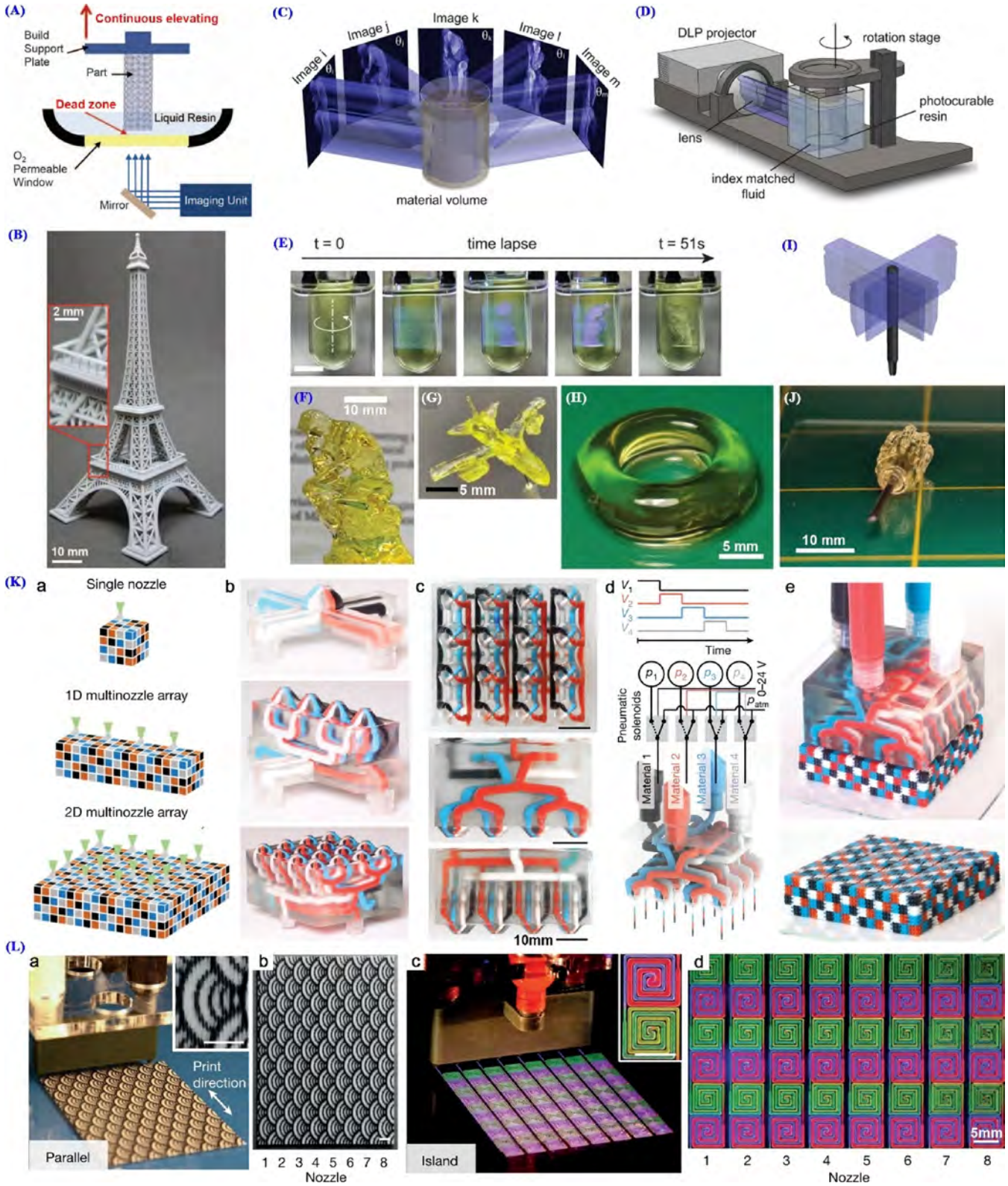
5. 3D bioprinting of cell-laden hydrogels

5.1. Multifaceted considerations

5.1.1. Definition of bioinks

Bioinks are very different from biomaterial inks. To be competent as a bioink, a biomaterial should act as a cell-laden medium/matrix during the formulation and printing. By definition from Groll and colleagues [153], bioinks refer to "a formulation of cells suitable for processing by

an automated biofabrication technology that may also contain biologically active components and biomaterials". Representative bioinks for 3D bioprinting may contain hydrogels or hydrogel precursors, decellularized matrices, separate cells, cell microcarriers, cell/tissue spheroids, mini-tissues, organoids [1579] and/or bioactive molecules e.g. adhesive peptides (RGD, KRSR, etc), growth factors (VEGF, TGF-β, etc), proteins (BMP-2, etc), cytokines (IL-4, etc), miRNA, DNA, etc. With regard to the types of selected cells, they can be human cells, animal cells, plant cells, microalgae, fungi cells, bacteria and a combination thereof. As a focus of this review, hydrogels are considered the most outstanding class of biomaterials for bioinks due to their tailorable physical, chemical, biological properties that can facilitate the use of



(caption on next page)

Fig. 12. Examples of some recently developed 3D printing techniques. (a–b) CLIP. (A) Schematic diagram to elucidate the working principle of CLIP printer in which the gyroid part is created continuously by simultaneously elevating build support plate and altering 2D cross-sectional UV images from imaging unit. (B) Eiffel Tower model, 10 cm tall. Reproduced with permission [1557]. Copyright 2015, American Association for the Advancement of Science. (C–J) CAL. (C) Patterned illumination from different directions can deliver a computed 3D exposure dose to a photopolymer. (D) Schematic diagram to elucidate the working principle of CAL printer. (E) Sequential view of the build volume (scale bar 10 mm) during CAL printing. (F) The 3D construct (40 mm tall) in (E) after rinsing away uncured photopolymer. (G) Airplane with sharp wing tips and overhanging wings. (H) Donut with smooth surface finish printed in highly deformable GelMA hydrogel. (I) Over-printing of 3D parts around pre-existing solid objects. (J) Example of a screwdriver handle printed by CAL to encase a metal shaft. Reproduced with permission [1558]. Copyright 2019, American Association for the Advancement of Science. (K–L) MM3D printing. (K) Multimaterial multinozzle 3D (MM3D) printheads. (a) Schematic of voxelated constructs through MM3D printing. (b–c) Photographs of the MM3D printheads. (d) Schematic of MM3D printhead operation. (e) Voxelated matter created by MM3D printing. (L) MM3D printing of voxelated soft matter. (a–b) Parallel printing of arbitrary pattern in wax (white) and silicone (black) with 8 × 1 two-material MM3D printhead. (c–d) Printing silicone spiral patterns with 8 × 1 four-material MM3D printhead. Reproduced with permission [1560]. Copyright 2019, The Authors, under exclusive licence to Springer Nature Limited 2019.

bioinks for 3D printing process and provide favorable extracellular microenvironment for various cell functions e.g. attachment, migration, proliferation and differentiation of encapsulated cells in 3D constructs (bioconstructs).

5.1.2. Bioprinting window

For the more detailed information on the rapidly developed 3D bioprinting field, we first refer the readers to some excellent reviews [10,11,13,1580]. Recapitulation of the hierarchically complex natural tissues in cell-laden 3D constructs is very attractive and meaningful for fabricating functional tissue/organ analogues for a wide range of biomedical applications. Both biopolymer hydrogels and synthetic polymer hydrogels have been extensively developed for formulating bioinks for 3D bioprinting, however, the formulation of bioinks requires a compromise between bioink printability and cell viability to enable the fabrication of complicated tissue- or organ-like constructs with high shape fidelity and resolution, which limits a narrow boundary for the use of hydrogels (Fig. 15). This is the concept termed bioprinting window or biofabrication window by researchers [185,1581].

5.1.3. Printability and biocompatibility

When formulating bioinks for 3D bioprinting, many factors should be deliberately taken into consideration to balance the printability and biocompatibility. Similar to that of biomaterial inks, the printability of bioinks is determined by the bioink rheological properties, cross-linking mechanisms and 3D printing parameters. In this section, we will focus on several main factors as follows.

(1) Material properties of bioinks: As will be further discussed in next section, both natural and synthetic polymers or prepolymers can be used to prepare hydrogels for bioink formulations. The compositions, structures, processability and properties of hydrogel materials are dependent on the types of used hydrogel precursors (polymers or prepolymers), precursor molecular weight, precursor concentration in bioinks and other added components e.g. nanoparticles. These variants will affect the rheological properties of the formulated bioinks. In materials science, the term rheology is the investigation of flow and deformation of materials (liquids, soft solids, solids) under the external condition of applied stress/force. Similar to 3D printing process (see section 4.1.3), important rheological parameters for printability in 3D bioprinting process are viscosity, shear thinning and yield stress. Viscosity is the resistance of a fluid to flow when applying stress, which can be tailored by changing precursor concentration and molecular weight as well as temperature; it can allow uniform cell encapsulation and retard collapse of printed structures. Shear thinning is a non-Newtonian behaviour of viscosity decreasing along with shear rate increasing, which is very important for the injectability, extrudability and printability of hydrogel materials. Yield stress refers to the stress that should be surmounted for the flow initiation of bioinks. In addition, viscoelasticity is also an important property for protecting cells from shear stress during the printing (e.g. extrusion) process.
(2) Cross-linking in the presence of cells: Both physical and chemical cross-linking methods are applicable to 3D bioprinting process. No matter prior to, during or after the 3D bioprinting, the solutoins/sols/pastes of hydrogels or hydrogel precursors will be physically and/or chemically cross-linked for gelation. In this context, it is inevitable for the encapsulated cells to suffer from this cross-linking procedure. Therefore, it is highly necessary to evaluate the cytocompatibility of the different cross-linking mechanisms (see section 3.1) with the embedded cells. Physical cross-linking methods are the most widely adopted due to their mild treatment and reversible gelation. Nevertheless, the printed bioconstructs are prone to mechanically weak and unstable, which is disadvantageous for long-term culture in vitro and in vivo. Chemical cross-linking methods can also be used for the gelation of bioinks, among which the UV cross-linking is the most commonly applied treatment for bioink gelation and cell encapsulation.
(3) 3D bioprinting parameters: The major bioprinting parameters include extrusion pressure, nozzle diameter, dispensing speed, bioprinting speed and biofabrication time. They must be flexibly optimised during 3D bioprinting process for optimal bioprinting outputs including the diameter, uniformity, precise placement and spacing distance of filaments and balanced with the post-printing viability of encapsulated cells. Biofabrication time determines the exposure duration of embedded cells to bioprinting conditions and too long bioprinting time will reduce the cell viability.
(4) Cell viability and biocompatibility: The 3D printed bioconstructs are expected to provide non-toxic or promotive excellular microenvironment for various embedded cell functions such as cell adhesion, migration, signaling, proliferation and differentiation. In addition, these printed bioconstructs should be able to avoid adverse immune responses or even elicit beneficial immunomodulatory functions when using them for in vivo

Table 26
Summaries of reported hydrogel-based materials for 4D printing.

Printing techniques	Materials	Applications	Refs
3D plotting	Alginate/PNIPAAm	Hydrogel actuators, sensors, soft robots, medical devices	[1565]
Extrusion printing	PDMAAm/PNIPAAm, cellulose nanofibrils, Laponite	Tissue engineering, biomedical devices, soft robotics	[1566]
Extrusion printing	Polyurethane/PHEMA, polyurethane/PNIPAAm	Foldable robots, electronics, biomedical devices	[1567]
Extrusion printing	PAAm/PNaAMPS/Laponite, Ecoflex/nanosilica	Soft devices for medicine and engineering	[1568]
Stereolithography	PEGDA/PHEMA/2-(2-methoxyethoxy) ethyl methacrylate (MEO_2MA)	Soft robotics, tissue engineering, actuators	[1569]
Stereolithography	PNIPAAm, poly(2-carboxyethylacrylate) (PCEA)	Hydrogel actuators	[1570]

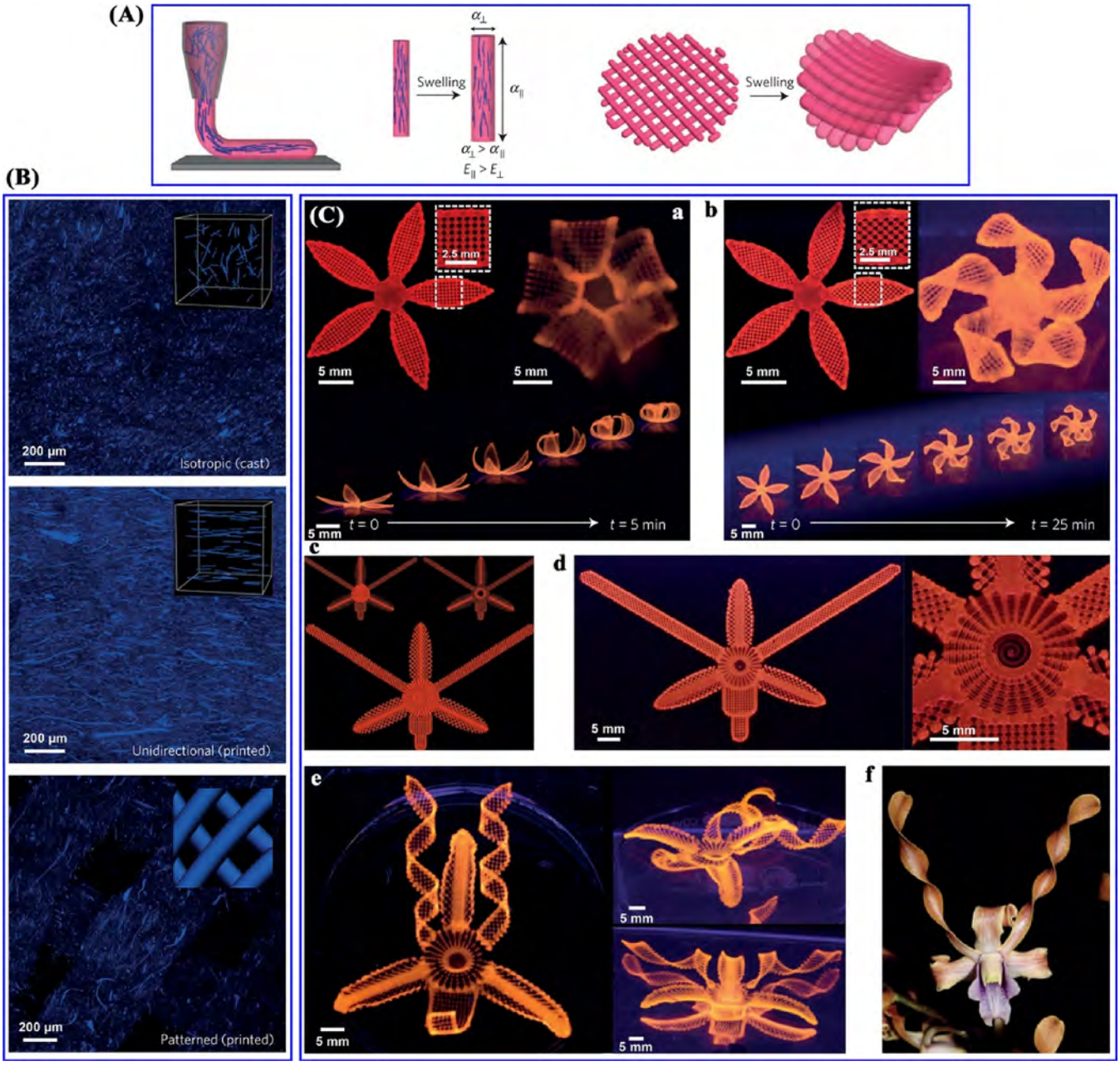


Fig. 13. Programmable localised anisotropy through biomimetic 4D printing: (A) Diagram of shear-induced one-step alignment of cellulose fibrils while printing composite hydrogel inks. (B) Distribution imaging of cellulose fibrils (blue) in the casted isotropic and the printed unidirectional and patterned specimens. (C) Complex flower morphologies fabricated by biomimetic 4D printing. (a) 90°/0° bilayer. (b) −45°/45° bilayer. (c) Print path, (d) printed structure and (e) swollen structure of a flower inspired by (f) Dendrobium helix.
Reproduced with permission [1566]. Copyright 2016, Nature Publishing Group.

investigation.

(5) Mechanical integrity and stability of printed bioconstructs: The formulated bioinks should ensure the high fidelity of the printed bioconstructs during and after the bioprinting process to maintain the structure, size, resolution and shape stability of the cell-laden bioconstructs for a desired period of time.
(6) Biodegradability of bioinks and printed bioconstructs: The satisfactory biodegradability and degradation rate of bioinks can promote the ECM production and remodeling of encapsulated cells in the printed bioconstructs.
(7) Biochemical and biophysical cues of bioinks: By definition in section 5.1.1, bioinks can contain various bioactive components such as growth factors, proteins, drugs and cytokines. In addition, the stiffness and elasticity of bioink materials can also be tailored by formulations to regulate cell differentiation toward different lineages according to the need given the fact that ECM elasticity and stiffness hold the potential to direct cell fate [1071,1582].
(8) Material biomimicry of bioinks: One should keep in mind that when formulating the bioinks for 3D bioprinting, designing the desirable material properties (structures, compositions, mechanics, functionalities, dynamic state, etc) should be on the basis of tissue- or organ-specifc endogenetic material components [13].

5.2. Selection and design of bioinks

Similar to the classifications of biomaterial inks, in this section we

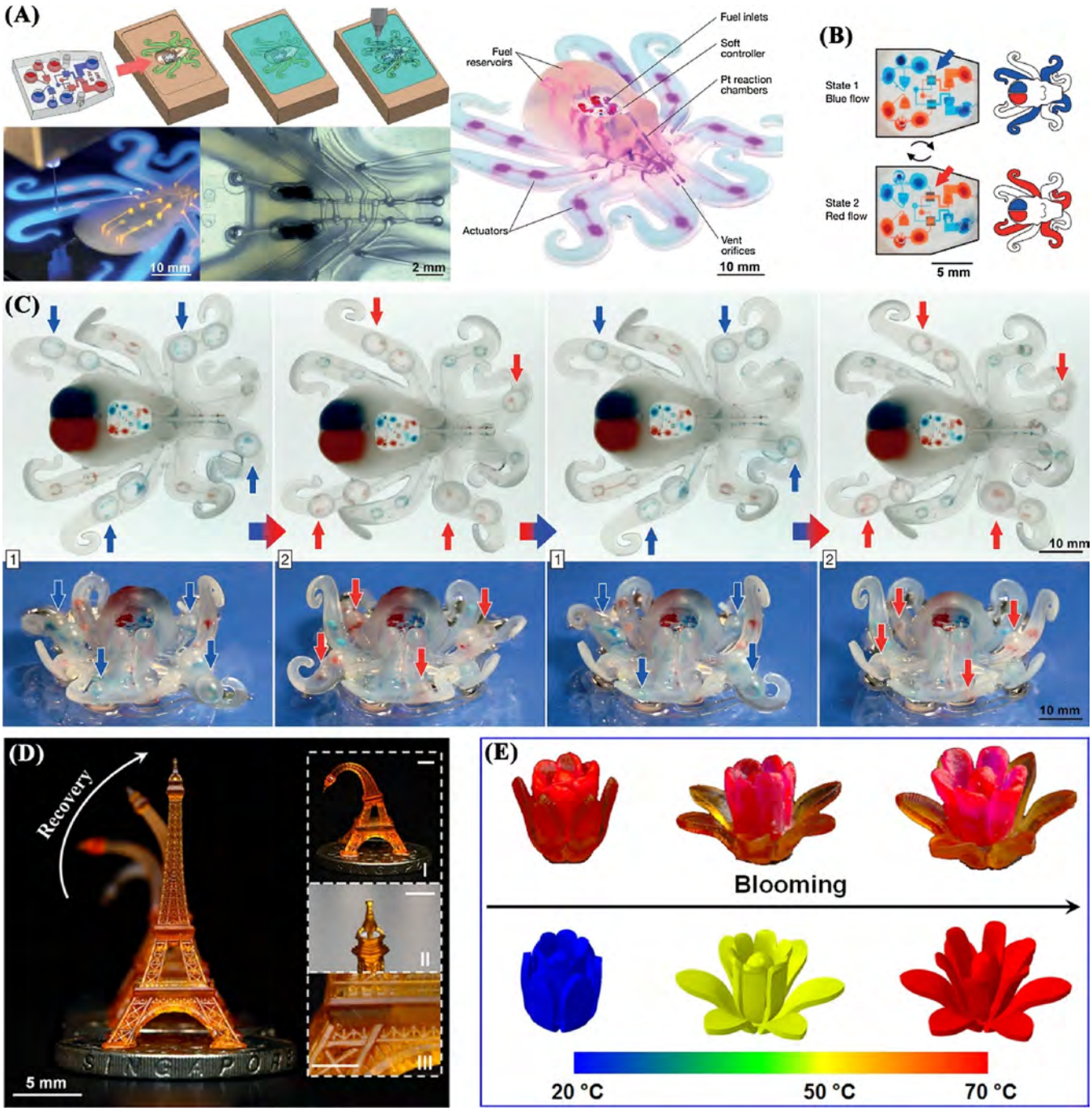


Fig. 14. Multimaterial 4D printing of an octobot: (A) Entirely soft autonomous robot design and fabrication. (B) The oscillator of soft controller trigger octobot to alternate between the blue (1) and red (2) actuation states. (C) Images of the octobot actuation autonomously alternating between the blue (1) and red (2) actuation states. Reproduced with permission [1577]. Copyright 2016, Nature Publishing Group. (D) Single-material 4D printing of Eiffel tower. (E) Sequential shape recovery of a flower from multimaterial 4D printing. Reproduced with permission [1578]. Copyright 2016, The Authors. This work is licensed under a Creative Commons Attribution 4.0 International License.

will mainly focus on five types of hydrogel-based bioinks (i.e. hydrogel materials + cells): single component bioinks, IPN bioinks, supramolecular bioinks, nanocomposite bioinks and multimaterial bioinks. For the more general information of bioink formulations for 3D bioprinting, we refer the readers to some excellent review papers [187,189,190,1583–1586].

5.2.1. Single component bioinks

The single component bioinks refer to bioink formulations of the single component hydrogels or hydrogel precursors and the encapsulated cells (i.e. single component bioinks = single component hydrogel materials + cells). Single component hydrogel inks have been well formulated with cells for 3D bioprinting. Representative examples of such established hydrogel systems include alginate-based and gelatin-based bioinks. To improve printability, one can increase the polymer concentration and cross-linking density in hydrogel networks. However, these changes may cause harm to the encapsulated cells due to the decrease in porosity, which has adverse effect on cell functions (migration, proliferation, etc) and mass transfer (nutrients, oxygen, etc). Table 27 lists the summaries of examples of reported single

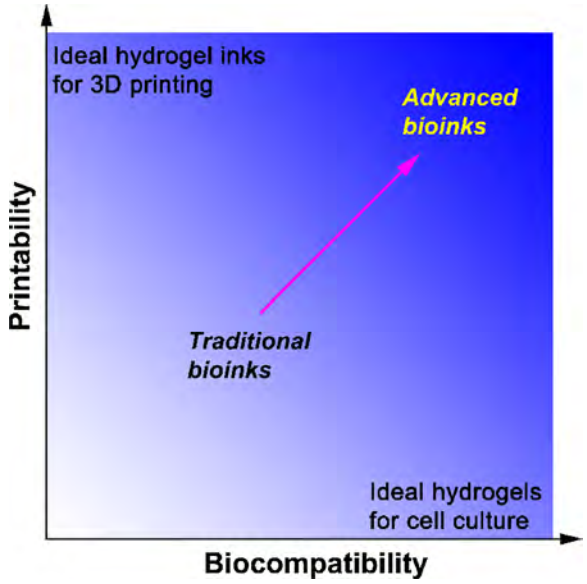


Fig. 15. The bioprinting window for the rational design of bioinks demands a compromise between the printability of bioinks and the biocompatibility of hydrogel constructs/scaffolds.
Adapted with permission [111]. Copyright 2016, Biomedical Engineering Society.

component bioinks for 3D bioprinting.

The human brain is an extremely complicated organ with various layered tissue structures. Researchers have extensively attempted to model and investigate the brain architecture through in vitro 2D cell culture approaches. To more truly mimic the in vivo environment and 3D structure of neuronal tissues, Wallace and colleagues [646] developed a facile method to 3D bioprint brain-like layered structures using a formulated bioink of RGD-modified gellan gum (RGD-GG) and primary cortical neurons (Fig. 16). The bioink was optimised with no need for massive bioprinting apparatus. The printed bioconstructs can support neuron viability and neural network formation. Overall, these bioprinted brain-like structures are expected to offer opportunities to replicate in vitro more realistic 3D brain architecture and improve our understanding of brain functions, brain injuries and neurodegenerative diseases at a tissue or organ level.

5.2.2. IPN bioinks

The IPN bioinks refer to the bioink formultions of designed IPN hydrogels or hydrogel precursors and encapsulated cells (i.e. IPN bioinks = IPN hydrogel materials + cells). Table 28 lists the summaries of examples of reported IPN bioinks for 3D bioprinting. To improve the printability of mechanically robust DN hydrogels for 3D bioprinting, the tough ionic-covalent entanglement (ICE) hydrogels, both physically and chemically cross-linked, have been developed to meet the requirements for 3D bioprinting process [1602]. Bakarich and colleagues [1603] demonstrated that the ICE hydrogels of alginate and PAAm can be manufactured through a coinstantaneous extrusion printing and in situ photopolymerisation. The rheological properties of ink formulations (alginate/acrylamide hydrogel precursors) were optimized to enhance the printability. Recently, Zhao and colleagues [1544] formulated an IPN hydrogel from covalently cross-linked PEG and ionically cross-linked alginate that can have high stretchability and toughness and allow cell encapsulation for 3D bioprinting (Fig. 17). These studies have demonstrated the potential of mechanically tough ICE hydrogels for 3D bioprinting for tissue engineering purposes.

5.2.3. Supramolecular bioinks

The supramolecular bioinks refer to the bioink formulations of supramolecular hydrogels or hydrogel precursors and encapsulated cells (i.e. supramolecular bioinks = supramolecular hydrogel materials + cells). The dynamic supramolecular interactions can impart distinct advantages to supramolecular hydrogels for 3D bioprinting. First, introducing these dynamic interactions will be able to promote printability and productivity. Incorporating such supramolecular functionalities can also offer tailorable surface structures to modulate cell–scaffold interactions [1630]. Furthermore, the introduced supramolecular interactions in hydrogel scaffolds can contribute to favorable mechanical properties. Duncan and colleagues [1631] designed a supramolecular polypeptide–DNA hydrogel system allowing 3D bioprinting. Designed cell-laden constructs can be printed through alternatively depositing two complementary bioink components A

Table 27
Summaries of reported single component bioinks for 3D bioprinting.

3D bioprinting techniques	Ink materials	Cells	Applications	Refs
3D bioplotting	Alginate	ATDC5	Cartilage tissue engineering	[1587]
Solid freeform fabrication	Alginate	Rat heart endothelial cells	Tissue engineering	[1581]
Extrusion bioprinting	Alginate	L929 mouse fibroblasts	Vascular tissue engineering	[1509]
Extrusion bioprinting	Norbornene modified alginate	Mouse L929 fibroblasts	Tissue engineering	[1588]
Extrusion bioprinting	Collagen	NIH 3T3 fibroblasts	Tissue engineering	[1589]
Extrusion bioprinting	Collagen	Bovine fibrochondrocytes	Soft tissue implants, cartilage tissue engineering	[1590]
Extrusion bioprinting	Collagen	Bovine primary articular chondrocytes	Cartilage tissue engineering	[1591]
Extrusion bioprinting	RGD modified gellan gum	Mouse primary cortical neurons	Brain-like structures for understanding of brain injuries and neurodegenerative diseases	[646]
Extrusion bioprinting	Pectin methacrylate	Human neonatal dermal fibroblasts	Dermal tissue engineering	[1592]
Extrusion bioprinting	Pluronic F127 diacrylate	Engineered E. coli	Wearable devices	[1593]
Digital light processing, extrusion bioprinting	Allylated gelatin	Human articular chondrocytes	Chondrogenic tissue engineering	[1594]
Extrusion bioprinting	Gelatin	Vascular smooth muscle cells	Vascular prosthesis	[1595]
3D bioplotting	GelMA	Hepatocarcinoma HepG2 cells	Tissue engineering	[1513]
Extrusion bioprinting	GelMA	Human umbilical vein endothelial cells (HUVECs)	Tissue engineering, pharmaceutical screening	[1596]
2 P P	GelMA	Human adipose-derived stem cells	Tissue engineering	[1597]
Extrusion bioprinting	MeHA	Human bone marrow-derived mesenchymal stromal cells	Bone regenerative medicine	[1598]
3D bioplotting	Spider silk	Mouse BALB/3T3 fibroblasts	Tissue engineering	[1599]
Stereolithography	PEGDA	Human adipose-derived stem cells (hADSCs)	Tissue engineering, tissue repair	[1600]
Stereolithography	PEGDMA	Human dermal fibroblasts	Tissue engineering	[1601]

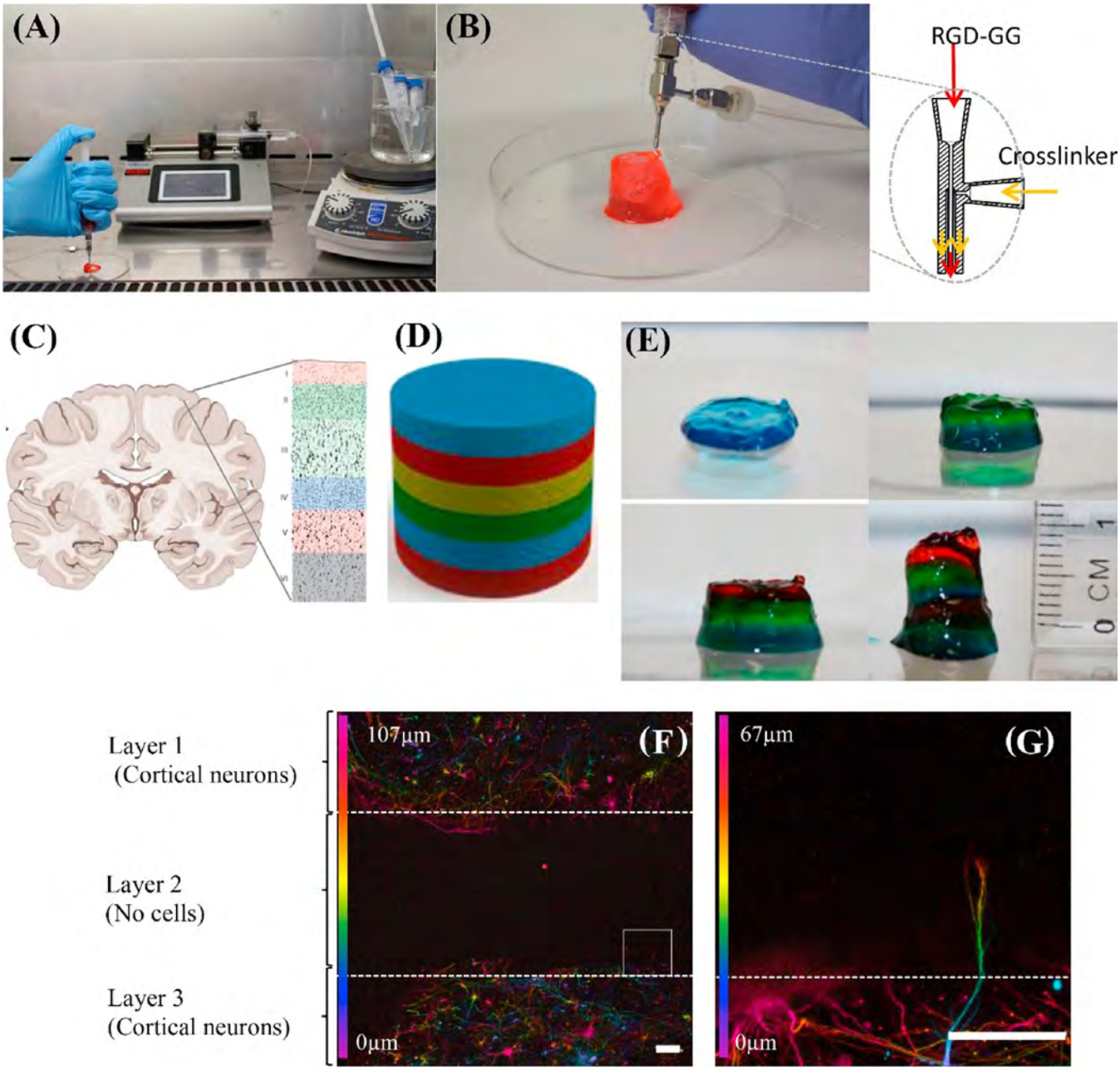


Fig. 16. 3D bioprinting of layered brain-like structures. (A) The 3D bioprinting system inside a biosafety cabinet. (B) A 3D printed construct. The right side is a diagram of the extrusion tip. (C) A representation of the six-layer brain structure in human cortex. (D) The proposed design for the artificial brain-like layered structure. (E) The 3D bioprinting process to fabricate the brain-like layer structure, in which each colour stands for one layer. (F) Confocal microscopy images of encapsulated neurons in each layer after culturing for 5 days. The image is coloured for neuron distribution through z-axis. (G) The enlarged view of the square area in (F) indicates an axon penetrating into the adjacent layer. (Scale bars 100 μm).
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(polypeptide–DNA conjugate) and B (complementary DNA linker). The self-healing and high mechanical strength allow printing millimeter-scale constructs without collapse. The encapsulated cells in printed bioconstructs can show normal cellular functions, indicating the favorable biocompatibility (Fig. 18). In the rapidly developing field of 3D bioprinting, supramolecular bioinks on the basis of host–guest interactions hold great promise for the two directions: (i) directly print gradient bioconstructs, and (ii) first print different bioconstructs and then self-assemble into more complex structures through molecular recognition. In this way one can achieve modular bioprinting and Lego-like assembly to reach larger and more complicated hydrogel-based bioconstructs (e.g. various tissues, organs at real size) with controllable physical, chemical and cellular components at higher resolutions. With regard to the concept of self-assembly, it can be applicable at all scales and achieve flexible manufacturing of 3D material structures organised on every scale, which is the distinct advantage that traditional manufacturing techniques do not have [1632]. Since the self-assembly process is independent on instruments, its parallel large-scale high-efficiency manufacturing can provide new ideas for 3D bioprinting with a particular significance on controlled macro-assembly biofabrication.

5.2.4. *Nanocomposite bioinks*

Nanocomposite bioinks refer to the bioink formulations of nanoengineered hydrogels or hydrogel precursors and encapsulated cells

Table 28
Summaries of reported IPN bioinks for 3D bioprinting.

3D bioprinting techniques	Ink materials	Cells	Applications	Refs
Extrusion bioprinting	Agarose/alginate	Chondrocytes	Cartilage tissue engineering	[1604]
Inkjet bioprinting	Alginate/collagen	Human amniotic fluid-derived stem cells, canine smooth muscle cells, bovine aortic endothelial cells	Tissue engineering	[1605]
Extrusion bioprinting	Alginate/Pluronic F127	Human bone marrow mesenchymal stem cells	Tissue engineering	[1606]
Extrusion bioprinting	Alginate/platelet-rich plasma (PRP)	human umbilical vein endothelial cells (HUVECs)	Growth factor delivery, vascular tissue engineering	[1607]
Extrusion bioprinting	Alginate/fibrinogen/gelatin	NIH 3T3 eGFP mouse fbroblasts, human dermal fbroblasts	Skin tissue engineering	[1608]
3D bioplotting	Alginate/carboxymethyl chitosan/agarose	Induced pluripotent stem cells (iPSCs), human neural stem cells	Pharmaceuticals development, regenerative medicine, neural tissue engineering	[1609,1610]
3D bioplotting	Alginate/gelatin	Human osteogenic sarcoma SaOS-2 cells	Bone tissue engineering	[1611]
Extrusion bioprinting	Alginate/gelatin	L929 mouse fbroblasts	Tissue engineering	[1612]
Extrusion bioprinting	Alginate/gelatin	Aortic root sinus smooth muscle cells, aortic valve leaflet interstitial cells	Aortic valve conduits	[1613]
3D bioplotting	Alginate/methylcellulose	Human mesenchymal stem cells	Tissue engineering	[1510]
3D bioplotting	HA/dextran methacrylate	Equine articular chondrocytes	Cartilage tissue engineering	[1514]
Extrusion bioprinting	HA/methylcellulose	Sheep adipose tissue-derived mesenchymal stem cells	Tissue engineering	[1614]
Extrusion bioprinting	HA-g-PNIPAAm/MeHA	Bovine articular chondrocytes	Cartilage tissue engineering	[1540]
3D bioplotting	Alginate dialdehyde/gelatin	Human dermal fibroblasts, human umbilical vein endothelial cells	Vessel tissue engineering	[1615]
Extrusion bioprinting	HA/κ-carrageenan/fumed silica, GMHA/κ-carrageenan/fumed silica	Pseudomonas putida, Acetobacter xylinum	Bioremediation, bacterial cellulose production	[1616]
3D bioplotting	Gelatin/alginate	Bacillus subtilis	Biomaterials, biotechnology, biomedicine	[1617]
Multiphoton lithography	Gelatin/bovine serum albumin	Staphylococcus aureus, Pseudomonas aeruginosa	Assessment of bacterial communication and antibiotic resistance	[1618]
3D bioplotting	GelMA/collagen	Breast adenocarcinoma cells MCF-7	Tissue engineering	[1619]
Microscale continuous optical bioprinting	GelMA/GMHA	Human umbilical vein endothelial cells (HUVECs), 10T1/2 s, HepG2	Tissue engineering, prevascularization	[1620]
Extrusion bioprinting	GelMA/gellan gum	Chondrocytes	Cartilage regeneration	[1621]
Extrusion bioprinting, stereolithography	GelMA–PEO	HepG2, human umbilical vein endothelial cells (HUVECs), NIH/3T3 fibroblasts	Tissue engineering	[1622]
Stereolithography	GelMA/PEGDA	Human bone marrow mesenchymal stem cells	Cartilage tissue engineering	[1623]
Stereolithography	GelMA/PEGDA	NIH 3T3 fibroblasts	Tissue engineering, bioengineering	[1624]
Digital light processing	GelMA/PVA methacrylate	Human endothelial colony forming progenitor cells, human bone marrow mesenchymal stromal cells	Osteochondral tissue engineering	[1523]
Extrusion bioprinting	Gellan gum/PEGDA	Murine bone marrow stromal cells, mouse MC3T3-E1	Human-on-a-chip, tissue engineering	[1625]
Extrusion bioprinting	Matrigel/alginate	Endothelial progenitor cells	Vascularization, bone regeneration	[1626]
Extrusion bioprinting	Silk fibroin/gelatin	Human nasal inferior turbinate tissue-derived mesenchymal stromal cells (hTMSCs)	Osteogenic and chondrogenic tissue engineering	[1627]
Digital light processing	Silk fibroin/PEGA	NIH 3T3 fibroblasts	Artificial skin model	[1628]
Extrusion bioprinting	Silk fibroin/PEG	Human bone marrow mesenchymal stem cells	Tissue regeneration	[1629]

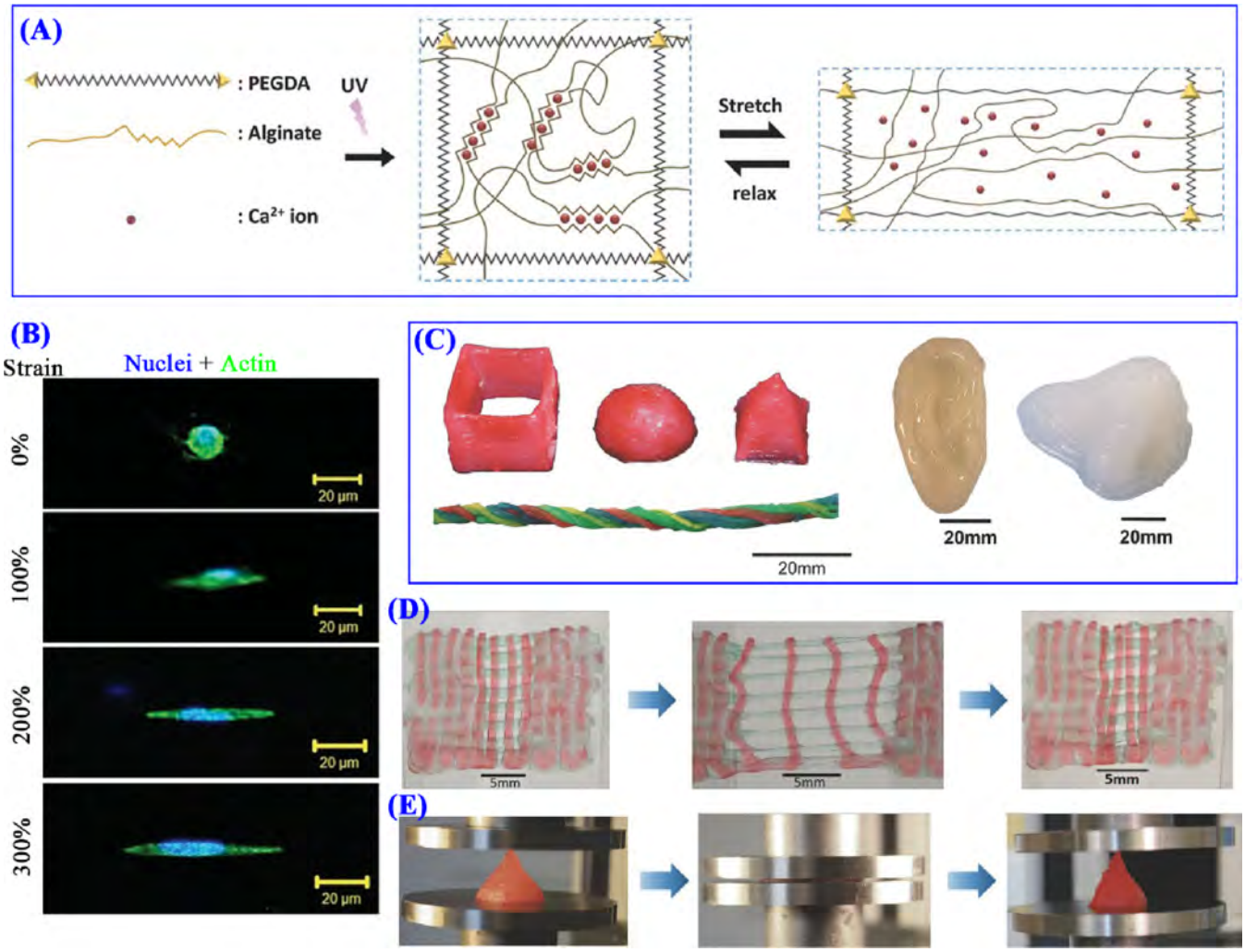

Fig. 17. 3D bioprinting of tough IPN hydrogels into cellularized constructs. (A) Diagram of the tough hydrogel made from covalently cross-linked PEG and ionically cross-linked alginate. (B) Deformation of the hMSC (nuclei in blue, actin in green) encapsulated in the hydrogel matrix and stretched to different strains. (C) 3D constructs printed with the hydrogel. Left to right: hollow cube, hemisphere, pyramid, twisted bundle, ear, nose. (D) A printed mesh with the hydrogel can undergo cyclic mechanical stretch and recovery. (E) A printed pyramid can undergo 95 % compressive strain and original shape recovery after relaxation.
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(i.e. nanocomposite bioinks = hydrogel materials + nanoparticles + cells). The incorporation of a small quantity of nanoparticles into the hydrogel matrices can lead to dramatic changes in a variety of physical, chemical and biological properties, offering many opportunities to regulate printability, functionality, degradability and usability. Due to the organic–inorganic composite feature, nanocomposite hydrogels have been increasingly formulated with cells into advanced bioinks for 3D bioprinting process. Table 29 lists the summaries of examples of reported nanocomposite bioinks for 3D bioprinting.

5.2.5. *Multimaterial bioinks*

Multimaterial bioinks refer to the bioink formulations of multiple ink materials and encapsulated cells. For the types of ink materials, at least one of them is the hydrogels or hydrogel precursors for cell encapsulation. Both multiple material types and multiple cell types can be used to formulate the multimaterial bioinks. The primary challenge of 3D bioprinting is to reproduce the complex ECM microstructures and multiple cell types at increasingly accurate high resolutions so as to recapitulate the various biological functions of tissues and organs. This demands the elaborate combinations of multimaterials and multiple cell types to print complex 3D bioconstructs that possess multiple morphological, structural, mechanical and functional compositions and characteristics. Nowadays, multimaterial hydrogels are being increasingly studied to surmount the limitation of single component hydrogels for 3D bioprinting applications. Although bioprinting a brain analogue does not mean it will develop into a real brain, multimaterial 3D bioprinting makes a closer step to the ultimate dream. Examples of the distinct advantages include: (i) establish gradient bioconstructs with biomimetic structural and functional properties, (ii) achieve multilevel mechanical match with surrounding tissues, (iii) combine multiple cell types in one complex construct with location specificity through printing multiple cell-laden hydrogels. Table 30 lists the summaries of examples of reported multimaterial bioinks for 3D bioprinting.

Recently, Khademhosseini and colleagues [1647] developed a continuous multimaterial extrusion bioprinting platform with the distinct ability to extrude coded multimaterial bioinks, whose reservoirs can be switched fast and smoothly, allowing for the rapid fabrication of complex tissue- and organ-like constructs (Fig. 19). With a wide range of bioink formulations from shear-thinning to conductive hydrogel materials, they demonstrated the multimaterial bioprinter capable of fabricating the miniaturized multiple cell-laden bioconstructs, gradient constructs and multicomponent bioelectronics with strong capacities in continuity and printing speed. Overall, this platform is expected to be extended to a broad variety of bioinks and offer convenience for 3D

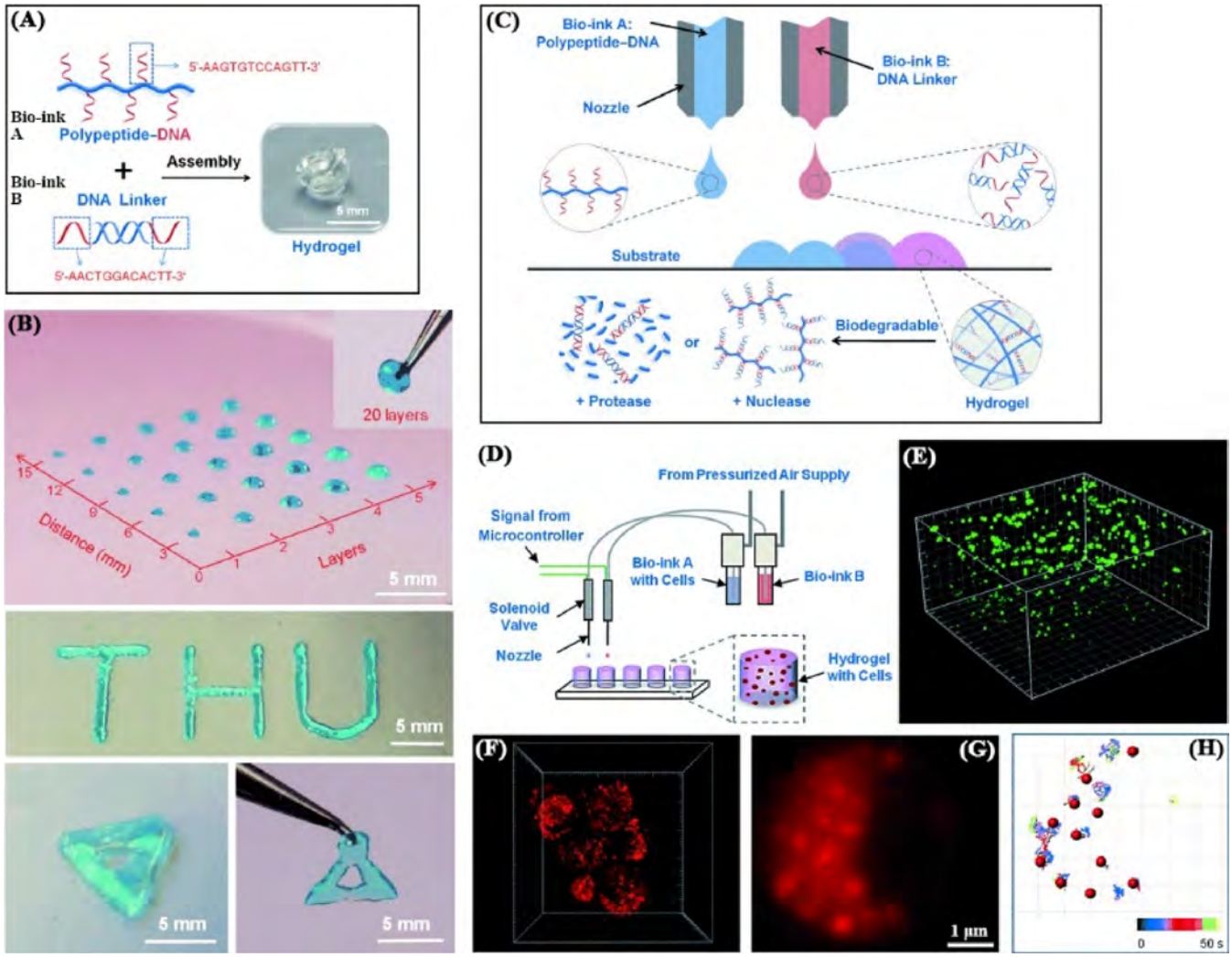

Fig. 18. 3D bioprinting of supramolecular hydrogels. (A) Fabrication of supramolecular polypeptide–DNA hydrogels by mixing two-component bio-ink A and bio-ink B. (B) 3D printing of the supramolecular hydrogels into various 3D constructs. (C) 3D printing of the hydrogels into arbitrarily designed 3D constructs that are responsive to proteases and nucleases for degradation. (D) 3D bioprinting of the cell-laden hydrogel bioink. (E, F) 3D stack images of the encapsulated cells in printed hydrogel scaffold stained in green and red. (G) Fluorescence image of a single cell in the printed scaffold. (H) Tracking of dynamic organelles and trajectories from inside the cell in (G).
Reproduced with permission [1631]. Copyright 2015, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

bioprinting of highly complicated bioconstructs and biomedical devices.

Lewis and colleagues [1652] reported an approach for engineering tissue constructs with vasculature, ECM and multiple cell types on the basis of multimaterial 3D bioprinting (Fig. 20**A-D**). The engineered bioconstructs can pave the way for screening drugs, studying angiogenesis, wound healing and stem cell niches and hopefully fabricating 3D functional tissues and organs. Rutz and colleagues [1553] developed bioink formulations from gelators (gelatin, GelMA, 4-arm PEG amine) and chemical cross-linker (PEGX). The PEGX cross-linking can impart tailorable viscosity and biodegradability without endangering cellular activity (Fig. 20**E-F**). The PEGX bioinks are expected to 3D bioprint tissue constructs that are more biomimetic, complicated and customized for tissue engineering applications.

5.3. Advances in hydrogel-based 3D bioprinting

5.3.1. New 3D bioprinting techniques

While developing a variety of advanced bioinks, researchers are also devoting to the development of advaced 3D bioprinting methods. To tackle the challenge of creating 3D vascularized constructs of complexity and large size, Atala and colleagues [1655] developed a 3D bioprinting system termed integrated tissue–organ printer (ITOP) capable of fabricating human-scale tissue constructs with any shape, structural integrity and microchannel incorporation (Fig. 21**A-C**). With further development, the ITOP system can produce more complicated tissues and organs for tissue engineering. Feinberg and colleagues [1649] reported development of the 3D bioprinting technique named freeform reversible embedding of suspended hydrogels (FRESH). The FRESH system utilised the thermoreversible gelatin slurry support bath to enable depositing hydrogels into 3D complex constructs. By extruding and embedding hydrogel inks in the gelatin hydrogel support bath, FRESH printing process can maintain the desired 3D structures and remarkably boost bioink printability. Based on 3D imaging data, the FRESH can 3D print complex bioconstructs such as explanted embryonic chick heart with high printing fidelity and spatial resolution (Fig. 21**D-E**). Despite requiring further investigation to achieve direct 3D bioprinting of intended tissues and organs, the FRESH technique should be a catalyst in academic or commercial biofabrication laboratories to elicit breakthroughs for a variety of tissue engineering applications. Recently, Kong and colleagues [1659] presented an embedded 3D bioprinting strategy termed freeform reconfigurable embedded all-

Table 29
Summaries of reported nanocomposite bioinks for 3D bioprinting.

3D bioprinting techniques	Matrix materials	Nanoparticles	Cells	Applications	Refs
Extrusion bioprinting	Agarose	Laponite	HeLa	Tissue engineering	[1633]
Extrusion bioprinting	Alginate	Nanocellulose	Human nasoseptal chondrocytes	Cartilage tissue engineering	[1634]
3D bioplotting	Alginate	Polylactic acid nanofiber	Human adipose-derived stem cells	Musculoskeletal soft tissue engineering	[1635]
3D bioplotting	Alginate/methylcellulose	Laponite	Human mesenchymal stem cells (hTERT-MSC)	Skeletal tissue engineering	[1552]
3D bioplotting	Alginate/methylcellulose	Pt(II) meso(2,3,4,5,6-pentafluoro)phenyl porphyrin	Human mesenchymal stem cells (hTERT-MSC), microalgae	O_2 imaging, tissue engineering	[1636]
3D bioplotting	Alginate dialdehyde/gelatin	Bioactive glass	Osteoblast-like MG-63	Bone tissue engineering	[1545]
Extrusion bioprinting	κ-Carrageenan	Laponite	MC3T3-E1 mouse preosteoblasts	Tissue regeneration	[1637]
Extrusion bioprinting	Chitosan	Hydroxyapatite	MC3T3-E1	Bone tissue engineering	[1638]
Extrusion bioprinting	PEG	Laponite	Murine MC3T3-E1 preosteoblasts	Tissue engineering	[1639]
Extrusion bioprinting	PEGDA	Nanocellulose	Saccharomyces cerevisiae	Biocatalyst	[1640]
Inkjet bioprinting	PEGDMA	Bioactive glass, hydroxyapatite	Human bone marrow-derived mesenchymal stem cells	Bone tissue formation	[1641]
Extrusion bioprinting	PEG/PEG dithiothreitol	Laponite	HUVECs	Therapeutic protein delivery, tissue engineering	[1642]
3D bioplotting	Gelatin	Laponite	Human bone marrow stromal cells	Hard and soft tissue reparation	[1643]
Extrusion bioprinting	GelMA	Au nanorod	Rat cardiac fibroblasts, cardiomyocytes	Cardiac tissue engineering	[1644]
Extrusion bioprinting	GelMA/κ-carrageenan	Laponite	Mouse preosteoblast MC3T3-E1	Tissue regeneration	[1645]

liquid (FREAL) bioprinting. They demonstrated the capability of FREAL to create the complicated, freeform and reconfigurable aqueous-in-aqueous 3D structures using the bioink formulation of aqueous two-phase systems, and the FREAL printed all-aqueous microarchitectures could be preserved for over 10 days. This bioprinting approach has the potential to be applied to create various vascularized complex constructs for tissue engineering and in vitro tissue-on-a-chip applications.

Recently, Grigoryan and colleagues [1660] successfully established the designs of multivascular networks and intravascular topologies in photopolymerizable hydrogels (PEGDA) for projection stereolithography with food dye additives as biocompatible photoabsorber. They demonstrated the elaborate design of entangled multivascular networks (e.g. axial vessel and helix, interpenetrating Hilbert curves, bicontinuous cubic lattice, torus and (3,10) torus knot) from the 3D space-filling mathematical topologies and investigated human red blood cell (RBC) oxygenation and flow during tidal ventilation and distension (Fig. 22). The developed 3D bioprinting platform termed stereolithography apparatus for tissue engineering (SLATE) can be used to create various tissue constructs encapsulating mammalian cells for rapid biofabrication. The production of biocompatible hydrogels comprising functional multivascular networks and intravascular topologies will provide a powerful toolbox for studying various biological phenomena related to valves, fluid mixers, nutrient delivery, intervascular transport, etc.

5.3.2. 4D bioprinting of cell-laden hydrogels

5.3.2.1. Definition of 4D bioprinting. The concept of 4D bioprinting has emerged recently as the next-generation biofabrication technology by integrating the time dimension with 3D bioprinting. Herein, according to the definition by Ashammakhi and colleagues [196], the term 4D bioprinting refers to 3D printing of cell-laden biomaterial inks (i.e. bioinks) where the printed bioconstructs is capable of shape transformation as active reponse to varieties of external and/or intrinsic (e.g. cell forces) stimuli (Fig. 23). This definition is different from the one given by Gao and colleagues [198], which also involves the maturation of 3D printed tissue constructs over time to recapitulate tissue/organ functions (the construct geometry may retain static). Through the combination of stimuli-responsive bioinks and advanced 3D bioprinting techniques, the 4D bioprinting is aimed at fabricating dynamic complex 3D bioconstructs capable of conformational changes in response to various stimuli. The capability of 4D bioprinting to manufacture such dynamic shape-transforming bioconstructs can enable more precise imitation of the dynamics of natural tissues and organs. In addition to the field of tissue engineering and regenerative medicine, a wide range of other research and application fields, such as biorobotics, bioelectronics, bioactuators, biosensors and biomedical devices, are expected to benefit from the advancements of 4D bioprinting technology. For the more detailed information of 4D bioprinting, we refer the readers to some excellent review papers [195,196,198,199].

5.3.2.2. Hydrogel-based 4D bioprinting. Kirillova and colleagues [1661] recently demonstrated an advanced 4D bioprinting strategy of shape-morphing hydrogels (methacrylated alginate and hyaluronic acid biopolymers) for fabricating hollow cell-laden self-folding tubes (Fig. 24). This approach was capable of unprecedented control of the tube diameters and structures at high resolution and allowed the production of internal tubes of 20 μm average diameter, which is comparable with the smallest blood vessels. The printed mouse bone marrow stromal cells can maintain favorable viability in the hydrogel-based self-folding tubes. Overall, this work has presented the 4D bioprinting of reconfigurable bioconstructs with dynamic tailorable responsiveness and functionality, opening a new avenue for creating cell-laden shape-shifting constructs for tissue engineering and regenerative medicine purposes. Using stereolithography-based 4D bioprinting method, Zhang and colleagues [1662] also developed

Table 30
Summaries of reported multimaterial bioinks for 3D bioprinting.

3D bioprinting techniques	Ink materials	Cells	Applications	Refs
Extrusion bioprinting	Agarose, hyaluronan/ PEG tetracrylate cross-linker	NIH 3T3	Bioartificial vascular grafts	[1646]
Extrusion bioprinting	Alginate/gelatin	Aortic root sinus smooth muscle cells, aortic valve leaflet interstitial cells	Heart aortic valve conduits	[1613]
Extrusion bioprinting	GelMA/alginate, GelMA/alginate/hydroxyapatite, Alginate/ carbon nanotubes/DNA, nanosilicate, alginate, PEGDA/ alginate	Human dermal fbroblasts (HDFs), HepG2 human hepatocellular cells, human mesenchymal stem cells (hMSCs), HUVECs, MC3T3-E1 preosteoblasts	Tissue engineering, biomedical devices	[1647]
Extrusion bioprinting	Alginate, agarose/gelatin	Human mesenchymal stem cells	Tissue engineering	[1648]
3D bioplotting	Alginate/methylcellulose, calcium phosphate cement	Human mesenchymal stem cells (hTERT-MSC)	Bone tissue engineering	[1522]
Extrusion bioprinting	Alginate, collagen, Matrigel, fibrinogen, hyaluronic acid, bovine serum albumin	C2C12 myoblasts, MC3T3-E1	Tissue engineering	[1649]
Extrusion bioprinting	Matrigel, gelatin/fbrin, GelMA	Human neonatal dermal fbroblasts, spinal neuronal progenitor cells, oligodendrocyte progenitor cells	Treatment of neurological diseases and spinal cord injury	[1650]
Extrusion bioprinting	MeHA/GelMA	Human valve interstitial cells	Heart valve tissue engineering	[1651]
Extrusion bioprinting	GelMA, Pluronic F127, silicone elastomer	10T1/2 fibroblasts, human neonatal dermal fibroblasts (hNDFs)	Vascular tissue engineering	[1652]
Extrusion bioprinting	GelMA, Pluronic F-127	articular cartilage-resident chondroprogenitor cells, bone marrow mesenchymal stromal cells	Cartilage regeneration, articular cartilage model	[1653]
Extrusion bioprinting	GelMA/PEG cross-linker, fibrinogen/PEG cross-linker	HUVECs	Tissue engineering	[1553]
Extrusion bioprinting	Gelatin/fibrinogen	Human bone marrow-derived mesenchymal stem cells (hMSCs), human neonatal dermal fibroblasts (hNDFs), human umbilical vein endothelial cells (HUVECs)	Vascular tissue engineering	[1654]
Extrusion bioprinting	Gelatin/fibrinogen/HA/glycerol, PCL, Pluronic F-127	3T3 fibroblasts, C2C12 myoblasts, human amniotic fluid-derived stem cells, rabbit primary auricular chondrocytes	Tissue engineering	[1655]
Extrusion bioprinting	PEGDA/Laponite, hyaluronic acid (HA)	Primary rat osteoblasts	Bone tissue regeneration	[1656]
Stereolithography bioprinting	PEGDA, GelMA	breast cancer cells (MCF7), human umbilical vascular endothelial cells (HUVECs), NIH/3T3 fbroblasts, C2C12 skeletal muscle cells, mesenchymal stem cells (MSCs), fbroblasts, osteoblasts	Tissue engineering, regenerative medicine, biosensing	[1657]
3D bioplotting	GelMA, gelatin	Human dermal fibroblasts (HDFs), human umbilical vein endothelial cells (HUVECs)	Tissue fabrication, hydrogel-based microfluidics, self-supported perfusable hydrogel constructs	[1658]

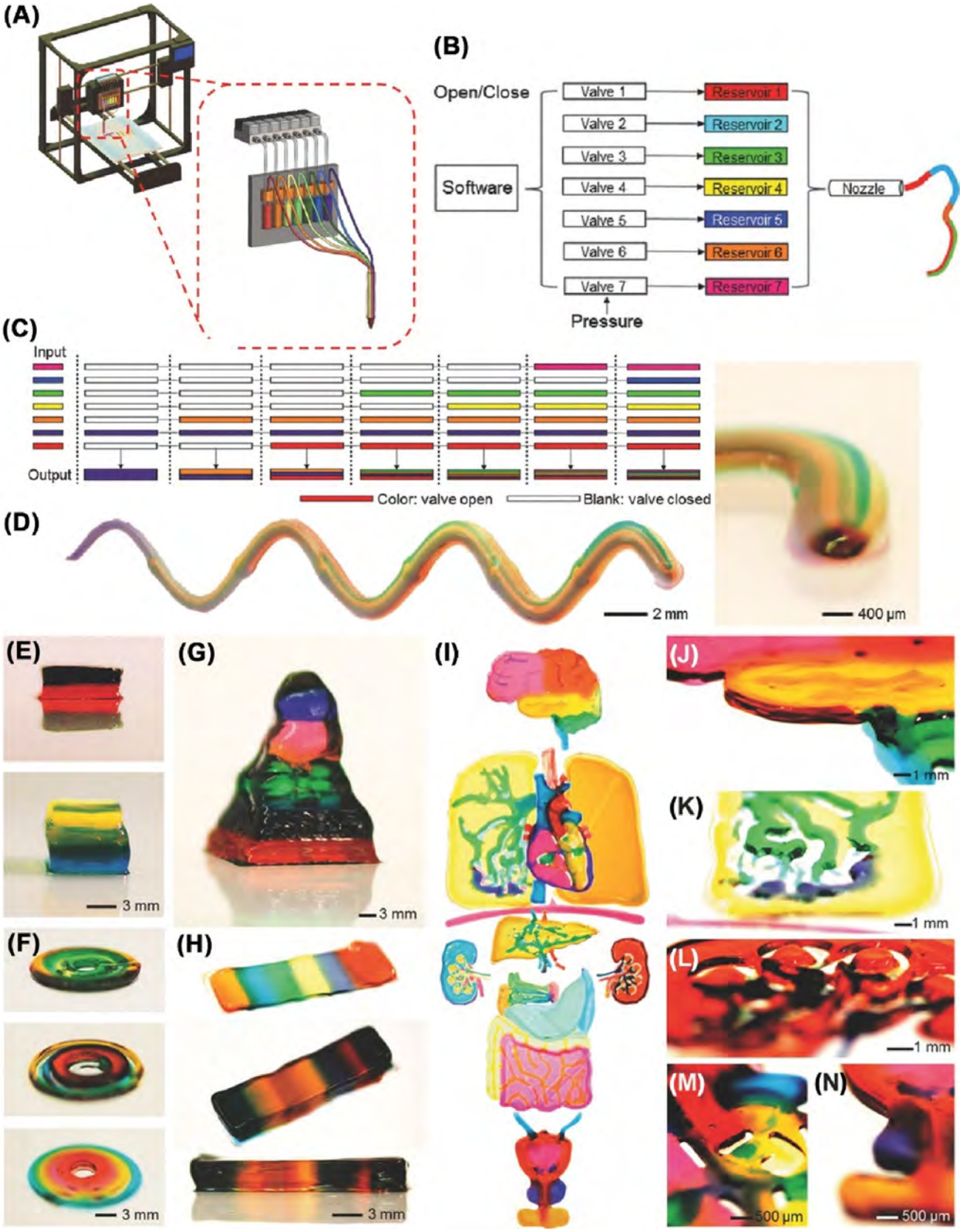

Fig. 19. Multimaterial 3D bioprinting. (A, B) Design of a digitally controlled multimaterial extrusion bioprinter with seven-channel printhead connected to reservoirs. (C) Sample code diagram for bioprinting single serpentine microfiber from 1 to 7 bioinks. (D) Photograph of the printed microfiber with the side view of the end (right). (E) Bioprinting of the 2- and 3-layer cuboid blocks. (F) Bioprinting of blood vessel analogues with 2, 3 and 4 materials. (G) Bioprinting of a 7-layer pyramid from different bioinks. (H) Bioprinting of the 3- and 10-layer blocks with 7 segments from different bioinks. (I) Bioprinting of human organ analogues from multimaterial bioinks, including brain, lung, heart, liver, kidneys, pancreas, stomach, small/large intestines, bladder, prostate. (J–N) 3D side views of organ-like structures: (J) brain, (K) lung vasculature, (L) kidney, (M) left atrium of heart, (N) bladder/prostate. Reproduced with permission [1647].

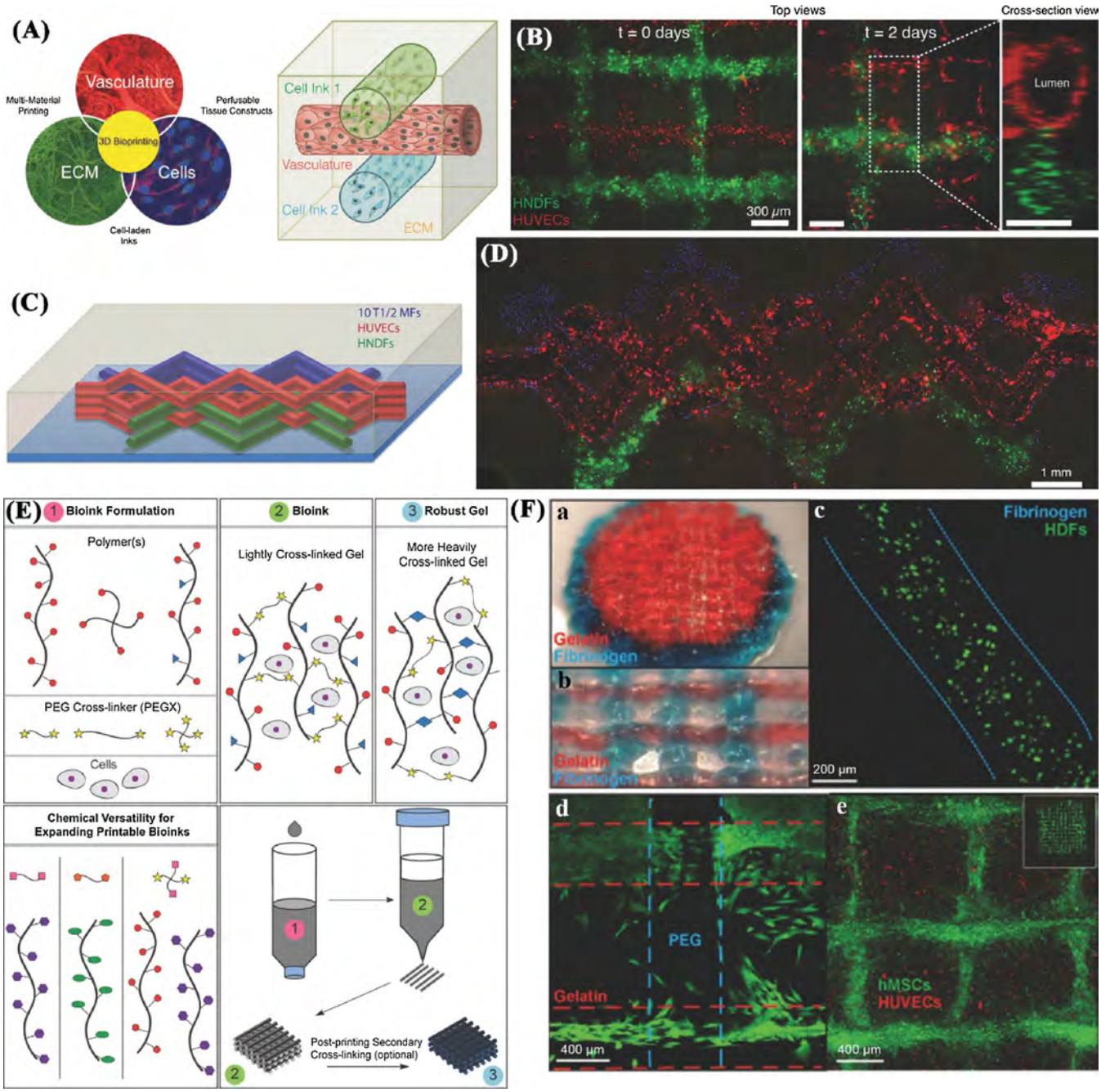

Fig. 20. Multimaterial bioinks for 3D bioprinting. (A) Diagram of a 3D bioprinting strategy to engineer tissue constructs with heterogeneous subunits. (B) Fluorescence images of an engineered tissue construct. (C) Schematic side view of a heterogeneous engineered tissue construct. (D) Composite top view image of 3D bioprinted tissue construct. Reproduced with permission [1652]. Copyright 2014, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (E) A multimaterial bioink formulated with GelMA, PEG cross-linker and cells. (F) 3D bioprinted constructs support high cellular viability and proliferation. Reproduced with permission [1553]. Copyright 2015, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

multi-responsive intelligent architectures for nerve tissue engineering, in which the stress-induced shape transformation was adopted to accomplish 4D reprogrammability. Another 4D bioprinting approach is based on the origami folding principle. By harnessing the cell traction force as biological driving force, one can achieve the self-folding and mass-production of various 3D cell-laden microstructures [1663], holding promise for biotechnology applications and biomedical devices. Last but not least, the shape chane in 4D bioprinting is expected to exert dynamic mechanical stimuli to the encapsulated cells and thus may be able to modulate cell fate e.g. adhesion, spread, migration, proliferation and differentiation [1070,1071,1664].

6. Emerging biomedical applications

6.1. Tissue engineering and regenerative medicine

Tissue engineering can apply the biology and engineering principles to develop functional substitutes for replacing, repairing and regenerating (3R) the lost, damaged or diseased tissues and organs. This field has been rapidly expanding to tackle the donor tissue limitations and transplant rejections. As detailed in section 4.1.2, scaffold designs should be capable of creating complex 3D constructs with hierarchical porous architectures and arbitrary anatomical shapes to achieve desired mass transport and mechanical properties, thereby coupling mechanical

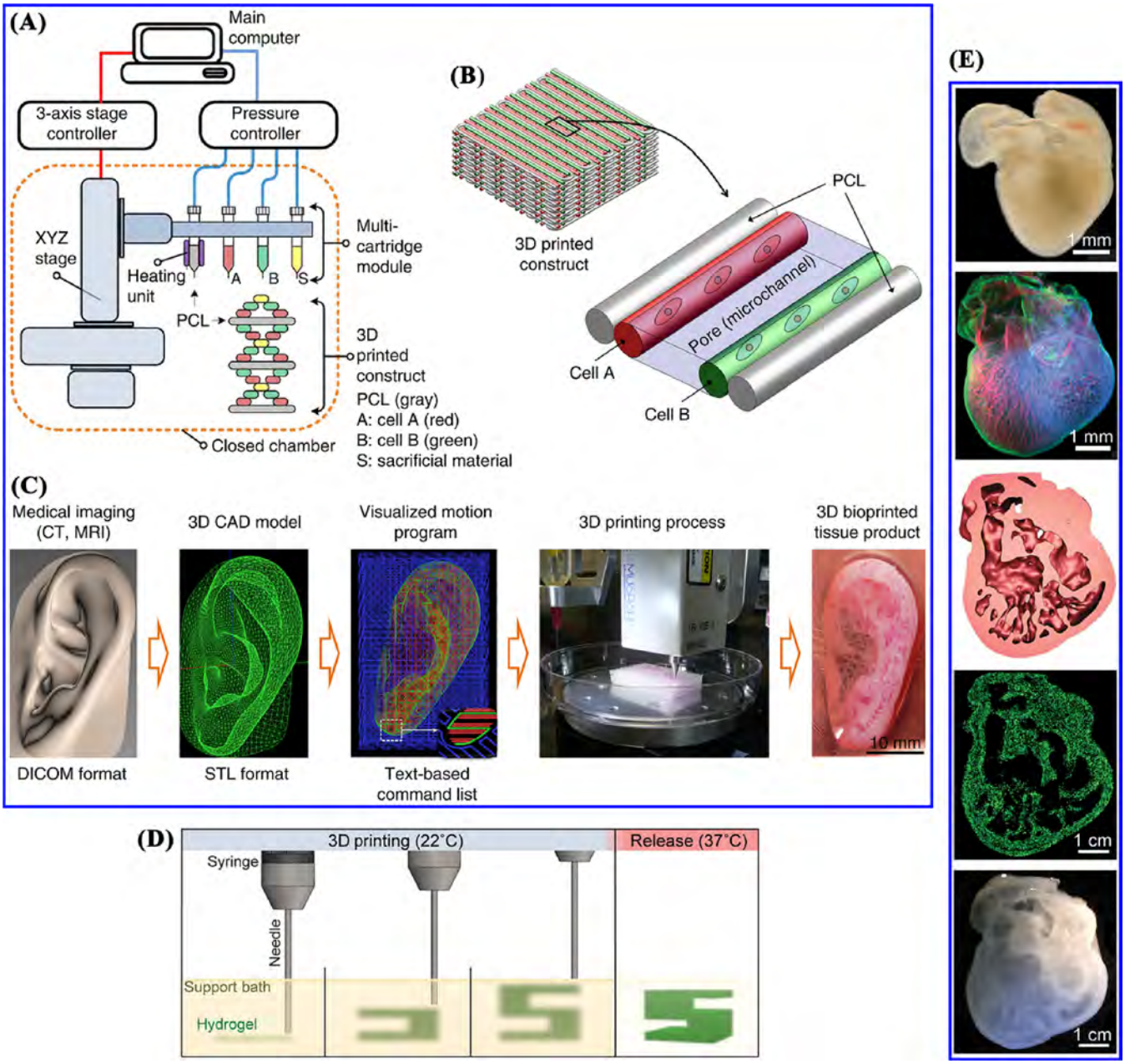

Fig. 21. ITOP system: (A) Major units of the system. (B) Schematic of 3D bioprinted construct with multiple cell-laden hydrogels and PCL support. (C) CAD/CAM process for 3D bioprinting of tissue- or organ-like structures. Reproduced with permission [1655]. Copyright 2016, Nature Publishing Group. FRESH system: (D) Schematic FRESH process of extruding hydrogel inks, cross-linking in gelatin slurry support bath and releasing 3D construct by heating and melting gelatin. (E) FRESH printed of an explanted embryonic chick heart. Reproduced with permission [1649]. Copyright 2015, The Authors, exclusive licensee American Association for the Advancement of Science. Distributed under a Creative Commons Attribution License 4.0 (CC BY).

functionality with tissue/organ regeneration. However, traditional fabrication methods e.g. solvent casting, phase separation, gas foaming, fiber bonding, freeze-drying and porogen leaching/particulate leaching have limitations for scaffold designs with complex topology structures and adequate mechanical properties [145]. As an advanced manufacturing technology with distinct advantages e.g. customizability and precision, 3D printing is an increasingly powerful tool for researchers, engineers and clinicians, and hold huge potential as an accelerator to translate tissue engineering concepts into rapid, cheap and promising clinical implementations.

6.1.1. Bone regeneration

Bone damages/defects caused by disease (e.g. osteoporosis), trauma (e.g. bone fracture), surgical resection (e.g. bone tumor therapy) and congenital bone condition require regeneration and reconstruction by customized substitutes (grafts) to repair the lost bone structure and physiological function. By now, the most widely used ways to regenerate bone defects (particularly large bone defects) include autografts and allografts. The use of autologous bone grafts is currently the clinical gold standard for bone regeneration due to superior osteoconduction and osteoinduction; however, it suffers from the problems like supply limitations and donor site complications [1665]. By contrast, bone allografts are the suboptimal choice owing to the wide availability; nevertheless, problems like immune rejection, infectious pathophoresis and decreased osteoinductivity may cause inferior bone healing. Alternatively, various synthetic biomaterials have been designed and developed as bone substitutes; nontheless, their structures and compositions often require elaborate designs to mimic native bone

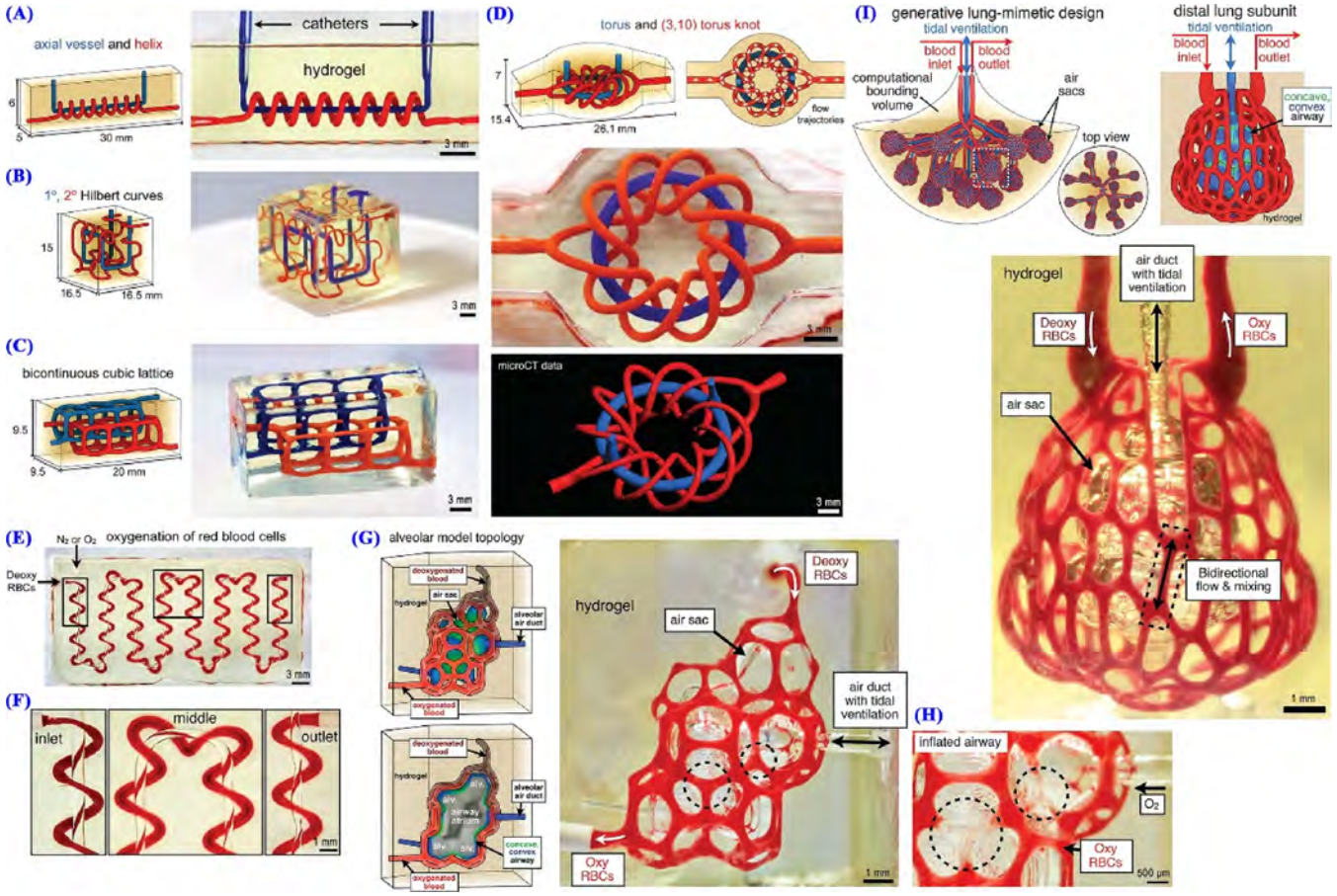

Fig. 22. Entangled multivascular networks: (A–D) Adaptation of the 3D space-filling mathematical curves to the entangled vessel topologies in hydrogel. (E) The tessellation of axial vessel along serpentine path. (F) Magnified images of the three rectangular areas in (E). Tidal ventilation and oxygenation: (G–H) Structural design of a vascularized alveolar model topology in hydrogel (left) and image of the printed hydrogel during RBC perfusion and O_2 ventilation (right). (I) Schematic of generative lung-mimetic design with distal lung subunit (upper) and image of the printed hydrogel during RBC perfusion and O_2 ventilation (lower). Reproduced with permission [1660]. Copyright 2019, American Association for the Advancement of Science.

tissues to enable bone regeneration function. Therefore, novel concepts and strategies for bone tissue engineering are urgently needed, among which 3D printing/bioprinting is expected to fabricate bone-biomimetic complex constructs having the anatomical characteristics of natural bone tissues, thereby enhancing the bone regenerative ability [1666,1667].

While engineering complex constructs for bone reconstruction and regeneration through 3D printing/bioprinting, the following information on bone structure and function should be fully taken into consideration to enable biomimetic designs: (1) Bone is hierarchically structured (Fig. 25) comprising macrostructures (cancellous bone, cortical bone), microstructures (trabeculae), submicrostructures (lamellae), nanostructures (fibrillar collagen, embedded minerals) and subnanostructures (minerals, proteins). (2) Bony hierarchical architectures provide mechanical function, guarantee mass exchange and maintain cell activities [1668]. (3) Bony structure is dynamic and needs dynamic modulation i.e. bone remodeling. (4) Bone tissues are highly vascularized and linked with rich blood vessels in periosteum, allowing active mass transfer e.g. oxygen, nutrients, wastes and blood cells with the outside. (5) The organic unity of bony structure, host cells (e.g. mesenchymal stem cells) and biomolecules endows bone tissues with the intrinsic powerful self-healing, self-renewing and self-regenerating capacity [1669].

Kang et al. [1655] adopted the home-made ITOP system to biofabricate a mandible bone fragment with the similar shape and size for traumatic facial reconstruction to meet the reconstruction requirements of arbitrary-shape mandible bone defects (Fig. 26A). The bioinks were formulated with the composite hydrogel (gelatin/fibrinogen/HA/glycerol) and human amniotic fluid-derived stem cells. It was demonstrated that the bioprinting process did not negatively affect cellular viability and that inducing the osteogenic differentiation for 28 d showed calcium deposition capacity in the cell-laden hydrogel. Khademhosseini and colleagues [1671] engineered a vascularized bone tissue construct using extrusion-based bioprinting strategy to mimic the whole bony architecture (Fig. 26B). The GelMA hydrogel cylinders were first individually printed out and then piled up to create pyramidal architectures comprising 28 rods. The individual printing process enables the composition customizability and precise control of the whole architecture at the single rod resolution. As a proof-of-concept model, they further printed the vascularized bone tissue constructs of predefined architectures using bioink formulations with GelMA and cells (HUVECs, hMSCs). The perfusable channels, formed by the fast degradation of the soft hydrogel inner core (5 % GelMA), can act as the central blood vessels in the cell-laden bioconstructs. The bioprinted bone tissue constructs are capable of supporting cell survival, proliferation and maturation and maintaining high structural stability over a desired period. Overall, this study has demonstrated a bone biofabrication approach enabling localized control of osteogenic and angiogenic niches and establishment of biophysical and/or biochemical gradients within the bioprinted bone constructs. Cui et al. [1672] developed a proof-of-concept design of dual 3D bioprinting platform for fabricating vascularized bone biphasic constructs comprising hard mineral structures encompassed by cell-laden GelMA hydrogels to simulate native bone, as shown in Fig. 26**C**. Such an integrated strategy can

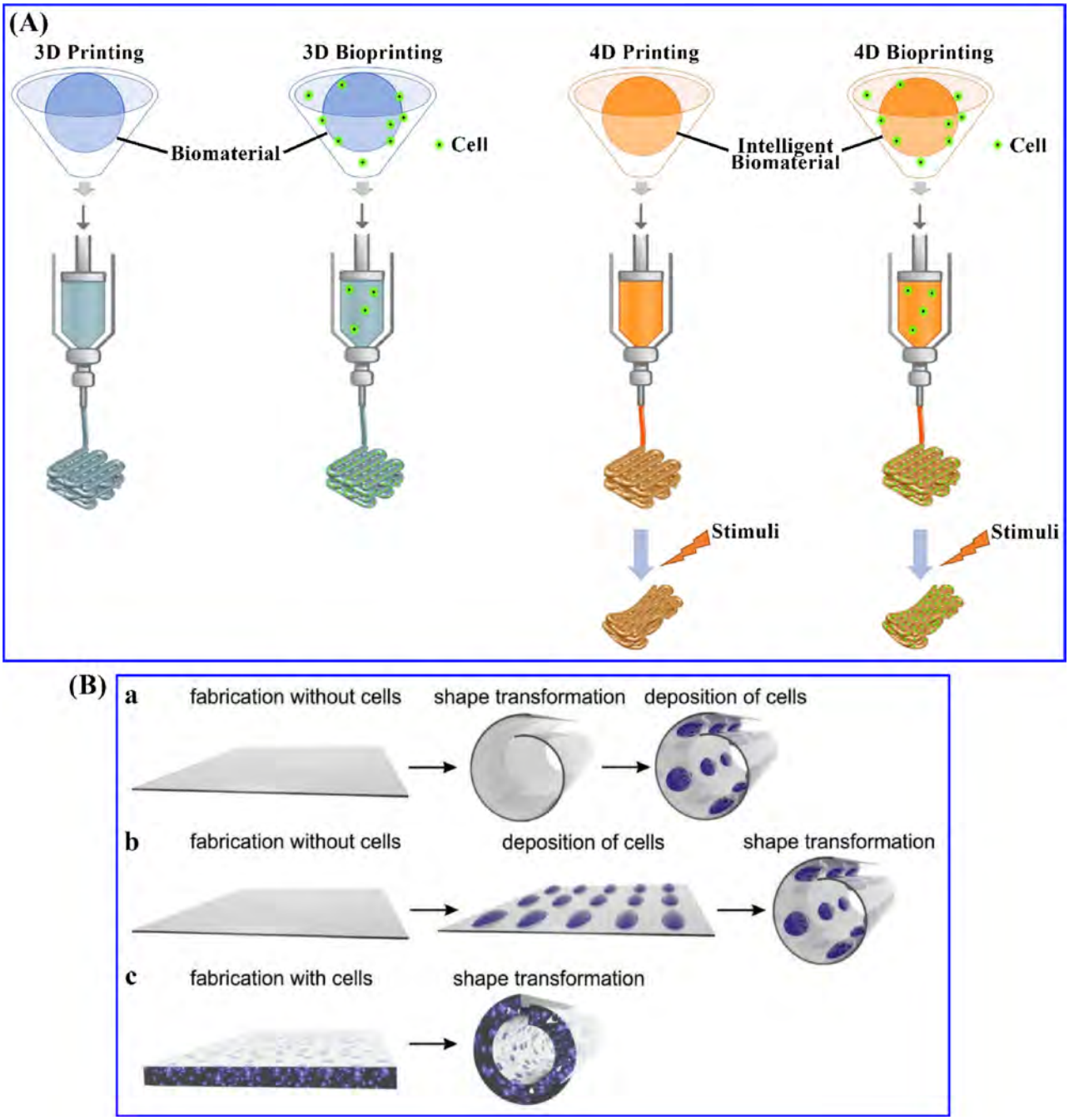

Fig. 23. (A) Schematic diagram of 3D printing, 3D bioprinting, 4D printing and 4D bioprinting with ink formulations from conventional biomaterials, cells and intelligent biomaterials. Cells are encapsulated within ink materials (e.g. hydrogels, hydrogel precursors) to formulate bioinks for bioprinting. Reproduced with permission [196]. Copyright 2018, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (B) Illustrative 4D biofabrication of 3D cellularized constructs through shape transformation. Reproduced with permission [199]. Copyright 2018, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

combine the advantages of multiple 3D bioprinting platforms with regional immobilization of bioactive factors to enable hierarchical bone-biomimetic reconstruction and regeneration, and is expected to provide an innovative approach for complex multicellular tissue regeneration. For the more detailed information of bone tissue biofabrication, we refer the readers to some excellent review papers [1665,1667,1669].

6.1.2. Cartilage regeneration

Cartilage is a smooth elastic connective tissue as the structural and functional component of many specific body parts such as ear, nose, trachea, meniscus, osteochondral, articular cartilage, intervertebral disc and costal cartilage. The cartilage matrices are composed of proteoglycans, collagen fibers, glycosaminoglycans and high content water; they are gelatinous, tough and rigid, thereby playing the role of holding and supporting in the body. Cartilage tissue only contains one kind of cells, i.e. chondrocytes, and can be classified into three types including hyaline cartilage (e.g. trachea, articular cartilage), elastic cartilage (e.g. ear, nose) and fibrocartilage (e.g. meniscus, intervertebral disc), which are different in the relative content of collagens and proteoglycans. Cartilage appears as a relatively simple tissue: it has no blood vessels (avascular) or nerves (aneural). Nutrients are supplied through diffusion to chondrocytes. Cartilage tissue has no self-repair capacity and once cartilage defect emergs, it will finally cause the degenerative and osteoarthritic changes [1673]. In spite of the simple

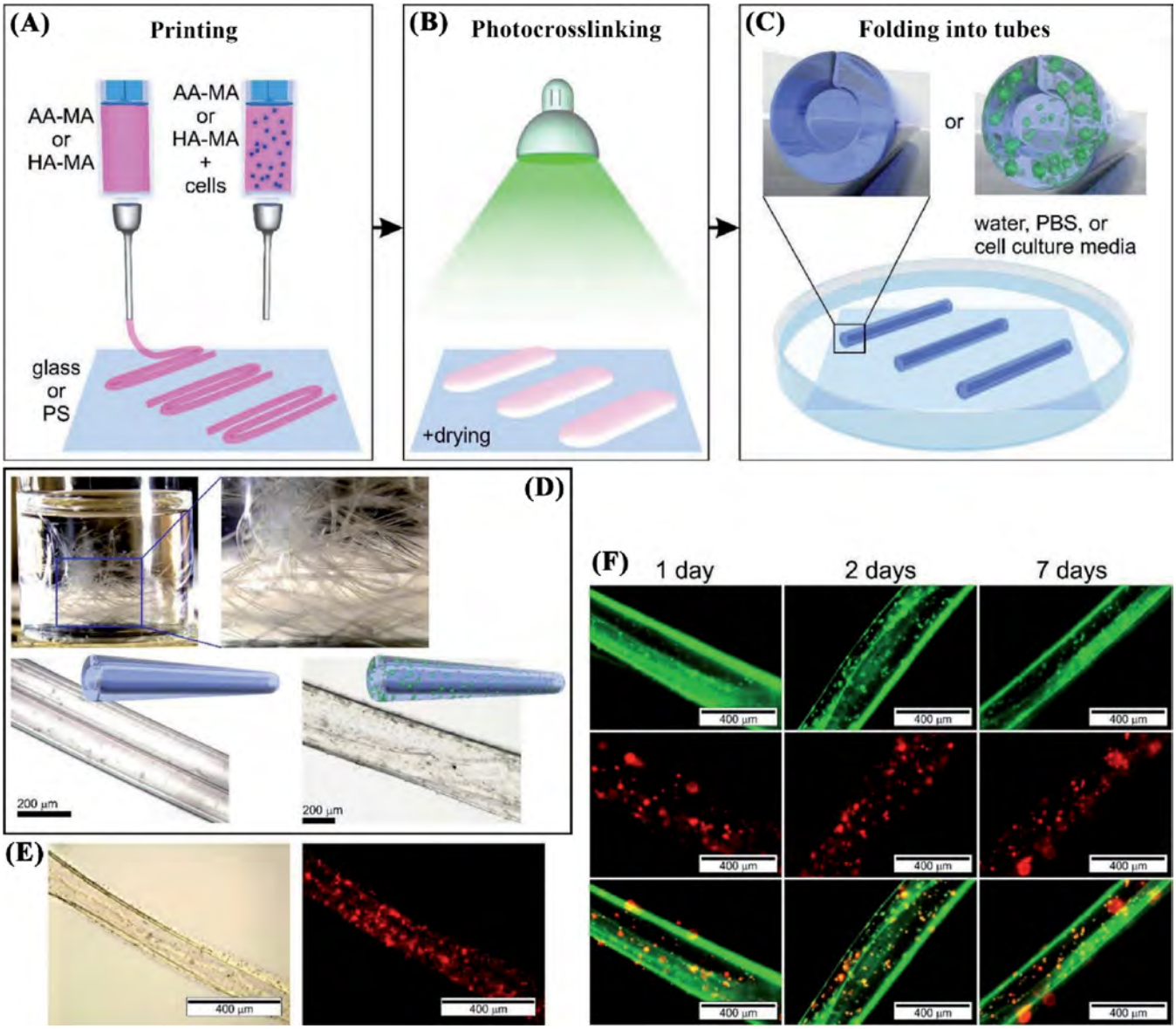

Fig. 24. Hydrogel-based 4D bioprinting of self-folding cell-laden tubes. (A) Printing of AA-MA (alginate methacrylate) or HA-MA (hyaluronan methacrylate) ink materials without/with cells onto glass or PS substrate. (B) Photocrosslinking of the printed films with 530 nm green light. (C) Instant folding of the cross-linked films into tubes when immersing in water, PBS or cell culture medium. (D) Large-scale fabrication of self-folding tubes (top) and images of single tube without/with cells (bottom). (E) Optical (left) and fluorescent (right) images of 4D bioprinted cell-laden self-folding AA-MA tubes. (F) Live/dead fluorescent images of cell-laden AA-MA tubes after different periods of culture: green for live cells, red for dead cells.
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appearance, cartilage tissue actually exhibits distinct heterogeneity with varying composition gradient in the depth. For example, the articular cartilage contains three zones: superficial zone (SZ, top 10 ~ 20 % region contacting with synovial fluid), middle zone (MZ, subsurface 40 ~ 60 % region) and deep zone (DZ, bottom 30 ~ 40 % region contacting with subchondral bone). With the depth increase, chondrocyte density decreases and glycosaminoglycan amount increases (leading to compressive modulus increase). Furthermore, the alignment of collagen fibers differs among the three zones, i.e. parallel to surface in SZ, random orientation in MZ and perpendicular to surface in DZ. These specific orientations combining with the distributed proteoglycan aggregates in between the fibrils can bring cartilage tissue unique biomechanics (resilience, compressive stiffness, shear resistance). Fig. 27 shows the morphology and structure of hyaline cartilage.

Kesti and colleagues [1675] developed a novel cartilage-specific bioink for 3D bioprinting of complex cartilaginous constructs. The bioink was formulated with two FDA-compliant polysaccharides (i.e. alginate and gellan) and combined with the clinical product cartilage ECM particles. The physical gelation of the formulated bioink is cell-friendly in the presence of cations through co-extrusion. As proof-of-concept study, 3D cartilage grafts e.g. ear, nose, and vertebral disk grafts were printed using patient-specific data (Fig. 28A). This 3D bioprinting approach is clinically compliant and can produce viable and high-resolution cartilaginous grafts to meet mechanical and biological requirements. Recently, Bernal et al. [1676] introduced the volumetric bioprinting concept, and demonstrated the 3D bioprinting of large-size living tissue structures using cell-friendly hydrogel bioresins (GelMA) on volumetric printer based on visible light laser. Such volumetric bioprinting can fabricate the entire cell-laden grafts of arbitrary size and structure in seconds to tens of seconds. The 3D meniscus grafts were bioprinted from the anatomical scan (Fig. 28B). The encapsulated articular chondroprogenitor cells revealed increasing metabolic activity

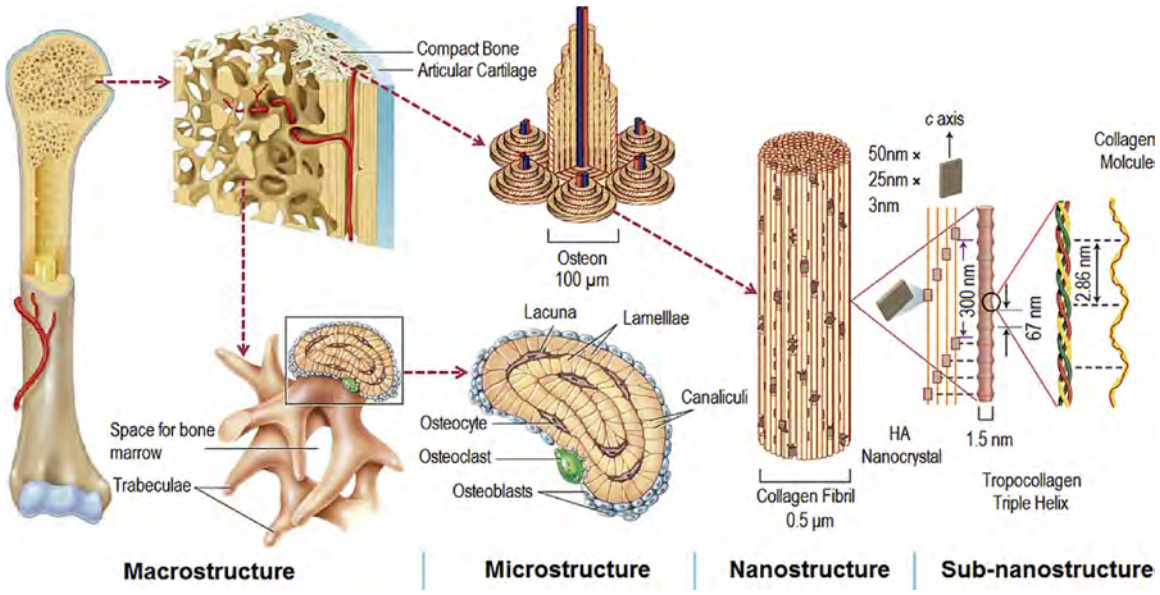

Fig. 25. Hierarchical architectures of native bone.
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and high cellular viability over time within the printed bioconstructs. The meniscus grafts can maintain shape in culture for at least 28 d. During this period, the neo-fbrocartilage matrix was synthesized, with the increase of mechanical properties and GAGs and collagen type I synthesis. This work explored the potential of volumetric bioprinting for rapid biofabrication of patient-specific grafts for a wide range of tissue regeneration applications. For the more detailed information of cartilage tissue biofabrication, we refer the readers to some excellent review papers [202,1673,1677].

6.1.3. Spinal cord regeneration

The spinal cord tissues are not structurally homogeneous, but composed of different cell types in high degree of spatial distribution. Traumatic spinal cord injury (SCI) can cause immediate loss of motor or sensory function below the site of injury, with poor prognosis. Fig. 29A gives an overview of the used experimental strategies to stop SCI progression and promote its repair. In the context of engineering functional spinal cord tissues, it is important to take into consideration the spatial distribution of different cell components to mimic spinal cord structure within the engineered tissue-like bioconstructs. To this point, the 3D bioprinting enables directly printing cells into scaffold for predefined localization, which is very advantageous compared with other fabrication methods involving printing scaffolds first and then seeding cells onto them after printing. Joung et al. [1650] bioengineered spinal cord tissues through extrusion-based multimaterial 3D bioprinting strategy (Fig. 29B). During the printing process, clusters of iPSC-derived spinal neuronal progenitor cells (sNPCs) and oligodendrocyte progenitor cells (OPCs) can be positioned precisely in defined locations within the 3D printed scaffolds. The successful 3D bioprinting of OPCs and sNPCs demonstrated a multicellular neural tissue engineering strategy, which is promising for the regeneration of functional axonal connections across the damage regions of central nervous system (CNS) tissues. It is expected that this 3D bioprinting approach enables the fabrication of novel hydrogel-based bioscaffolds to mimic the complex structures of CNS tissues for treating some neurological diseases, for example, repairing/regenerating the SCI.

Tuszynski and colleagues [1679] recently reported using microscale continuous projection printing (μCPP) to fabricate complex CNS constructs for tissue regeneration applications in SCI repair (Fig. 30). The μCPP approach can print biomimetic 3D hydrogel scaffolds; the scaffold parameters can be tailored to meet the needs of the human spinal cord dimensions and the actual lesion geometries. The injured axons are able to regenerate into the biomimetic 3D printed scaffolds and the implanted NPCs can extend axons out of scaffold and into spinal cord below injury to significantly repair synaptic transmission and functions. Therefore, the biomimetic 3D scaffold fabrication by μCPP may offer an effective way for improving SCI regeneration.

6.1.4. Skeletal muscle regeneration

The skeletal muscle tissues are highly organised cable-like architectures and meanwhile have hierarchical structure units at different sizes (Fig. 31A). The tiniest functional unit is referred to as myofibril. Bundles of myofibrils can generate myofibers that further group to produce fascicles. Then the fascicles can be bundled to create muscle belly. Skeletal muscle tissue engineering aims to repair/regenerate the skeletal muscle defects. The utilization of 3D bioprinting is expected to provide a promising strategy and tool for skeletal muscle tissue regeneration [1680,1681]. Merceron et al. [1682] adopted the 3D integrated organ printing (IOP) system to print four different ink components, which enabled fabricating single integrated muscle–tendon unit (Fig. 31B). The printed bioconstructs revealed over 80 % cell viability at day 1 and 7 post-printing and initial tissue differentiation, thus demonstrating the potential of IOP system to manufacture integrated tissue-like constructs having region difference in cellular types and mechanical property for skeletal muscle tissue engineering. The same group later developed the ITOP system for the 3D bioprinting of skeletal muscle grafts (Fig. 31**C**) [1655]. The 3D printed bioconstructs were able to mature into functional muscle in vivo and the implanted muscle grafts could respond to electrical stimulation to a degree accordant with the developing immature muscles. Costantini and colleagues [1683] demonstrated a novel 3D bioprinting strategy to create skeletal muscle tissues of functional morphologies. The microfluidic printing head was coupled with co-axial needle extruder, thus enabling the 3D bioprinting of myoblast-laden PEG-fibrinogen hydrogel fibers at high resolution and the fabrication of 3D multicellular constructs with precise location of encapsulated cells (Fig. 31D). Histological analysis showed muscle-like tissue formation within the 3D bioprinted implants with significant parallel organization and orientation. This strategy is robust and promising for the 3D bioprinting of skeletal muscle tissues for regenerative applications.

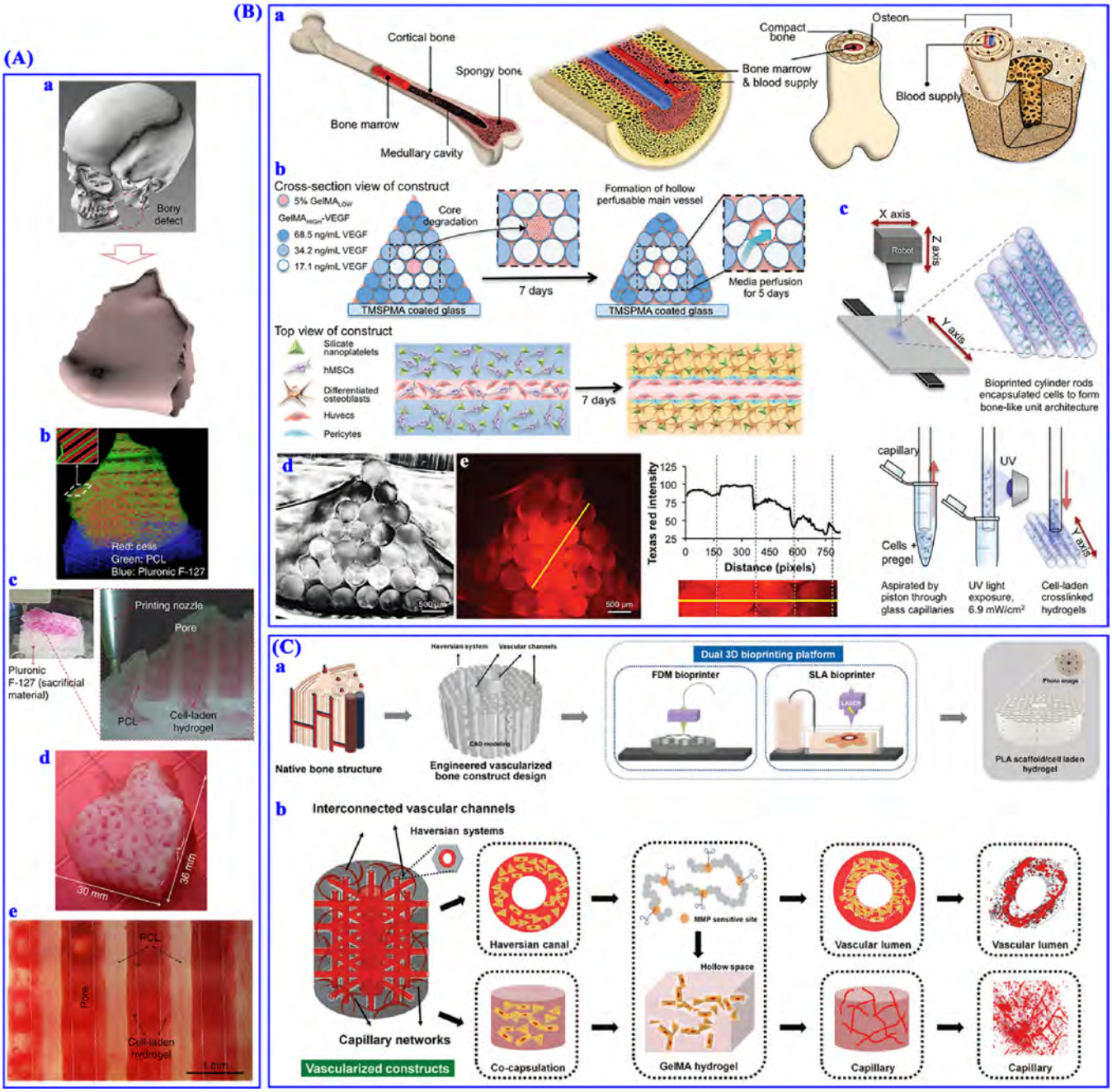

Fig. 26. 3D bioprinting for bone regeneration. (A) Reconstruction of mandible bone defect. (a) 3D CAD model from human CT image data. (b) Visualized motion program to establish 3D defect architecture. (c) 3D bioprinting process with the ITOP system. (d) Picture of the 3D bioprinted defect construct. (e) Calcium deposition showing osteogenic differentiation in the printed bioconstruct. Reproduced with permission [1655]. Copyright 2016, Nature Publishing Group. (B) Biofabrication of osteogenic and vasculogenic 3D bone architectures. (a) Schematic of native bony structure. (b) Schematic of the bone bioprinting strategy. (c) Schematic of 3D bioprinting individual cell-laden cylinders. (d) Cross-section of the bioprinted pyramidal architectures. (e) Conjugation of gradient Texas Red to −COOH modified GelMA. Reproduced with permission [1671]. Copyright 2017, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (C) Biofabrication of vascularised bony biphasic constructs. (a) Schematic of dual 3D bioprinting to engineer the vascularised bony constructs. (b) Design schematic of the vascularised bone constructs. Reproduced with permission [1672]. Copyright 2016, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

6.1.5. Skin regeneration

The human skin (average thickness ~2.5 mm) has unique anatomy composed of three different layers i.e. epidermis, dermis and hypodermis (Fig. 32A). Different types of skin cells are positioned at high degree of region specificity including keratinocytes and melanocytes in epidermis, fibroblasts in dermis and adipose tissue at hypodermis base; meanwhile, collagen can be found in all the three layers. Tissue-engineered skin substitutes are believed to surmount the limitations of conventional skin therapy methods, and 3D bioprinting is a promising strategy to enable rapid, reliable and scalable production of biomimetic skin substitutes to meet the clinical requirements for human skin regeneration [1685]. Koch et al. [1686] demonstrated the 3D spatial arrangement of fibroblasts and keratinocytes in collagen hydrogel by laser cell printing to fabricate multicellular 3D skin grafts (Fig. 32B). This study shows the potential of laser cell printing for engineering functional skin substitutes. Recently, Kim and colleagues [1687] suggested a novel 3D cell printing platform to increase the complexity of the printed skin anatomy (Fig. 32C), which can engineer the perfusable

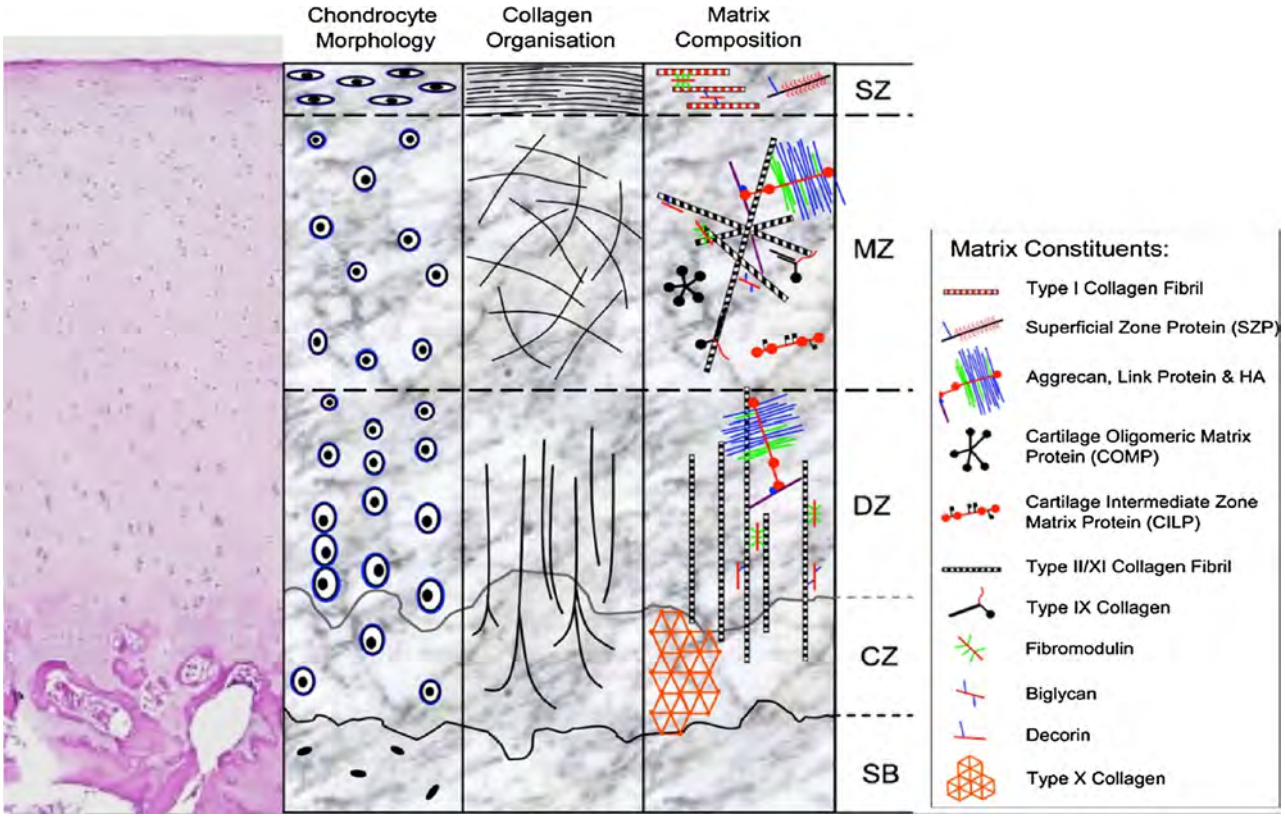

Fig. 27. H&E staining image and schematic illustration of the morphology and structure of hyaline cartilage. Note: CZ, calcified zone; SB, subchondral bone. Reproduced with permission [1674]. Copyright 2015, The Authors. This is an open-access article distributed under the terms of the Creative Commons Attribution License (CC BY).

vascularized 3D human skin equivalent. The full-thickness skin model composed of epidermis, dermis and hypodermis demonstrated tissue maturation and better recapitulation of native human skin. For the more detailed information of skin tissue biofabrication, we refer the readers to some excellent review papers [1688–1690].

6.1.6. Vasculature regeneration

Almost all the tissues in human body own complicated branched internal vascular system to satisfy oxygen and nutrient supply. To promote the performances and functions of engineered tissues, developing 3D vascular networks interconnected within the tissue-engineered constructs can play a crucial role [1652,1691]. However, mimicking the natural vascular networks for blood transport, mass exchange and long-term tissue survival remains a major challenge for tissue engineering [1691,1692]. To this point, 3D bioprinting has been emerging as a promising approach for manufacturing functional 3D vascular networks via predefined deposition of cell and material components [1646,1693–1695]. Lei et al. [1696] presented a versatile 3D printing strategy to create biomimetic vasculature for tissue regeneration applications (Fig. 33A). The vasculature fabrication was based on 3D printed caramel construct as sacrificial template and polymer coating with phase separation. The patterned microchannel networks have biomimetic three-level vascular architectures i.e. 3D framework, interconnected microchannels and permeable walls. This 3D fabrication method is applicable to varied polymers and integrated with different matrices e.g. hydrogels. Mirabella and colleagues [1697] demonstrated the implantation of 3D printed vascular grafts with endothelial cell lined lumens are able to engineer functional vascular patches for guiding rapid therapeutic angiogenesis and perfusion for treatment of the ischaemic diseases (Fig. 33B). Previously, Miller et al. [1693] demonstrated these 3D printed rigid sacrificial template are cytocompatible and able to form patterned vascular networks in various hydrogels for creating 3D perfusable engineered tissues with living cells (Fig. 33C). The perfused vascular channels can maintain metabolic functions of rat primary hepatocytes in tissue-engineered bioconstructs. A coaxial 3D bioprinting approach was developed to create the GelMA microfibers encapsulated in alginate and produce mini tissues containing human umbilical vein endothelial cells; these cells can gradually migrate and connect to generate lumen-like blood vessels [1698]. For the more detailed information of vascular tissue biofabrication, we refer the readers to some excellent review papers [1699,1700].

6.1.7. Ovary regeneration

Patients that undergo treatments of cancers and autoimmune diseases may suffer from weakened ovary function and cause puberty loss, infertility and early menopause [1701,1702]. Current artificial reproduction and hormone restoration approaches e.g. ovary transplant and assisted fertilisation cannot give long-term solution and may leave metastatic diseases to paediatric patients [1703,1704]. Thus, developing whole organ replacement strategy is crucial for oncofertility field in order to restore the long-term fertility and hormone functions for all cancer patients. In this context, tissue engineered hydrogel scaffolds e.g. alginate, fibrin, matrigel, PEG based [1705–1708] play an increasingly important role for ovarian tissue regeneration and function restoration. Due to its unique advantages, 3D printing technology has huge potential to fabricate human ovary bioprostheses for ovarian tissue replacement. As the first attempt towards this aim, Laronda et al. [1709] used 3D printed porous hydrogel scaffolds to investigate the influence of varied scaffold pore geometry on the survival and function of ovarian follicles (Fig. 34). They demonstrated that the 30° and 60° scaffolds can produce the corners surrounding follicles on multiple sides and promote the interactions of follicles with scaffolds. After implantation in sterilized mice, the follicle-seeded 3D scaffolds became highly vascularized and their ovarian function was completely restored.

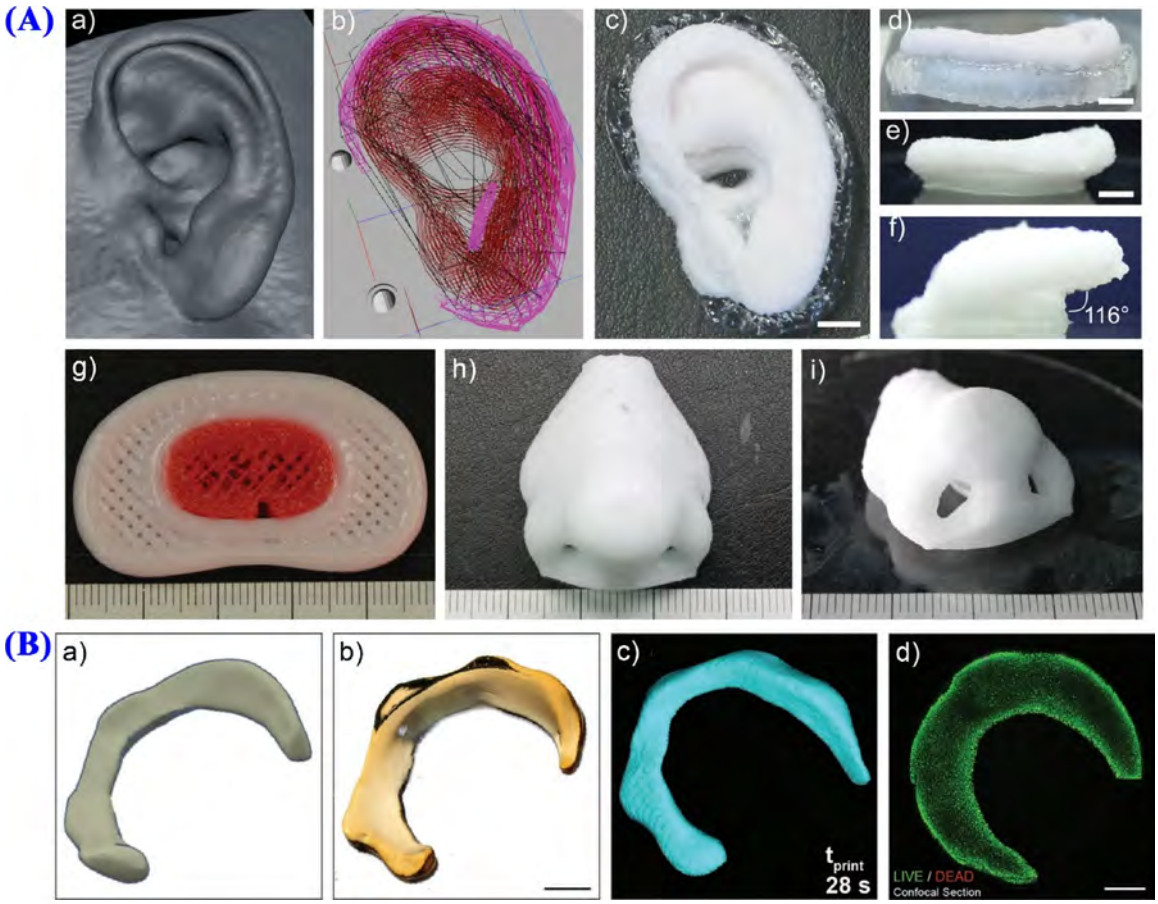

Fig. 28. 3D bioprinting for cartilage regeneration. (A) 3D bioprinting of cartilaginous tissues of complex shapes with clinically used materials as the bioink. 3D printing of external ear graft (a–f), intervertebral disk (g), and nose (h–i) based on patient-specific data. Note: scale bar 5 mm, ruler smallest division 1 mm. Reproduced with permission [1675]. Copyright 2015, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (B) Volumetric bioprinting of meniscus graft. (a) 3D model. (b) The GelMA graft after 28 d of culture in vitro. (c) The μ-CT image of the 3D printed meniscus. (d) The high cellular viability within the graft over 7 d. Note: scale bar 2 mm. Reproduced with permission [1676]. Copyright 2019, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

More excitingly, mouse pups were successfully born via natural mating and thrived by maternal lactation. This study shows that 3D printing can be used for creating bioprosthetic ovarian implants with predefined scaffold designs to achieve whole ovary replacement and restore ovary functions.

6.1.8. Islet transplantation

Type 1 diabetes (T1D), also termed juvenile-onset diabetes, is an autoimmune disease where immune system will attack and damage the insulin-producing islet cells in pancreas. Over the last decades, pancreatic islet transplantation has become a promising therapy for the Type 1 diabetes to give sustained extrinsic insulin supply [1710,1711]. However, after transplantation, factors like the lack of vasculature and allo- or autoimmune attack will cause the substantial loss of islet mass and function. To tackle these problems, encapsulating islets in hydrogel matrix can protect them from immune system and 3D printing can be used for favorable hydrogel scaffold designs, therefore, the combination of islet encapsulation and 3D printing of hydrogel scaffolds is a promising strategy to enhance the islet viability and functions. Liu et al. [1712] formulated the alginate/gelatin bioink for islet and islet-related cell encapsulation and 3D printing (Fig. 35A). The designed coaxial printer allowed for 3D printing of the multicellular islet-containing bioconstructs with a precise control of distribution. After 3D printing, the viability of pancreatic islets was well maintained, showing the potential for islet transplantation. Marchioli and colleagues [1713] adopted the 3D bioplotting to fabricate 3D alginate-based porous hydrogel scaffolds as the islet delivery system (Fig. 35B). They demonstrated that the INS1E β-cells, mouse and human islets can be successfully encapsulated into the 3D plotted hydrogel constructs without influence on their viability and morphology. These islets can be confined at one site in the 3D plotted hydrogel scaffolds and blood vessels can grow into the scaffold pores and come in closer contact with the encapsulated islet tissue. In a recent work, Duin et al. [1714] 3D bioprinted the pancreatic islets in alginate/methylcellulose hydrogel constructs. With the maintained viability, morphology and function within scaffolds, these embedded islets can continuously generate insulin and glucagon during observation.

6.1.9. Heart regeneration

Cardiovascular diseases are the leading reason of mortality and morbidity in the world [1715]. Cardiovascular tissues e.g. heart valves, arteries and myocardium are highly differentiated and incessantly load-bearing. So far, heart transplantation is the only therapy choice for patients with heart failure at the end stage. However, given the fact that the number of available cardiac donors is quite limited, it is of huge demand to develop novel strategies to regenerate/repair infarcted hearts, thereby reducing the excessive dependence on cardiac donors. To solve this issue, cardiac tissue engineering plays an important role through the integration of cardiac cells within 3D hydrogel scaffolds/biomaterials [1716,1717]. Increasing attentions have been paid to 3D bioprinting, a very promising method to create customized cardiac valves, patches or even whole heart [1649,1700,1718]. Recently, Feinberg and colleagues [1719] adopted their home-made FRESH 3D bioprinting technique to fabricate collagen hydrogel scaffolds for

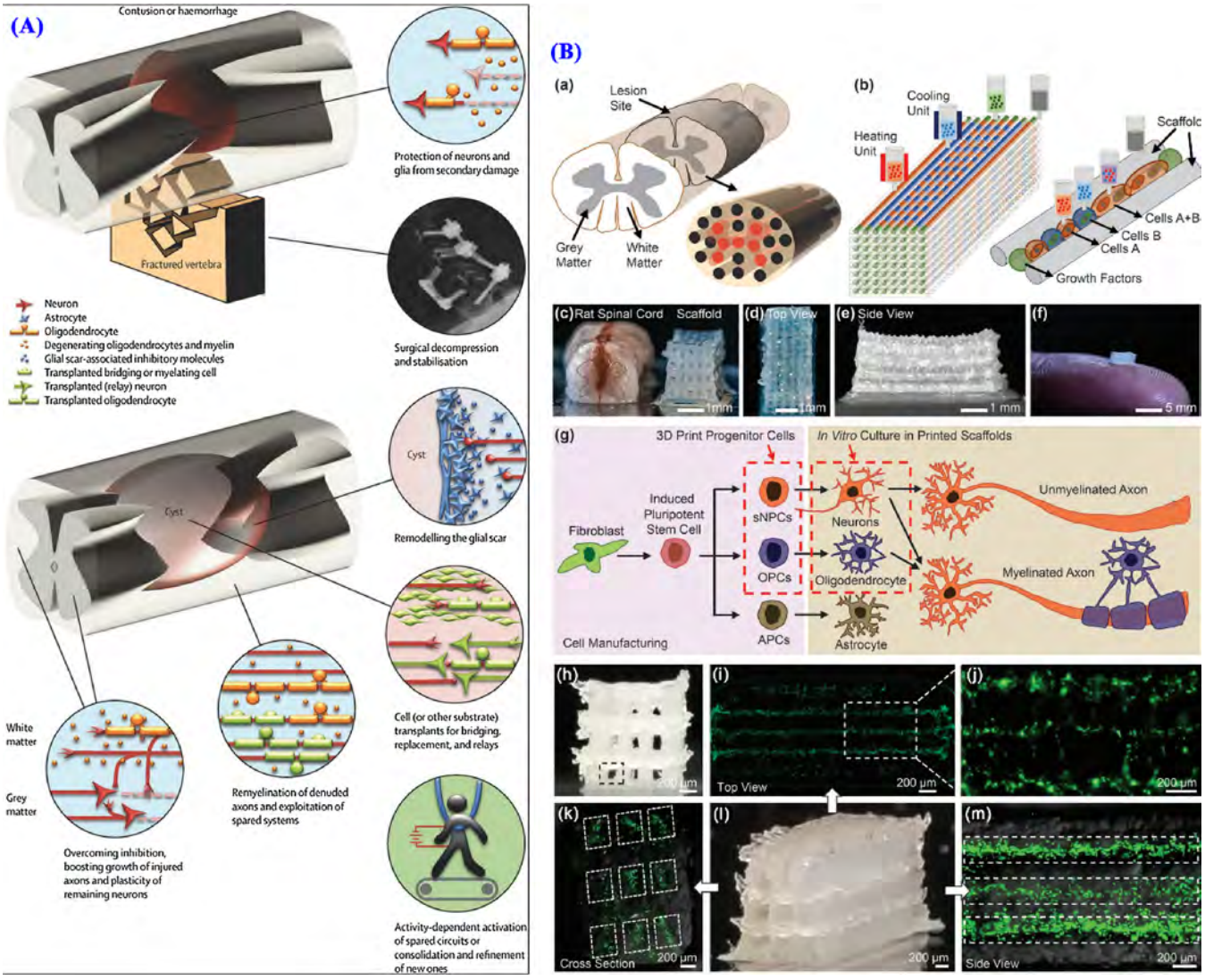

Fig. 29. (A) Experimental strategies to stop progression and promote repair of the spinal cord injury (SCI). Reproduced with permission [1678]. Copyright 2014, Elsevier Ltd. (B) 3D bioprinting for spinal cord regeneration. (a) Schematic spinal cord and model design for 3D bioprinting multichannel scaffold. (b) Schematic 3D bioprinting process through extruding bioinks. (c) Comparison of transected rat spinal cord and the 3D bioprinted scaffold. (d–f) Images of the 3D bioprinted scaffold. (g) Schematic of the progenitor cells for 3D bioprinting and in vitro culture. (h–m) 3D bioprinting of neurocompatible alginate-based bioscaffolds. Reproduced with permission [1650]. Copyright 2018, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

rebuilding the components of human heart at multiple scales, ranging from the capillaries to the whole heart (Fig. 36). They demonstrated that the FRESH 3D bioprinted hearts are able to precisely reproduce the patient-specific anatomy architecture. Morever, using human cardiomyocytes, the bioprinted cardiac ventricles can exhibit synchronic contraction, directional propagation of action potential and wall thickening of ~14 % during the peak systole. Although there is still a long way to achieve 3D bioprinting of totally functional organs, this work shows the potential of 3D bioprinting to rebuild tissue engineered bioconstructs that can begin to recapitulate the complex architectures, mechanics and biological properties of natural tissues/organs.

6.2. *In vitro tissue modeling*

6.2.1. *Tissue models*

Tissue engineering and regenerative medicine has been witnessing the unprecedented success and driving the thrilling field of in vitro tissue models toward the structural, mechanical, biological and functional recapitulation of native human tissues, which is very important for a wide range of applications such as improving tissue/organ regeneration (see section 6.1), investigating natural biological processes, evaluating the safety and efficacy of drugs, antibiotics and cosmetics. From tissue models (hard tissues e.g. bone, cartilage, soft tissues e.g. skin, cardiac tissue, adipose tissue) to organ models (e.g. eye, ear, liver, lung, heart, kidney), from vascularized models to tissue-on-a-chip or organ-on-a-chip platforms, these versatile tissue/organ models hold great promise for in vitro disease modeling and drug development and high-throughput screening. 3D bioprinting has unique technical advantages and enables the precise customization of spatio-temporal placement of cells, ECM, biomaterials and growth factors within the 3D bioengineered constructs/scaffolds, thereby creating the biomimetic tissue and organ analogues for different biomedical applications. For the more detailed information of in vitro tissue model biofabrication, we refer the readers to some excellent review papers [15,1720–1723].

6.2.2. *Integration with microfluidics*

The tissue-on-a-chip and organ-on-a-chip platforms are developed to mimic the complex real microenvironment of native tissues and organs, which is achieved by the precise control over cell components,

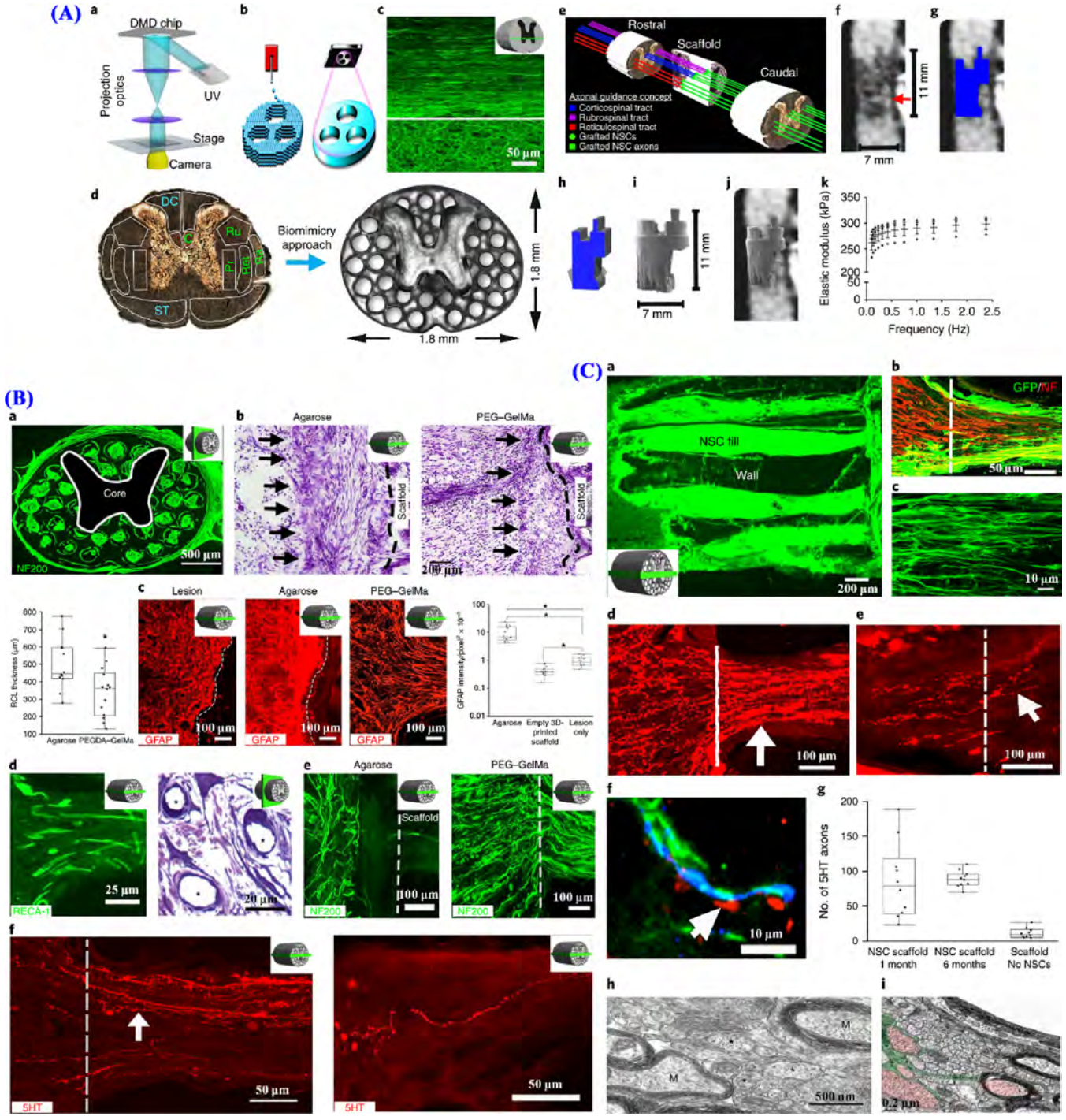

Fig. 30. (A) 3D printing of functional scaffolds to mimic the architecture of spinal cord tissues. (a) 3D printer setup. (b) Comparison of μCPP layerless 3D printing and extrusion 3D printing. (c) Axon neurofilament labeling in intact rat spinal cord. (d) Axonal projections in spinal cord. (e) Schematic of axonal alignment and guidance hypothesis. (f–g) Magnetic resonance image of human clinically complete SCI and the traced outline of cystic lesion cavity. (h–j) 3D model design and 3D printing of the scaffold to fit contusion cavity. (k) Scaffold mechanical measurement. (B) In vivo performance of 3D printed scaffold after 4 weeks. (a) Axon labeling in cross-section. (b) Nissl staining of reactive cell layer (arrows). (c) Glial scar shown by GFAP immunoreactivity. (d) Scaffold vascularization. (e) Comparison of axons to enter scaffolds. (f) Axons (arrow) regenerate into empty scaffold (left) and reach channel caudal end (right). (C) In vivo performance of NPC-laden 3D printed scaffold after 4 weeks. (a) Channels filled with GFP-expressing NPCs. (b–c) GFP-expressing NPCs can extend the linear axons within scaffold. (d) Axons enter NPC-filled channel and regenerate linearly (arrow). (e) Axons regenerate into spinal cord distal to lesion (arrow). (f) Axons regenerating into scaffold can produce appositional contacts (arrow) with NPC dendrites. (g) Quantification of axons reaching scaffold caudal end. (h–i) Ultrastructural images of oligodendrocyte, myelinate and ensheath axons within channels.
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ECM and other factors e.g. hydrogels and growth factors to elucidate the pathological and physiological mechanism. The combination of microfluidics and 3D printing technologies in tissue-on-a-chip and organ-on-a-chip enables more powerful designs for creating complicated flow channels/chambers and functional bioconstructs with 3D heterogeneous structure, cell placement and tissue specificity to

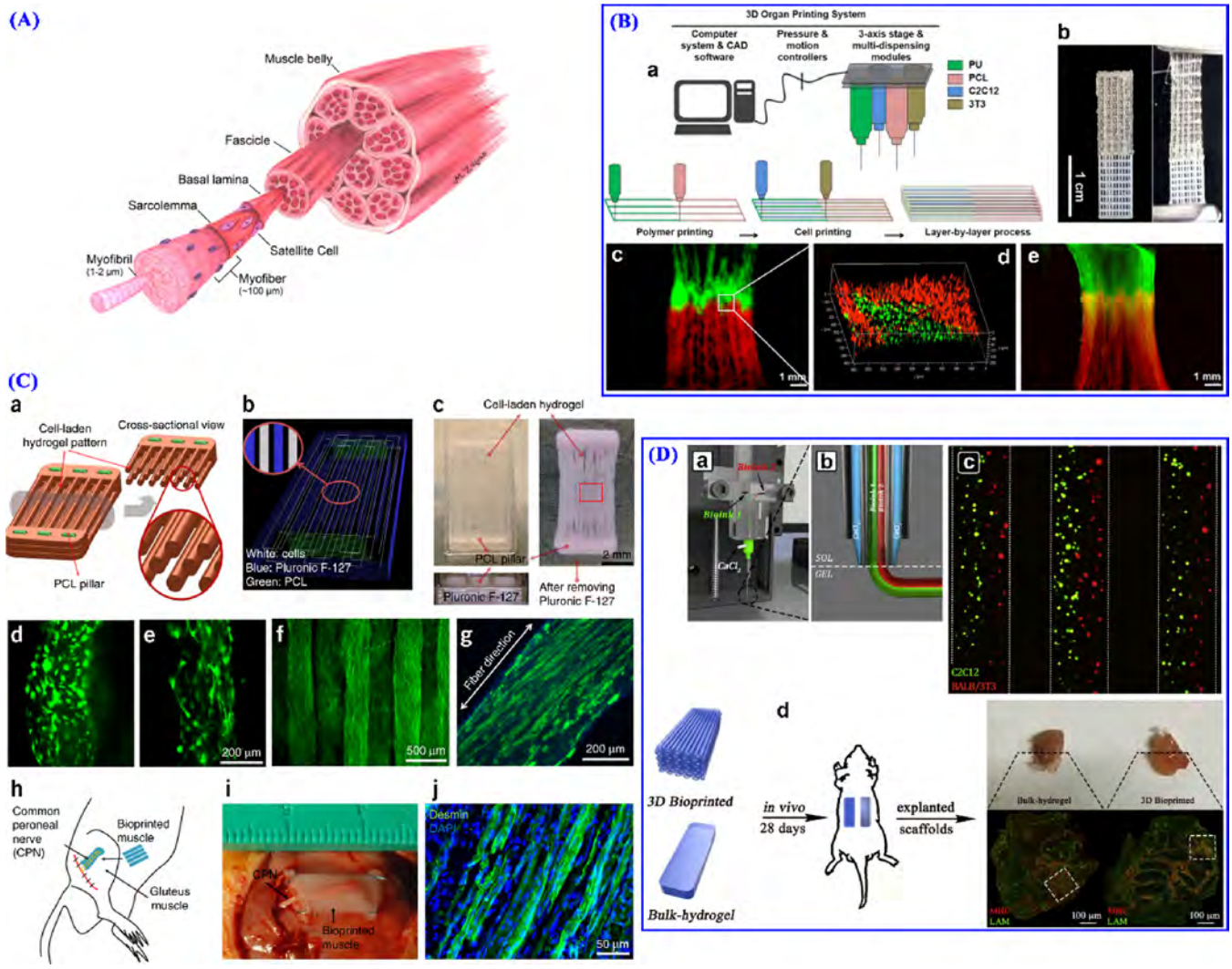

Fig. 31. 3D bioprinting for skeletal muscle regeneration. (A) Schematic of hierarchical arrangement of the skeletal muscle tissues. Reproduced with permission [1684]. Copyright 2015, Elsevier Ltd. (B) 3D bioprinting of muscle–tendon unit. (a) Schematic of 3D IOP system. (b) Image of the 3D printed bioconstruct. (c–e) Fluorescence labeling of dual cell 3D printed construct (green, C2C12; red, NIH/3T3). Reproduced with permission [1682]. Copyright 2015, IOP Publishing Ltd. (C) 3D bioprinting of skeletal muscle. (a–g) In vitro bioprinted muscle. (h–j) In vivo study on structure maintenance and nerve integration of 3D bioprinted muscle graft. Reproduced with permission [1655]. Copyright 2016, Nature Publishing Group. (D) Microfluidic 3D bioprinting of skeletal muscle. (a–b) Schematic 3D bioprinting system. (c) Multicellular 3D bioprinting (green, C2C12; red, BALB/3T3). (d) In vivo study on PEG-fibrinogen hydrogel scaffolds. Reproduced with permission [1683]. Copyright 2017, The Authors. Published by Elsevier Ltd. This is an open access article under the CC. BY-NC-ND.

recapitulate and reproduce the natural tissue/organ features [1724–1726]. On the one hand, 3D printing, without the use of replica molding master, can offer much more simplified fabrication process for microfluidic devices. One of the earliest applications of 3D printing for microfluidics was the fabrication of complicated 3D microvascular networks (Fig. 37A), which were composed of 10 ~ 300 μm smooth cylindrical channels for promoting fluid mixing via chaotic advection [1727]. The multimaterial 3D printing method was used to manufacture the instrumented cardiac microphysiological device with formulated six functional inks, which can integrate the strain gauge sensors in human laminar cardiac tissues to investigate their drug response and contractile development (Fig. 37**B**) [1728]. The soft 3D model of human kidney phantom with collection system was fabricated with 3D printing and medical imaging data (Fig. 37C), with real anatomy architectures and mechanical properties for surgical preparation and simulation [1729]. 3D cell encapsulation and patterning can also be coupled into GelMA hydrogel to improve the properties of 3D printed microfluidic chip, thus forming the tailorable 3D cell culture environment (Fig. 37**D**) [1730].

On the other hand, with elaborate design, microfluidic printheads can be couple with multimaterial 3D printing/bioprinting technique for the on-the-fly seamless switching of inks/bioinks [1683,1731,1732]. Colosi et al. [1733] developed a new 3D bioprinting exemplification which can deposit multimaterials simultaneously and fabricate 3D cell-laden bioconstructs (Fig. 38A). The bioinks, integrated with microfluidic platform, can be formulated to promote cell spreading and migration inside the bioconstructs, and meanwhile allow additional cell seeding on the top of the 3D bioprinted construct, thereby creating new inhomogeneous 3D in vitro tissue models to mimic native tissues for regenerative medicine or drug development purposes. Recently, a promising microfluidic 3D bioprinting technique i.e. multichannel coaxial extrusion system was established to achieve the single-step fabrication of circumferentially multilayered tubular tissues with the bioink formulations of GelMA, alginate and PEGA derivative (Fig. 38**B**) [1734]. This study reveals a basic step toward the fabrication of biomimetic human cannular tissues for tissue regeneration and in vitro tissue modeling. The same group also developed a stereolithography-based multimaterial bioprinting system for the creation of

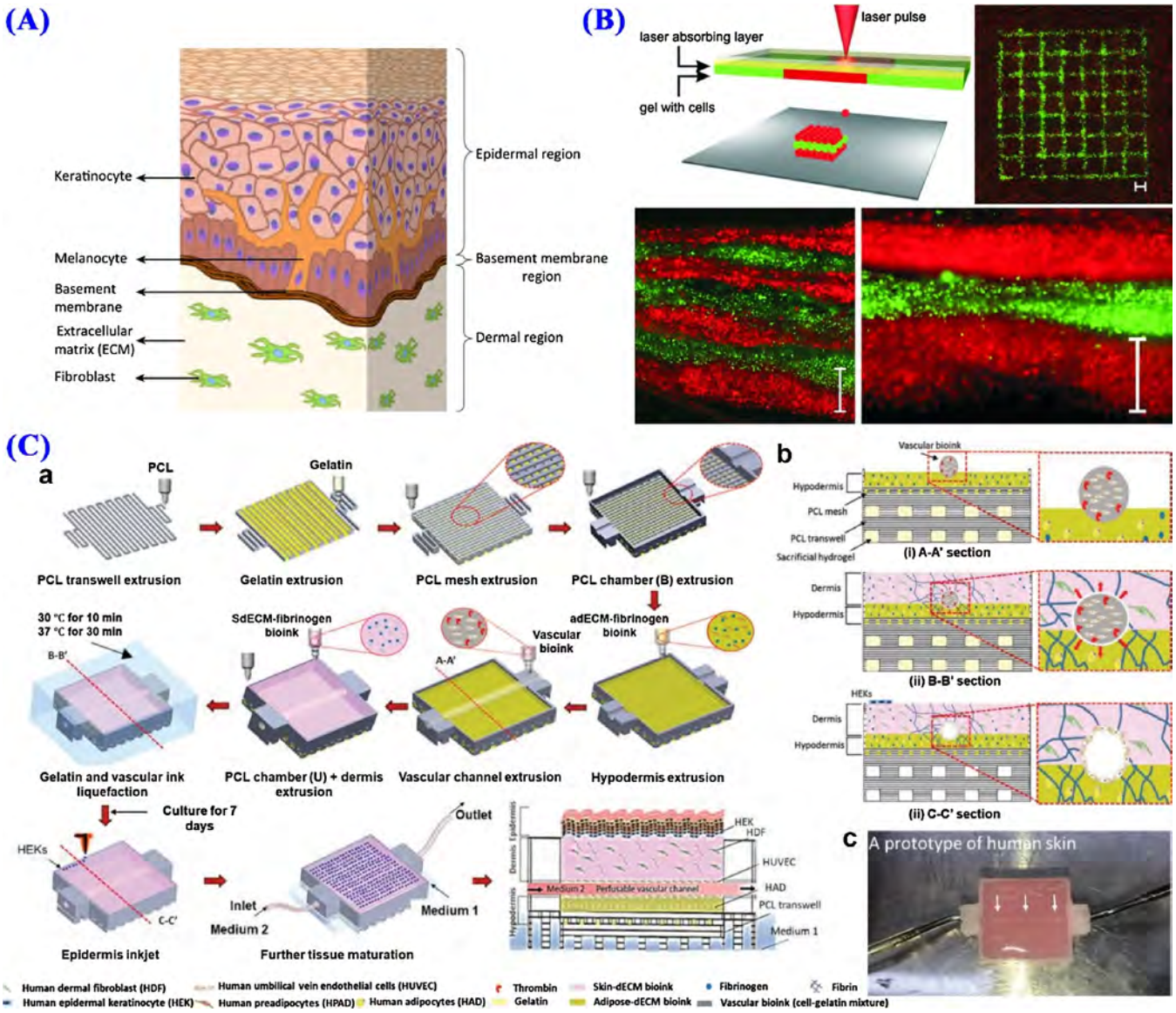

Fig. 32. 3D bioprinting for skin regeneration. (A) Schematic of 3D human skin. Reproduced with permission [1689]. Copyright 2016, Elsevier Ltd. (B) Laser cell printing of skin tissues (Scale bar 500 μm). Reproduced with permission [1686]. Copyright 2012, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (C) 3D cell printing of full-thickness human skin tissues. (a) Schematic of biofabrication process. (b) Section views from (a). (c) A prototype of bioprinted skin graft. Reproduced with permission [1687]. Copyright 2018, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

inhomogeneous 3D hydrogel-based bioconstructs through the combination of microfluidic apparatus having 4 on/off pneumatic valves [1657]. The microfluidic apparatus can switch fast among diverse hydrogel bioinks and thus enable the layer-by-layer multimaterial 3D bioprinting of 3D tissue-like constructs (Fig. 38C). The unique advantage of this bioprinting platform is the multimaterial 3D biofabrication performance at the high spatial resolution, which is superior to the traditional stereolithography bioprinting. 3D human kidney tissue models capable of recapitulating the native human kidney behaviors and responses are very promising for kidney organ regeneration, renal disease modeling and new drug screening. Lewis and colleagues [1735] demonstrated a 3D bioprinting strategy to fabricate the 3D in vitro human renal proximal tubules. These 3D convoluted renal proximal tubules were totally embedded in gelatin/fibrinogen ECM and sheathed in perfusable tissue chip (Fig. 38D), and the 3D tissue-on-a-chip can be maintained for over 2 months. This work represents a new approach to create biomimetic human kidney tissue model for a variety of applications e.g. regenerative medicine, drug mechanism study, drug development and screening and kidney disease modeling.

6.2.3. *Disease modeling*

For decades researchers and clinicians have been profoundly dependent on the use of 2D cell culture experiments and animal models to clarify the underlying cellular and molecular mechanisms that cause various human diseases. However, conventional 2D cell culture and animal models have their limitations and cannot reproduce/recapitulate the complicated tissue microenvironment of natural human tissues. In this context, increasing attempts have been made to develop more complex and physiologically relevant 3D in vitro disease models [1499,1736]. In the field of tissue engineering and regenerative medicine, the research community has been focusing on the development of 3D cell culture and in vitro disease models from the very beginning [1737]. Recently, microphysiological platforms, the biomimetic microfluidic devices, including tissue-on-a-chip and organ-on-a-chip, have been introduced for recapitulation of the biological features and functions of native human tissues, organs and circulation through integrating the in vitro 3D cell culture models into the controlled perfusable microphysiological systems [1738]. The combination of 3D bioprinting and microfluidics can provide more diverse and versatile

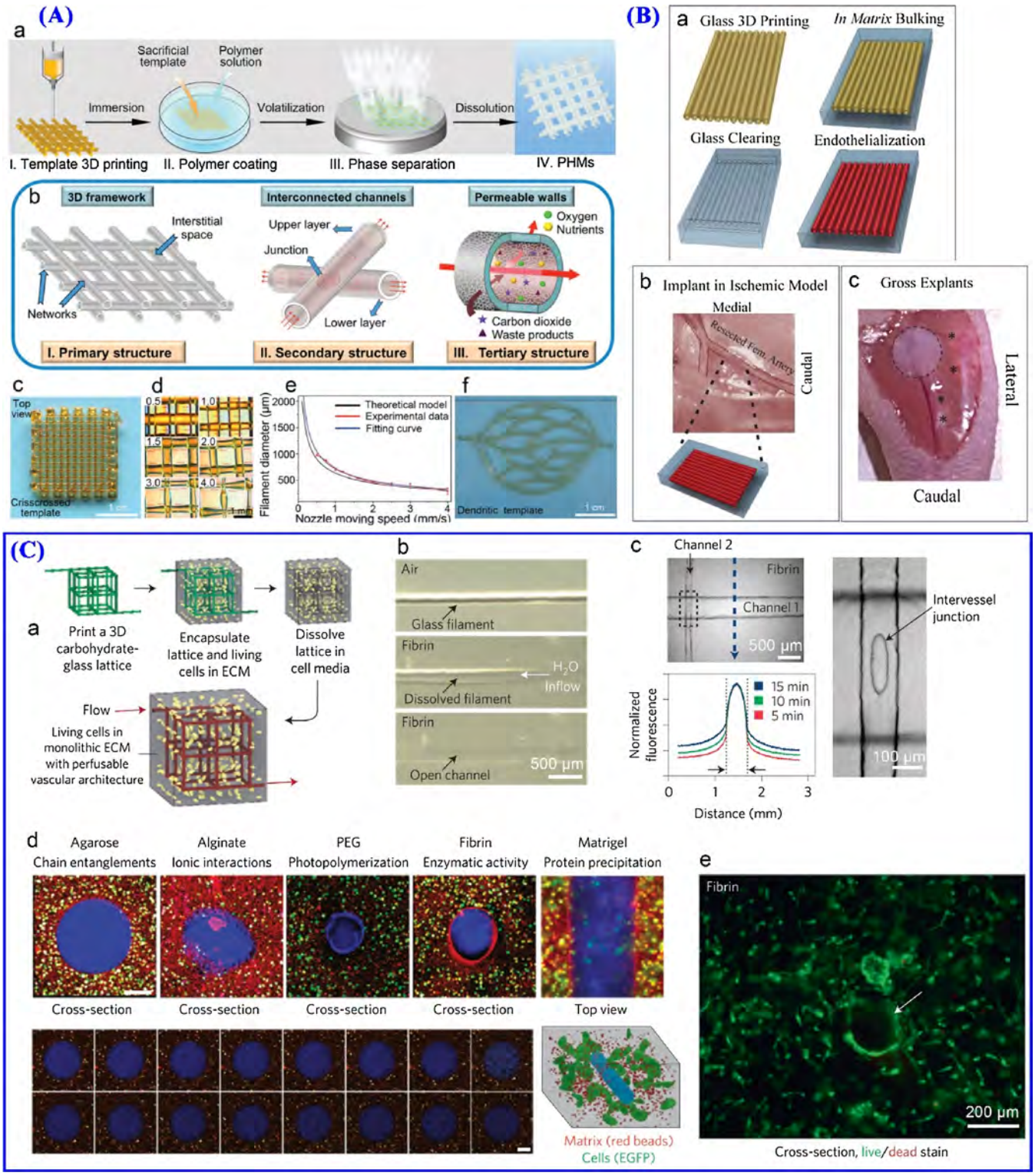

Fig. 33. 3D printing for vascular and vascularized tissue regeneration. (A) Design and printing of biomimetic vasculature. (a–b) Schematic of fabrication process and printed constructs. (c–f) 3D printed caramel template with tunable filament diameter. Reproduced with permission [1696]. Copyright 2019, The Royal Society of Chemistry. (B) 3D printing of vascular patches. (a) Schematic of fabrication. (b–c) Illustration of patches implantation. Reproduced with permission [1697]. Copyright 207, Nature Publishing Group. (C) Engineered tissue constructs with patterned vascular networks and living cells. (a) Schematic overview. (b) Removal of filament in fibrin gel. (c) Transport of fluorescent dextran in fibrin gel channels. (d) Patterned vascular channels in various gels (Scale bar 200 μm). (e) Live/dead staining image of cross-section in fibrin gel. Reproduced with permission [1693]. Copyright 2012, Nature Publishing Group.

designs for creating tissue-on-a-chip and organ-on-a-chip systems for 3D in vitro disease modeling (see section 6.2.2). For the more detailed information of 3D bioprinting for in vitro disease models, we refer the readers to some excellent review papers [1739–1742].

6.2.4. *Drug screening*

The traditional drug discovery road from the preclinical study to the clinical trial and to the final commercial use is a quite long and costly process. Although continuous efforts have been made to increase the

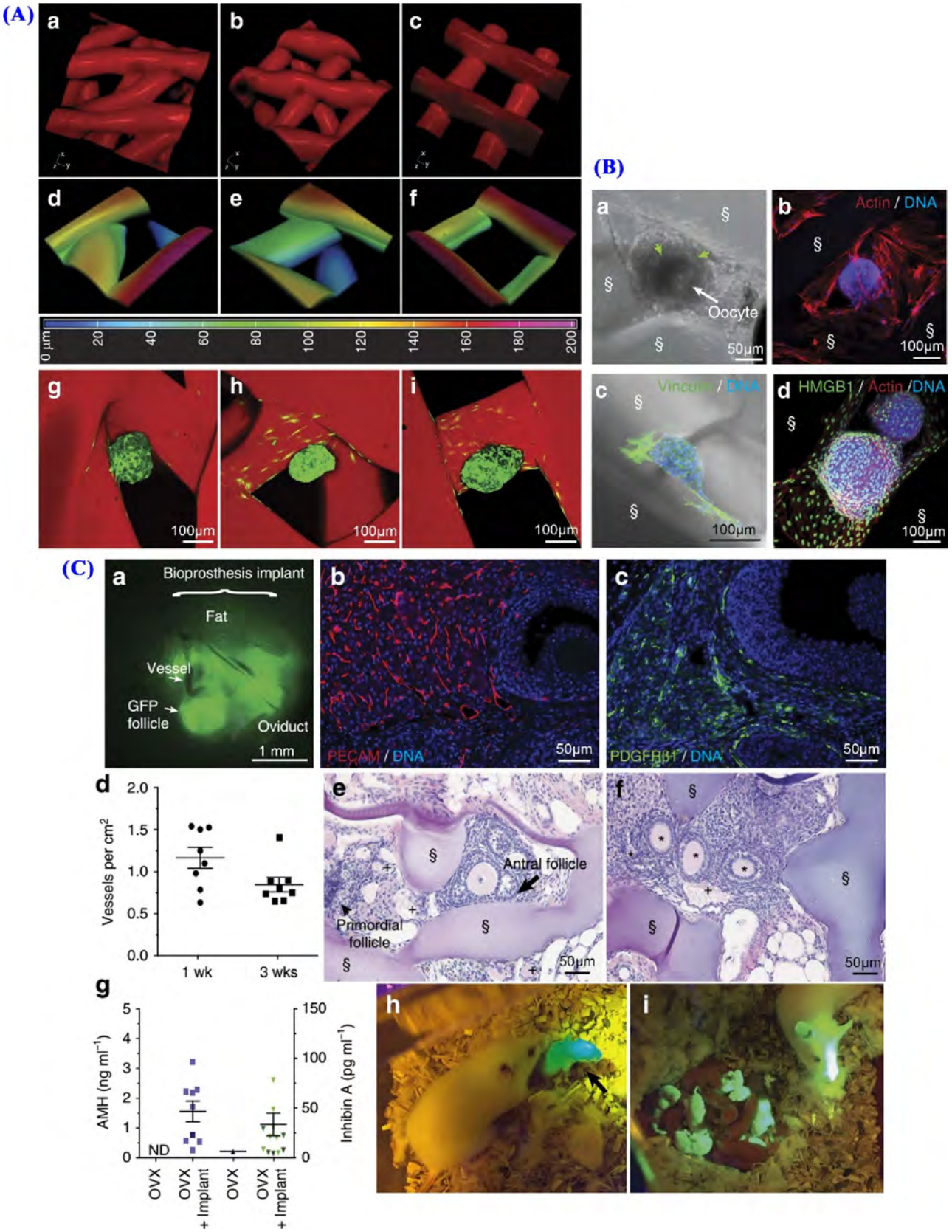

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Fig. 34. (A) Dependence of ovarian follicle survival on follicle–strut contacts from 30° and 60° scaffold pore geometry. (a–c) 3D reconstruction of 30°, 60° and 90° scaffolds. (d–f) 3D pore reconstruction. (g–i) Maximum intensity projection of GFP + follicles in pores after 2 day culture. (B) Interactions of follicles with struts. (a) Image of follicle in 30° scaffold for 8 day culture with maintained basement membrane (green arrow) and centralized oocyte (white arrow). (b) Image of follicle in 60° scaffold for 6 day culture revealing cells can surround and support follicle. (c) Vinculin staining showing cells can adhere to gelatin struts of the 3D printed scaffold after 8 day culture. (d) Image of follicle in 60° scaffold for 2 day culture showing stromal cells surrounding and anchoring follicle. (C) Bioprosthetic ovaries supporting vascular infiltration and restoring function in vivo. (a) Implantation of GFP + follicles (green) in GFP- mice ovarian bursa. (b–c) Vascularization. (d) Quantification of vessels. (e–f) H&E staining of ovarian bioprostheses. (g) Serum analysis. (h–i) Photos of GFP + pup. Reproduced with permission [1709]. Copyright 2017, The Authors. This work is licensed under a Creative Commons Attribution 4.0 International License.

production efficiency of new drug development, only 1 out of tested 10,000 new chemical entities can be finally approved and enter market [1743]. Therefore, it is very necessary and meaningful to develop novel strategies and concepts for drug screening and development, thus being able to speed up the new drug discovery and clinical use. Numerous attempts have been showing that 3D in vitro tissue and organ models are very ideal platforms to mimic the native tissues and organs, and they can be integrated in various microarrays and microfluidic systems, thereby enabling the high-throughput drug trial and screening. Due to the unique technological superiorities, 3D bioprinting has been widely used to create 3D in vitro tissue and organ models with huge potential for the drug toxicology testing and high-throughput drug screening [226]. For the more detailed information of 3D bioprinting for drug screening applications, we refer the readers to some excellent review papers [85,1739,1744,1745].

6.2.5. Cancer research

Nowadays, cancer is still among the most life-threatening diseases in the world. Many factors may affect cancer prognosis; these factors include cancer type and stage, tumor microenvironment, patient age and gender, body response to cancer treatment, etc. To implement basic cancer research and develop more effective cancer therapy, new methodologies are needed prior to clinical trials, for example, establishment of various in vivo cancer-bearing animal models and in vitro cancer models. The cancer models can provide promising cancer research platforms with no need for the animal models, thus being able to largely reduce the use of animals in cancer research; however, it is still a challenge to create fully functional cancer models in imitation/replication of the anatomical structure, biological feature and drug response of human solid tumors [1746,1747]. 3D bioprinting is a powerful way to tackle this issue through creating 3D tissue engineered bioconstructs that are able to mimic/recapitulate the heterogeneity, vascular networks, multicellular spheroids and native ECM of solid tumor tissues. Moreover, 3D bioprinting can also create the tissue engineered bioconstructs into the microfluidic devices, thereby together producing the “tumor-on-a-chip” platforms suitable for in vitro tumor modeling and high-throughput screening of new drugs or therapeutic targets in biomimetic tumor microenvironment [1748]. Hydrogel-based microfluidic chips with multiscale vascular network were constructed through 3D printing to model the interactions of tumors and blood vessels [1749]. Cao et al. [1750] recently developed a tumor-on-a-chip with 3D bioprinted blood and lymphatic vessel pair (TOC-BBL, Fig. 39A). The bioprinted blood vessel was hollow perfusable and the lymphatic vessel was one end blinded. Such devices revealed diverse diffusion profiles for biomolecules or anticancer drugs with the varied combination of 3D bioprinted blood and lymphatic vessels. It is anticipated that such in vitro tumor model may enable mimicking the drug transport within tumor microenvironment and thereby improve the screening accuracy of anticancer drugs. Cho and colleagues [1751] utilized the 3D bioprinted glioblastoma-on-a-chip system to investigate patient-specific body responses toward chemoradiotherapy (Fig. 39B). First, they 3D bioprinted the in vitro glioblastoma model using patient-derived tumor cells and vascular endothelial cells within decellularized ECM hydrogels. The bioprinted tumor model was capable of recapitulating the architectural, biophysical and biochemical features of natural glioblastoma tumor, and reproducing the patient-specific resistance to temozolomide and chemoradiation therapy in clinic. This model can also screen different anticancer drug combinations and thus assist in identifying effective therapies for the glioblastoma patients. 3D bioprinted in vitro microchip in hydrogels can also be used for studying the cancer cell migration behavior [1752]. Meng et al. [1753] demonstrated the use of 3D bioprinting to create in vitro metastatic model through predefined positioning of tumor cells, hydrogels and release capsules (Fig. 39C). On the basis of guided migration of the tumor and endothelial cells in the presence of growth factors and stromal cells, the 3D vascularized in vitro model can simulate the cancer progression and metastasis including invasion, intravasation and vascularization. Meanwhile, this bioprinted metastatic model can serve as a useful platform to identify and screen new anticancer drugs for animal-free preclinical study. Recently, Xu and colleagues [1754] established a 3D microtissue model of growth-hormone-secreting pituitary adenoma (GHSPA) through 3D bioprinting. Compared to those in 2D environment, tumor cells in the 3D model showed the enhanced activity of cell cycle progression, secretion, proliferation, invasion and tumorigenesis. This 3D bioprinting strategy of tumor microtissue can create a promising in vitro model at tissue level for cancer research, and thereby hopefully boost the further investigation of GHSPA etiology, therapy, drug resistance and long term prognosis.

6.3. Surgical preparation

3D printing technology enables developing patient-specific treatment protocols through printing the patient anatomy, thereby promoting the development of individual patient care. The individualized variance and complexity of human body anatomy make the 3D printed models an ideal tool for the surgical preparation. For surgeons or physicians, studying a tangible 3D model of patient anatomy for surgery simulation and planning is preferable and more instructive than solely viewing the MRI or CT scan images on a flat screen [1755]. Being able to fully simulate all complex surgical procedures prior to a surgery with the help of 3D printed models can assist in predicting the possible intraoperative accidents or complications, with already having numerous cases for preoperative planning [97,1756]. A proof-of-concept study adopted 3D printing to develop a useful benchtop workflow to estimate the interactions of customized expandable valve sizer with patient-specific aortic root anatomy, which can help to predict the leak location of heart valve and direct the optimal selection of artificial valve size for every individual patient, thus enabling preoperative planning for successful transcatheter aortic valve replacement [1757]. The 3D printing technology can allow for high-resolution fabrication of rigid and flexible parts of the flexible 3D models of aortic root and transcatheter aortic valve [1758]. In clinic, 3D printed models have been used for preoperative planning of liver transplantation [1759]. The surgeons utilized patient liver replicas to foresee how to carve donor liver with minimal tissue loss to match recipient abdominal cavity. It should be noticed that these 3D liver models were printed with partially transparent acrylic resin or PVA hydrogels. The combination of hydrogel materials and 3D printing can create soft tissue and organ models with needed water content and vasculature networks similar to native living soft tissues and organs; 3D printing of tough hydrogels can even be used to fabricate hard tissues such as cartilages, tendons and ligaments. Using these 3D models for preoperative planning will be able to allow

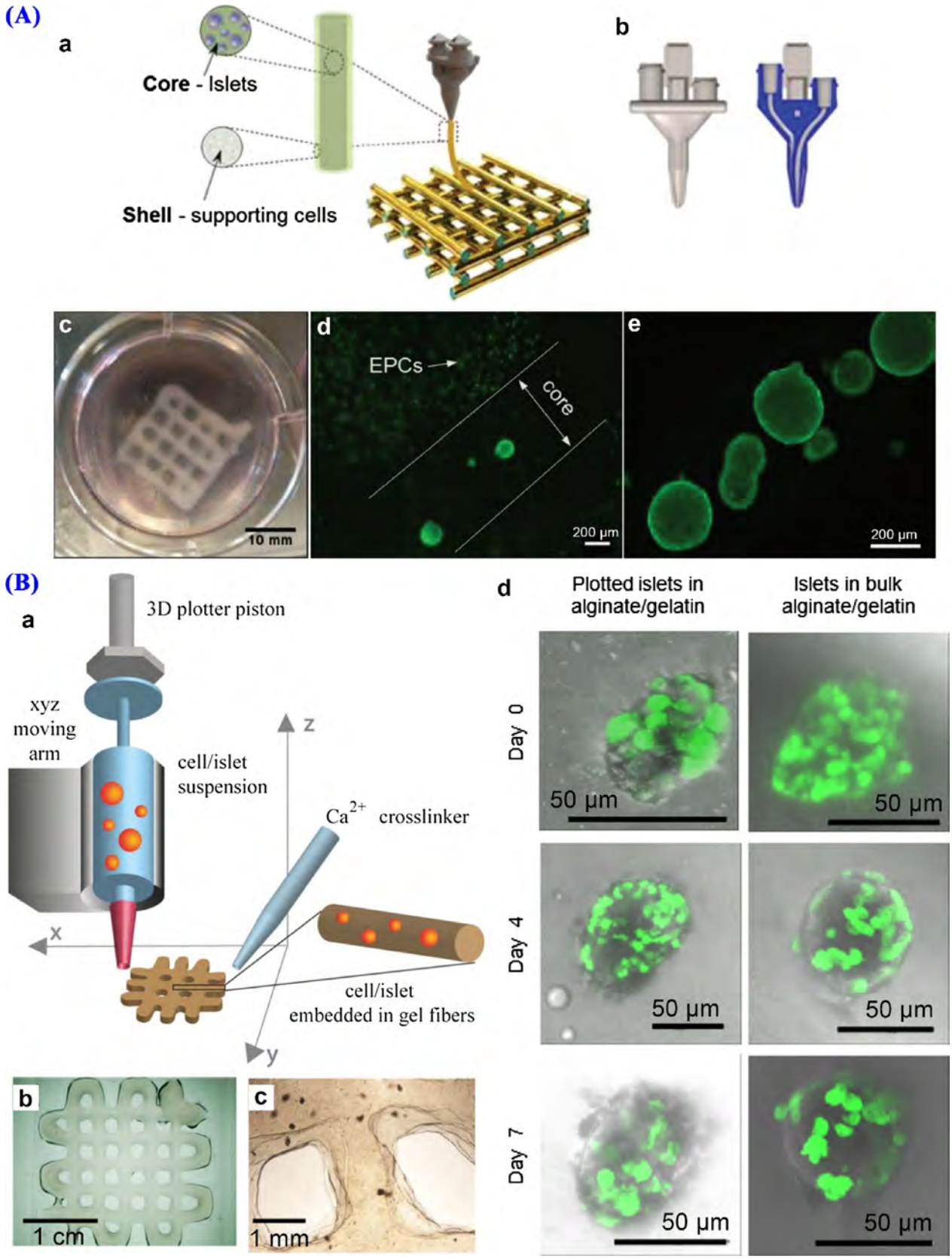

Fig. 35. 3D bioprinting for islet transplantation. (A) Coaxial 3D bioprinting of vascularized pancreatic bioconstructs. (a) Schematic of bioprinting. (b) Nozzle. (c–e) Printed bioconstruct with the encapsulated islets and endothelial progenitor cells. Reproduced with permission [1712]. Copyright 2019, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (B) 3D bioplotting of hydrogel scaffolds for islet delivery. (a) Schematic of bioplotting. (b) 3D plotted alginate/gelatin scaffold. (c) 3D plotted islets in scaffold. (d) Fluorescence image of islets in scaffold. Reproduced with permission [1713]. Copyright 2015, IOP Publishing Ltd.

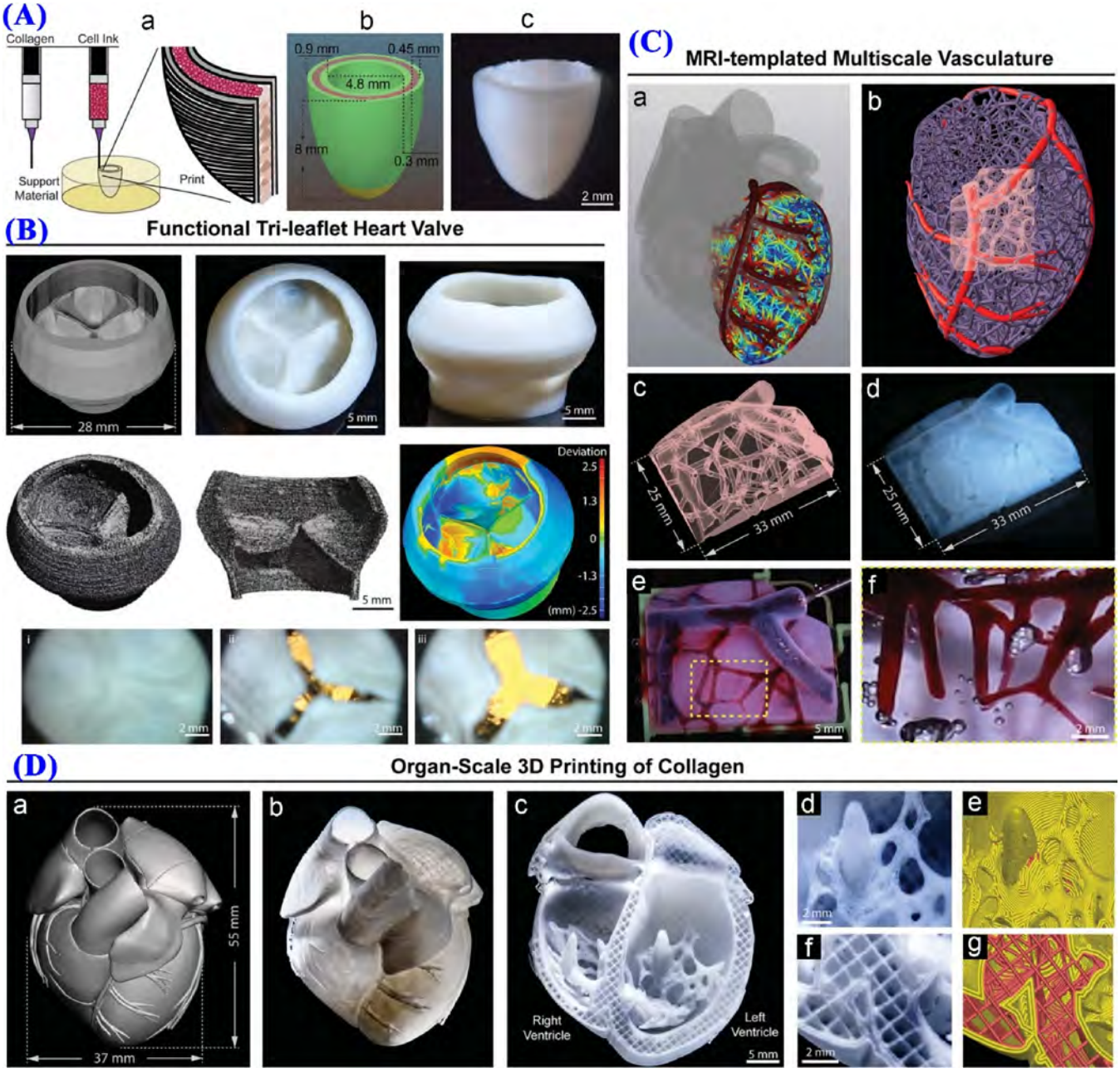

Fig. 36. 3D bioprinting for cardiovascular tissue engineering. (A) FRESH 3D bioprintig of contractile human cardiac ventricle. (a) Schematic of bioprinting. (b) Ventricle model. (c) Printed ventricle. (B) Organ-scale bioprinting of tri-leaflet heart valve. (C) Bioprinting of multiscale vasculature. (a) 3D human heart model. (b) Left ventricle. (c) 3D vascular network. (d) Bioprinted vascular network. (e–f) Glycerol perfusion (red) of vascular network. (D) Bioprinting of neonatal-scale human heart. (a–c) 3D model and printed collagen heart. (d–e) Image of trabeculae in left ventricle. (f–g) Image of septal wall between the ventricles. Reproduced with permission [1719]. Copyright 2019, American Association for the Advancement of Science.

surgeons and physicians to cut into them with surgical blades with a more realistic 3D perception and appreciation relative to the anatomical, mechanical and functional characteristics of natural or pathological tissues and organs.

6.4. Soft robotics

Design and development of mechanically strong and stimuli-responsive hydrogel materials have enabled the rapid advancement of hydrogel-based biomimetic soft robotics [1760–1762]. To develop soft robots and soft actuators through 3D printing has been a thrilling research field. 3D printing can allow for the customization design and one-step production of soft robots/actuators with complicated architecture at a high resolution, thereby facilitating more precise control over their shape morphing ability. The combination of stimuli-responsive hydrogels with 3D printing can create novel 4D shape transforming or actuating systems that can make asymmetrical response e.g. swelling/shrinking to a variety of external stimuli e.g. temperature, pH, humidity, light, electric field or magnetic field (see section 3.6,4.3.2 and 5.3.2). The commonly used hydrogel materials include alginate, agarose, chitosan, methylcellulose, PEG, PNIPAAm, GelMA, pluronic and their derivatives [106], with the unique superiorities of porous network, high water content, shape memory effect and governable sol-gel phase transition relative to other traditional materials. The technical advantages of 3D printing e.g. layer-by-layer deposition and multimaterial deposition can allow feasible anisotropic

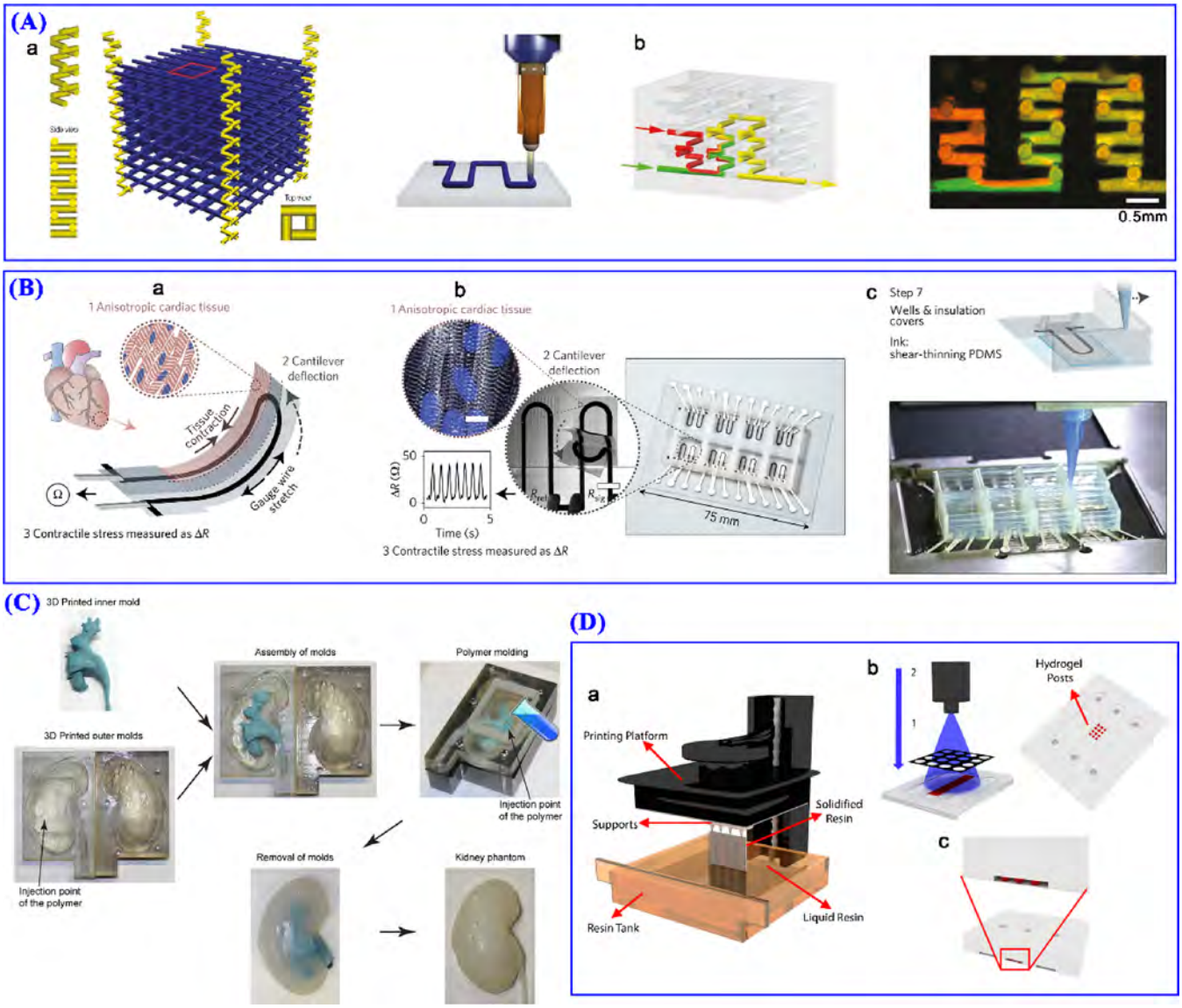

Fig. 37. 3D printing for microfluidics. (A) 3D microvascular networks fabrication. (a) Schematic of fabricating microvascular scaffold. (b) Microfluidic mixing test. Reproduced with permission [1727]. Copyright 2003, Nature Publishing Group. (B) Multimaterial 3D printing of cardiac microphysiological device. (a) Device principle schematic. (b) Fully printed final device. (c) Microscale 3D printing process (step 7 shown here). Reproduced with permission [1728]. Copyright 2017, Nature Publishing Group. (C) 3D printing of human kidney phantom. Reproduced with permission [1729]. Copyright 2016, The Authors. This article is distributed under the terms of the Creative Commons Attribution 4.0 International License (http://creativecommons.org/licenses/by/4.0/), which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited. (D) 3D printing of microfluidic chip. (a) Stereolithography printing process. (b) Hydrogel post formation. (c) 3D chip cross-section. Reproduced with permission [1730]. Copyright 2016, IOP Publishing Ltd.

designs of soft robotics and soft actuators and create predefined anisotropic structures to achieve better control over their actuating and shape-morphing performance. A hydraulic hydrogel actuator and robot can be created by 3D printing [1763]. With prescribed architectures and properties, the designed hydrogels exhibited hydraulic actuations that endow soft robots and actuators with optical and sonical camouflage ability in water at a high speed. An autonomous, fully soft robot (octobot) was designed and fabricated via the multimaterial 3D printing of two hydrogel-based fugitive and catalytic inks into moulded base materials (Fig. 14**A-C**) [1577]. Using the stimuli-responsive nanocomposite hydrogel ink composed of GelMA and aligned iron oxide nanoparticles, a magnetic starfish soft robot was 3D printed, showing magnetic remote control and cell guidance effect in which C2C12 skeletal myoblasts can differentiate into myotubes without inducing medium [1764]. Soft helical GelMA hydrogel microswimmers were microfabricated by 2 P P 3D printing method and then decorated with magnetic nanoparticles (Fig. 40**A**) [1765]. Such hydrogel soft robots can be enzymatically biodegraded after implementing tasks and thus hold promise for targeted cell delivery or drug delivery into human body for wide biomedical applications. Through stereolithography 3D printing, Bashir and colleagues developed hydrogel “bio-bots” of asymmetrical physical design, which were powered by the engineered skeletal muscle strip to move (Fig. 40**B**) [1766]. Electric stimulation was able to trigger the contraction of C2C12 cells within the skeletal muscle strip and the net locomotion of hydrogel bio-bot at maximum speed of more than 1.5 body lengths per min ($\sim 156\,\mu m\ s^{-1}$). Such biorobot designs can integrate cell components within soft robotic systems, thus enabling a wide range of biomedical applications e.g. active drug delivery, drug screening, bioinspired machine designs and programmed tissue engineering. Morimoto et al. [1767] developed a biohybrid robot that was powered with an antagonistic pair of skeletal muscle tissues (Fig. 40C). They demonstrated the biohybrid robot can

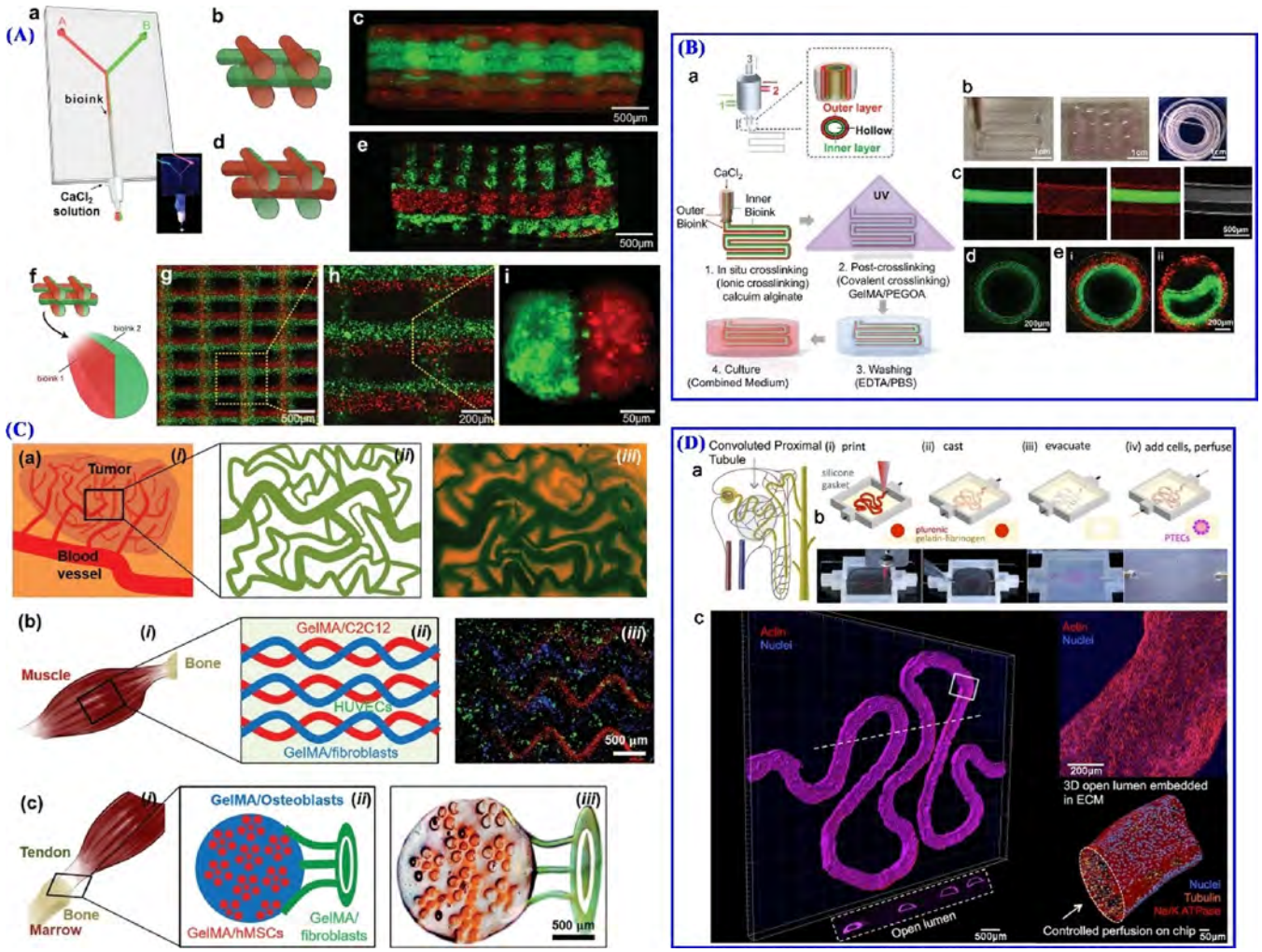

Fig. 38. Microfluidic-enhanced 3D bioprinting. (A) Multimaterial bioprinting of heterogeneous 3D tissue constructs. (a) Microfluidic system. (b–c) Alternate extrusion. (d–e) Alternate/simultaneous extrusion. (f–i) Simultaneous extrusion. Reproduced with permission [1733]. Copyright 2015, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (B) Bioprinting of multilayered cannular tissues. (a) Bioprinting schematic of multilayered hollow tube. (b) Bioprinted perfusable tubes. (c) Image of double layered hollow fibres. (d–e) Image of monolayered (i) and double layered (ii) hollow fibres. Reproduced with permission [1734]. Copyright 2018, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (C) Multimaterial maskless stereolithography bioprinting. (a) Tumor angiogenesis model. (b) Skeletal muscle model. (c) Tendon-to-bone insertion model. Reproduced with permission [1657]. Copyright 2018, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (D) Bioprinting of 3D renal proximal tubules. (a) Nephron schematic. (b) Creation of 3D perfusable tubules. (c) Bioprinted convoluted tubule with 3D rendering. Reproduced with permission [1735]. Copyright 2016, The Authors. This work is licensed under a Creative Commons Attribution 4.0 International License.

allow for the pick-and-place object manipulation, thereby having the potential to replicate/reproduce diverse life-like motions. In addition, the biohybrid robot can be integrated with muscle-on-a-chip platform to develop the in vitro tissue models for new drug screening and discovery. For the more detailed information of 3D printing for soft robotics, we refer the readers to some excellent review papers [106,107,1768].

6.5. Printable electronics

The wearable flexible electronics represent one highly important field of future technical development, which include far more than the smartwatch but the whole range of flexible wearables e.g. biosensors, programmed biomedical implants/devices, supercapacitors and electrochemical cells powered by human body fluids. These wearable devices hold enormous potential to greatly improve the quality of our life, health and safety. Due to the stretchability, transparency, biocompability, ionic conductivity and similarity to the native biologic tissues, hydrogel materials have been recently considered as a promising candidate for developing the new generation flexible bioelectronic devices [1769–1771]. Compared with traditional fabrication methods, 3D printing technology can allow for the creation of customizable flexible electronic devices with complex structures by precisely predefining to meet the individualized use requirements. The combination of 3D printing with wearable flexible electronics enables the rapid advancement of 3D printable electronics for personalized flexible wearables [1772–1774]. These 3D printable flexible wearables have huge potential for a wide range of customized applications including individual wearable devices, patient-specific prostheses, human–machine interfaces, etc. Using 3D printing strategy, Zhao and colleagues [1593] developed a novel living material system consisting of hydrogels and programed bacteria, wherein bacteria were able to communicate and process various signals programmably. Such 3D printing of programed bacteria enabled the fabrication of living wearable devices (Fig. 41A), and have potentials for practical applications e.g. on-demand medicine and drug delivery. Inspired by human skin, a conductive self-healing DN hydrogel (PAA/polypyrrole) was developed and used for 3D printing of wearable sensors for human movement detection (Fig. 41**B**) [1775]. A hydrogel–elastomer device of softness, transparency, stretchability and conductivity was developed by 3D extrusion printing

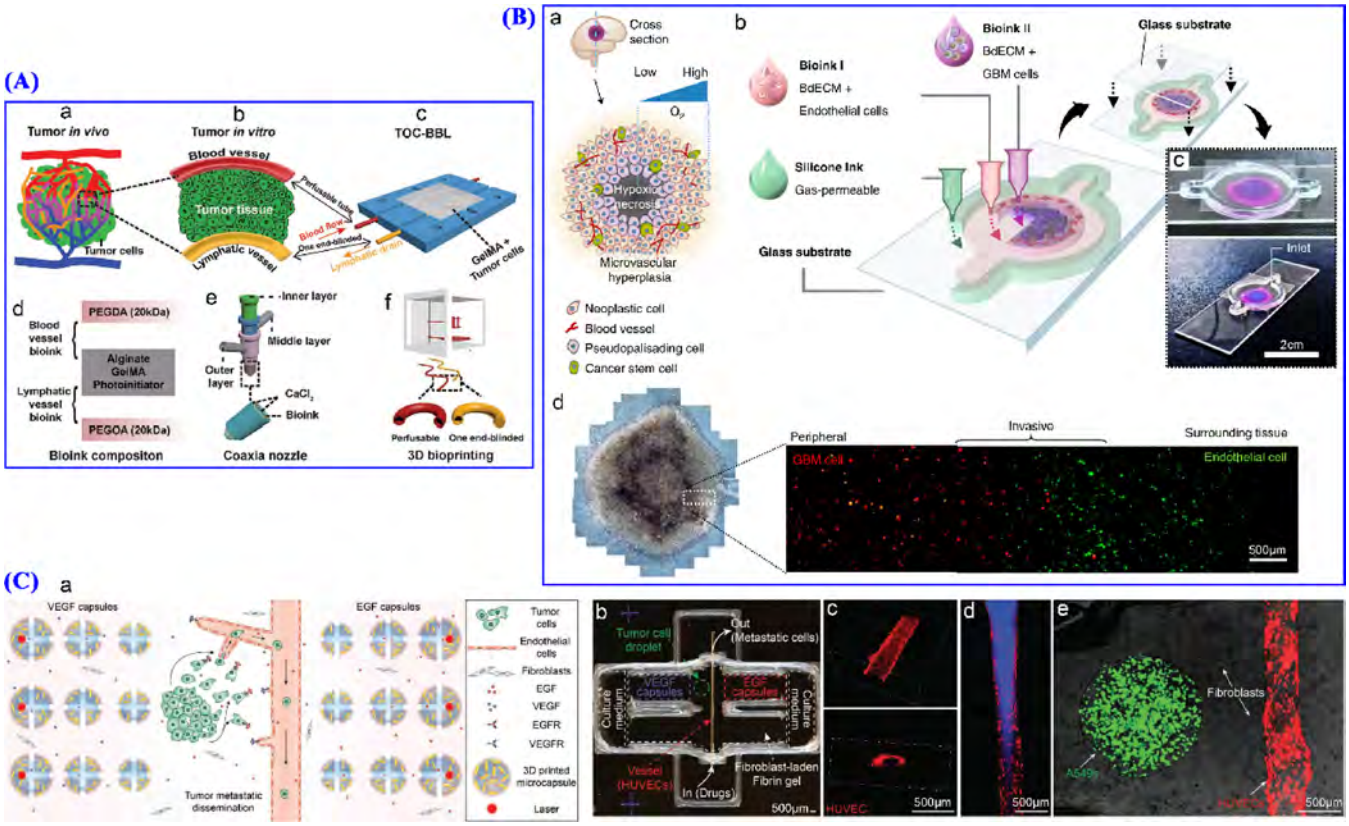

Fig. 39. 3D bioprinting for cancer research. (A) Tumor-on-a-chip with 3D bioprinted blood and lymphatic vessel pair (TOC-BBL). (a) Tumor structure. (b) Blood and lymphatic vessel pair model. (c) TOC-BBL. (d) Bioink formulations. (e) Coaxial nozzle. (f) Bioprinting of two different hollow tubes. Reproduced with permission [1750]. Copyright 2019, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (B) 3D bioprinting of glioblastoma-on-a-chip. (a) Schematic of glioblastoma cross-section. (b) Bioprinting schematic. (c) Photo of glioblastoma-on-a-chip. (d) Microscopy image. Reproduced with permission [1751]. Copyright 2019, The Authors, under exclusive licence to Springer Nature Limited 2019. (C) 3D bioprinting of in vitro metastatic tumor model. (a) Schematic of bioprinted in vitro model to mimic metastatic dissemination. (b) Photo of metastatic tumor model. (c) Lumen image of vessel. (d) Image of vessel perfused with fluorescent fluid. (e) Image of tumor model before capsule rupture by laser. Reproduced with permission [1753]. Copyright 2019, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

technique at a sub-millimeter resolution (Fig. 41**C**) [1776]. Such integrated design and fabrication enabled by 3D printing of hydrogels with dielectric elastomers hold promise for applications in stretchable electronics as well as soft robotics. A proof-of-concept study demonstrated the use of chondrocyte-laden hydrogels for 3D printing a bionic ear, with Ag NPs infused silicone as antenna [1777]. Such 3D printed ear can allow for the readout of inductively coupled signals from the cochlea electrode and improve the auditory sensing ability of radio frequency and stereophonic audio music (Fig. 41D), thereby showing the promising applications of 3D printing for cybernetics to create cyborg organs. The organic integration of wearable electronics/bioelectronics with our human bodies could make a cybernetic future for cyborgs (cybernetic organisms) (Fig. 42) [1778].

7. Conclusions

This Review summarizes recent progress related to the rational design of hydrogels for 3D printing and emerging biomedical applications. Representative techniques of hydrogel-based 3D printing are introduced and discussed. Hydrogel synthesis methods, crosslinking mechanisms, modification approaches and material, physical, chemical or biological properties are discussed. On this basis, the design strategies and recent advancements of biomaterial inks and bioinks for 3D printing and 3D bioprinting are reviewed, along with 4D printing and 4D bioprinting. Combining the material and technical superiorities, hydrogel-based 3D printing enables a wide range of promising biomedical applications to satisfy individualized requirements in a customizable manner.

8. Outlook and future directions

The number of papers has been witnessing an explosive increase and 3D printing will play an increasingly important role in the tissue engineering and regenerative medicine field. In the future, 3D printing can be applied to fabricate multifunctional patient-specific constructs, implants and devices, which are biomimetic, bioactive and biocompatible (3B) to enable the functional replacement, repair or regeneration (3R) of the damaged/diseased tissues and organs in human bodies. Meanwhile, by virtue of the human–machine interactions, the 3D printable wearable electronic devices can achieve on-the-fly health monitoring of human bodies in a customized and individualized way. Furthermore, well established 3D in vitro tissue and organ models by 3D printing of cell-laden natural and/or synthetic hydrogels can allow for high-throughput new drug screening and discovery with the reduced or even avoided animal testing. Enabled by the 3D printing technology, customized tissue/organ grafts and drug delivery systems holds great promise for patient-specific healthcare, personalized medicine and precision medicine.

It is a rising trend to combine multiple 3D printing techniques together to additively manufacture one object, thereby being able to reduce the production period and add the complexities of architecture and functionality. At the same time, new 3D printing techniques and/or instruments will be developed with the advancement of this field. For example, applying external electric, magnetic or acoustic fields during 3D printing can assist in the orientation, alignment, distribution and assembly of functional nanoparticles within the 3D printed objects. Such field-assisted 3D printing enables the design of bioinspired

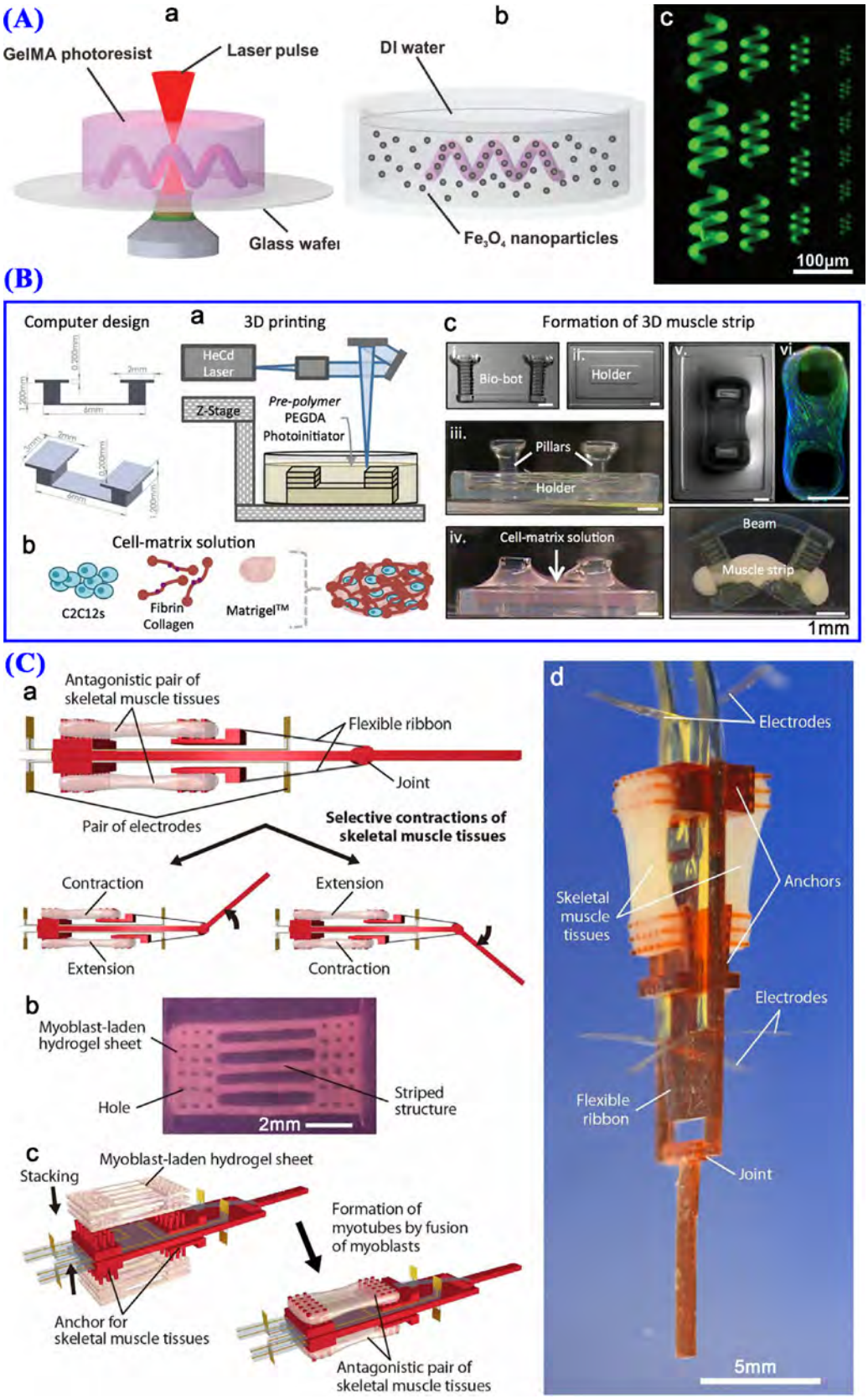

Fig. 40. 3D printing for soft robots. (A) Creation of GelMA hydrogel helical microswimmers. (a) 2 P P printing. (b) Decoration with Fe_3O_4 nanoparticles. (c) Image of helical microstructures. Reproduced with permission [1765]. Copyright 2018, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (B) Printing of hydrogel bio-bot. (a) Design and stereolithography printing. (b) Cell–matrix solution. (c) Bio-bot fabrication. Reproduced with permission [1766]. Copyright 2014, National Academy of Sciences. (C) Creation of biohybrid robot having antagonistic skeletal muscle tissue pair. (a) Schematic of biohybrid robot. (b) Photo of myoblast-laden hydrogel sheet shaped by PDMS stamp. (c) Creation of antagonistic skeletal muscle tissue pair. (d) Photo of biohybrid robot. Reproduced with permission [1767]. Copyright 2018, The Authors, exclusive licensee American Association for the Advancement of Science.

composite reinforcement constructs, e.g. nacre-mimetic structures. Moreover, multimaterial 3D printing is anticipated to increase in demand to create novel functional objects of gradient, heterogenity, anisotropy and complicacy, wherein those sophisticated microstructure characteristics are so far only achievable or accessible by the natural biologic materials/structures. In addition, hand-held 3D printing devices will become increasingly popular due to flexibility and convenience for diverse field applications.

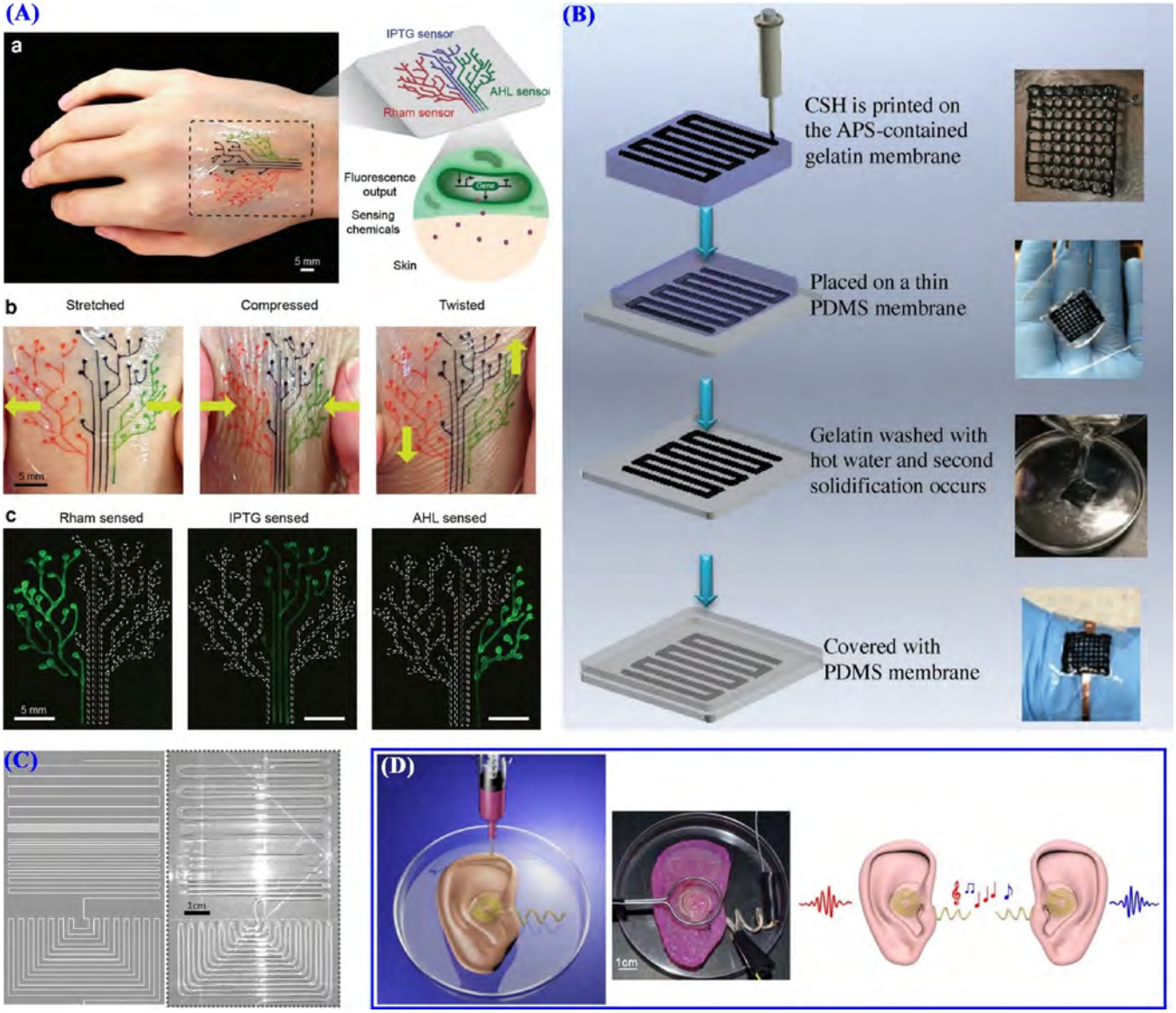

Fig. 41. 3D printable electronics. (A) 3D printing of living tattoo for on-skin chemical detection. (a) Living tattoo design. (b) Different states of on-skin living tattoo. (c) Response of on-skin living tattoo. Reproduced with permission [1593]. Copyright 2017, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (B) 3D printing of wearable sensor with conductive self-healing hydrogels. Reproduced with permission [1775]. Copyright 2017, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (C) Comparison photo of the patterned design (left) and 3D printed hydrogel-on-PDMS sample (right). Reproduced with permission [1776]. Copyright 2017, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (D) 3D printing of bionic ears. Reproduced with permission [1777]. Copyright 2013, American Chemical Society.

Although 3D printing enables the precision manufacturing on the basis of patient-specific anatomy, more attempts and efforts need to be made for developing new biomaterial inks and bioinks with tailorable/appropriate material, mechanical, physical, chemical and biological properties for 3D printing and practical applications. To this end, supramolecular hydrogels point out a charming direction for the development of advanced biomaterial inks and bioinks. On the basis of supramolecular chemistry, each individual building block can be 3D printed first and thereafter, these printed building blocks can self-assemble into larger, more complex, more exquisite and more functional 3D objects through molecular recognition and multivalent interaction, thereby leading to the concept of self-assembly 3D printing. In addition, tough hydrogel-based 3D printing and bioprinting hold great promise for the fabrication of synthetic connective tissues to mimic biologic gel composites such as menisci, tendons, ligaments and articular cartilages, which are very strong and tough structural solids in human body.

Implementation of the 3D printing process under a microgravity or zero-gravity environment will be very meaningful considering the levitation state of biomaterial inks and bioinks, and such explorations enable the promising applications for astronauts in space. Besides, 3D printing may enable developing hydrogel-based metamaterials that present unprecedented properties e.g. mechanical reinforcement. Herein, we define the concept of life materials. Life materials refer to advanced materials that not only contain cells such as mammalian cells or microorganisms (e.g. bacteria, fungi, microalgae), but also can respond to external stimuli such as chemicals or physical fields (e.g. light, electric, magnetic, acoustic, heat). These living materials have life and can self-assemble and shift shapes and achieve functions. 3D bioprinting of cell-laden hydrogels can be used to create a wide range of life materials for applications in drug screening, drug resistance study, photosynthetic therapy, catalysis/biocatalysis, environmental remediation and other industrial applications. All in all, some day the 3D bioprinting scenes of an organ or a human in science fiction movies will no longer be science fiction but become reality, entering our real world.

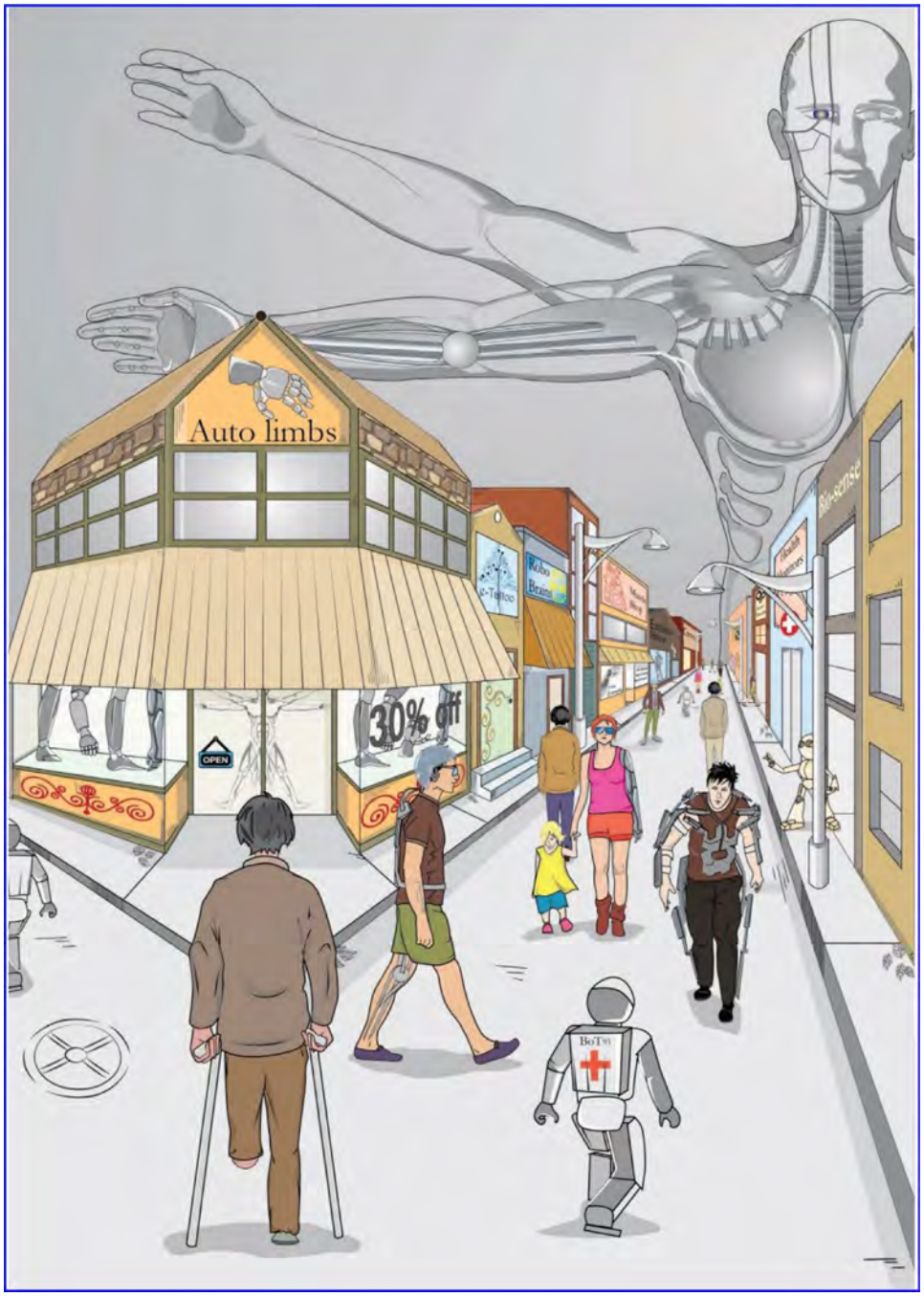

Fig. 42. A cybernetic future for cyborg-like humans. Reproduced with permission [1778]. Copyright 2018, The Authors. Published by WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. This is an open access article under the terms of the Creative Commons Attribution License, which permits use, distribution and reproduction in any medium, provided the original work is properly cited.

Declaration of Competing Interest

The authors declare no competing financial interest.

Acknowledgments

Dr Jinhua Li gratefully acknowledges the funding support from the Alexander von Humboldt Stiftung/Foundation. This work was jointly supported by the German Research Foundation (GE 1133/24-1), National Natural Science Foundation of China (51761135103), Guangdong - Hong Kong Technology Cooperation Funding Scheme (TCFS) (No. GHP/085/18SZ), and Hong Kong Research Grants Council (RGC) General Research Funds (GRF) (No. CityU 11205617).

References

[1] C.W. Hull, Google Patents (1986) US 4575330 A.
[2] B. Mueller, Assem. Autom. 32 (2012).
[3] I. Gibson, D. Rosen, B. Stucker, Additive Manufacturing Technologies: 3D Printing, Rapid Prototyping, and Direct Digital Manufacturing, Springer, 2014.
[4] W. Gao, Y. Zhang, D. Ramanujan, K. Ramani, Y. Chen, C.B. Williams, C.C.L. Wang, Y.C. Shin, S. Zhang, P.D. Zavattieri, Comput. Des. 69 (2015) 65–89.
[5] S.H. Huang, P. Liu, A. Mokasdar, L. Hou, Int. J. Adv. Manuf. Technol. 67 (2013) 1191–1203.
[6] B.C. Gross, J.L. Erkal, S.Y. Lockwood, C. Chen, D.M. Spence, Anal. Chem. 86 (2014) 3240–3253.
[7] D.L. Bourell, Annu. Rev. Mater. Res. 46 (2016) 1–18.
[8] T. Rayna, L. Striukova, Technol. Forecast. Soc. Change 102 (2016) 214–224.
[9] M. Colaco, D.A. Igel, A. Atala, Nat. Rev. Urol. 15 (2018) 213–221.
[10] B. Derby, Science 338 (2012) 921–926.

[11] F.P.W. Melchels, M.A.N. Domingos, T.J. Klein, J. Malda, P.J. Bartolo, D.W. Hutmacher, Prog. Polym. Sci. 37 (2012) 1079–1104.
[12] C.J. Ferris, K.G. Gilmore, G.G. Wallace, M. in het Panhuis, Appl. Microbiol. Biotechnol. 97 (2013) 4243–4258.
[13] S.V. Murphy, A. Atala, Nat. Biotechnol. 32 (2014) 773–785.
[14] L. Moroni, T. Boland, J.A. Burdick, C. De Maria, B. Derby, G. Forgacs, J. Groll, Q. Li, J. Malda, V.A. Mironov, C. Mota, M. Nakamura, W. Shu, S. Takeuchi, T.B.F. Woodfield, T. Xu, J.J. Yoo, G. Vozzi, Trends Biotechnol. 36 (2018) 384–402.
[15] L. Moroni, J.A. Burdick, C. Highley, S.J. Lee, Y. Morimoto, S. Takeuchi, J.J. Yoo, Nat. Rev. Mater. 3 (2018) 21–37.
[16] G. Jürgen, B. Thomas, B. Torsten, A.B. Jason, C. Dong-Woo, D.D. Paul, D. Brian, F. Gabor, L. Qing, A.M. Vladimir, M. Lorenzo, N. Makoto, S. Wenmiao, T. Shoji, V. Giovanni, B.F.W. Tim, X. Tao, J.Y. James, M. Jos, Biofabrication 8 (2016) 013001.
[17] T. DebRoy, H.L. Wei, J.S. Zuback, T. Mukherjee, J.W. Elmer, J.O. Milewski, A.M. Beese, A. Wilson-Heid, A. De, W. Zhang, Prog. Mater. Sci. 92 (2018) 112–224.
[18] D.D. Gu, W. Meiners, K. Wissenbach, R. Poprawe, Int. Mater. Rev. 57 (2012) 133–164.
[19] W.E. Frazier, J. Mater. Eng. Perform. 23 (2014) 1917–1928.
[20] D. Herzog, V. Seyda, E. Wycisk, C. Emmelmann, Acta Mater. 117 (2016) 371–392.
[21] J.J. Lewandowski, M. Seifi, Annu. Rev. Mater. Res. 46 (2016) 151–186.
[22] W.J. Sames, F.A. List, S. Pannala, R.R. Dehoff, S.S. Babu, Int. Mater. Rev. 61 (2016) 315–360.
[23] P.C. Collins, D.A. Brice, P. Samimi, I. Ghamarian, H.L. Fraser, Annu. Rev. Mater. Res. 46 (2016) 63–91.
[24] W.E. King, A.T. Anderson, R.M. Ferencz, N.E. Hodge, C. Kamath, S.A. Khairallah, A.M. Rubenchik, Appl. Phys. Rev. 2 (2015) 041304.
[25] C. Körner, Int. Mater. Rev. 61 (2016) 361–377.
[26] A. Basak, S. Das, Annu. Rev. Mater. Res. 46 (2016) 125–149.
[27] L.E. Murr, S.M. Gaytan, D.A. Ramirez, E. Martinez, J. Hernandez, K.N. Amato, P.W. Shindo, F.R. Medina, R.B. Wicker, J. Mater. Sci. Technol. 28 (2012) 1–14.
[28] K. Zhang, Y. Zhou, C. Xiao, W. Zhao, H. Wu, J. Tang, Z. Li, S. Yu, X. Li, L. Min, Z. Yu, G. Wang, L. Wang, K. Zhang, X. Yang, X. Zhu, C. Tu, X. Zhang, Sci. Adv. 5 (2019) eaax6946.
[29] J.W. Halloran, Annu. Rev. Mater. Res. 46 (2016) 19–40.
[30] J.A. Lewis, J.E. Smay, J. Stuecker, J. Cesarano, J. Am. Ceram. Soc. 89 (2006) 3599–3609.
[31] C. Minas, D. Carnelli, E. Tervoort, A.R. Studart, Adv. Mater. 28 (2016) 9993–9999.
[32] Z.C. Eckel, C. Zhou, J.H. Martin, A.J. Jacobsen, W.B. Carter, T.A. Schaedler, Science 351 (2016) 58–62.
[33] J.A. Lewis, Curr. Opin. Solid State Mater. Sci. 6 (2002) 245–250.
[34] H. Seitz, W. Rieder, S. Irsen, B. Leukers, C. Tille, J. Biomed. Mater. Res. Part B Appl. Biomater. 74B (2005) 782–788.
[35] F. Kotz, K. Arnold, W. Bauer, D. Schild, N. Keller, K. Sachsenheimer, T.M. Nargang, C. Richter, D. Helmer, B.E. Rapp, Nature 544 (2017) 337–339.
[36] G. Marchelli, R. Prabhakar, M. Ganter, D. Storti, Rapid Prototyp. J. 17 (2011) 187–194.
[37] D.T. Nguyen, C. Meyers, T.D. Yee, N.A. Dudukovic, J.F. Destino, C. Zhu, E.B. Duoss, T.F. Baumann, T. Suratwala, J.E. Smay, R. Dylla-Spears, Adv. Mater. 29 (2017) 1701181.
[38] Q. Fu, E. Saiz, A.P. Tomsia, Adv. Funct. Mater. 21 (2011) 1058–1063.
[39] C. Wu, Y. Luo, G. Cuniberti, Y. Xiao, M. Gelinsky, Acta Biomater. 7 (2011) 2644–2650.
[40] F. Kotz, P. Risch, K. Arnold, S. Sevim, J. Puigmartí-Luis, A. Quick, M. Thiel, A. Hrynevich, P.D. Dalton, D. Helmer, B.E. Rapp, Nat. Commun. 10 (2019) 1439.
[41] S.C. Ligon, R. Liska, J. Stampfl, M. Gurr, R. Mülhaupt, Chem. Rev. 117 (2017) 10212–10290.
[42] X. Wang, M. Jiang, Z. Zhou, J. Gou, D. Hui, Compos. Part B Eng. 110 (2017) 442–458.
[43] J.W. Stansbury, M.J. Idacavage, Dent. Mater. 32 (2016) 54–64.
[44] B. Wendel, D. Rietzel, F. Kühnlein, R. Feulner, G. Hülder, E. Schmachtenberg, Macromol. Mater. Eng. 293 (2008) 799–809.
[45] D.M. Kirchmajer, R. Gorkin Iii, M. in het Panhuis, J. Mater. Chem. B 3 (2015) 4105–4117.
[46] N. Guo, M.C. Leu, Front. Mech. Eng. 8 (2013) 215–243.
[47] M. Hofmann, ACS Macro Lett. 3 (2014) 382–386.
[48] A. Bandyopadhyay, B. Heer, Mater. Sci. Eng. R Rep. 129 (2018) 1–16.
[49] D. Kokkinis, M. Schaffner, A.R. Studart, Nat. Commun. 6 (2015) 8643.
[50] D. Gräfe, A. Wickberg, M.M. Zieger, M. Wegener, E. Blasco, C. Barner-Kowollik, Nat. Commun. 9 (2018) 2788.
[51] E. Atzeni, A. Salmi, Int. J. Adv. Manuf. Technol. 62 (2012) 1147–1155.
[52] S. Mellor, L. Hao, D. Zhang, Int. J. Prod. Econ. 149 (2014) 194–201.
[53] C. Weller, R. Kleer, F.T. Piller, Int. J. Prod. Econ. 164 (2015) 43–56.
[54] E. MacDonald, R. Wicker, Science 353 (2016) aaf2093.
[55] C.K. Chua, K.F. Leong, C.S. Lim, Rapid Prototyping: Principles and Applications, World Scientific, 2010.
[56] M. Ficko, I. Drstvenšek, M. Brezočnik, J. Balič, B. Vaupotic, J. Mater. Process. Technol. 164–165 (2005) 1327–1335.
[57] B. Berman, Bus. Horiz. 55 (2012) 155–162.
[58] M. Attaran, Bus. Horiz. 60 (2017) 677–688.
[59] I.J. Petrick, T.W. Simpson, Res. Manag. 56 (2013) 12–16.
[60] J.D. Prince, J. Electron. Resour. Med. Libr. 11 (2014) 39–45.
[61] C. Schubert, M.C. van Langeveld, L.A. Donoso, Br. J. Ophthalmol. 98 (2014) 159.
[62] L.E. Murr, J. Mater. Sci. Technol. 32 (2016) 987–995.
[63] L. Nickels, Met. Powder Rep. 70 (2015) 300–303.
[64] S.C. Joshi, A.A. Sheikh, Virtual Phys. Prototyp. 10 (2015) 175–185.
[65] N. de Beer, D. Dimitrov, K. Schreve, W. van Wijck, Rapid Prototyp. J. 12 (2006) 42–52.
[66] M.M. Stanton, C. Trichet-Paredes, S. Sánchez, Lab Chip 15 (2015) 1634–1637.
[67] J.Z. Gul, M. Sajid, M.M. Rehman, G.U. Siddiqui, I. Shah, K.-H. Kim, J.-W. Lee, K.H. Choi, Sci. Technol. Adv. Mater. 19 (2018) 243–262.
[68] N.W. Bartlett, M.T. Tolley, J.T.B. Overvelde, J.C. Weaver, B. Mosadegh, K. Bertoldi, G.M. Whitesides, R.J. Wood, Science 349 (2015) 161–165.
[69] K. Sun, T.-S. Wei, B.Y. Ahn, J.Y. Seo, S.J. Dillon, J.A. Lewis, Adv. Mater. 25 (2013) 4539–4543.
[70] F. Zhang, M. Wei, V.V. Viswanathan, B. Swart, Y. Shao, G. Wu, C. Zhou, Nano Energy 40 (2017) 418–431.
[71] X. Tian, J. Jin, S. Yuan, C.K. Chua, S.B. Tor, K. Zhou, Adv. Energy Mater. 7 (2017) 1700127.
[72] A. Ambrosi, M. Pumera, Chem. Soc. Rev. 45 (2016) 2740–2755.
[73] K. Fu, Y. Yao, J. Dai, L. Hu, Adv. Mater. 29 (2017) 1603486.
[74] P.P. Rodenbough, W.B. Vanti, S.-W. Chan, J. Chem. Educ. 92 (2015) 1960–1962.
[75] P. Lolur, R. Dawes, J. Chem. Educ. 91 (2014) 1181–1184.
[76] S. Rossi, M. Benaglia, D. Brenna, R. Porta, M. Orlandi, J. Chem. Educ. 92 (2015) 1398–1401.
[77] P.G. McMenamin, M.R. Quayle, C.R. McHenry, J.W. Adams, Anat. Sci. Educ. 7 (2014) 479–486.
[78] F.C. Godoi, S. Prakash, B.R. Bhandari, J. Food Eng. 179 (2016) 44–54.
[79] J. Sun, W. Zhou, D. Huang, J.Y.H. Fuh, G.S. Hong, Food Bioproc. Tech. 8 (2015) 1605–1615.
[80] J. Sun, Z. Peng, W. Zhou, J.Y.H. Fuh, G.S. Hong, A. Chiu, Procedia Manuf. 1 (2015) 308–319.
[81] F. Ilievski, A.D. Mazzeo, R.F. Shepherd, X. Chen, G.M. Whitesides, Angew. Chemie 123 (2011) 1930–1935.
[82] K.I. Galkin, V.P. Ananikov, Russ. Chem. Rev. 85 (2016) 226.
[83] B. Gross, S.Y. Lockwood, D.M. Spence, Anal. Chem. 89 (2017) 57–70.
[84] R.D. Johnson, Nat. Chem. 4 (2012) 338–339.
[85] S.J. Trenfield, A. Awad, A. Goyanes, S. Gaisford, A.W. Basit, Trends Pharmacol. Sci. 39 (2018) 440–451.
[86] A. Goyanes, J. Wang, A. Buanz, R. Martínez-Pacheco, R. Telford, S. Gaisford, A.W. Basit, Mol. Pharm. 12 (2015) 4077–4084.
[87] J. Norman, R.D. Madurawe, C.M.V. Moore, M.A. Khan, A. Khairuzzaman, Adv. Drug Deliv. Rev. 108 (2017) 39–50.
[88] I.D. Ursan, L. Chiu, A. Pierce, J. Am. Pharm. Assoc. 53 (2013) 136–144.
[89] J. Goole, K. Amighi, Int. J. Pharm. 499 (2016) 376–394.
[90] A. Awad, S.J. Trenfield, A. Goyanes, S. Gaisford, A.W. Basit, Drug Discov. Today 23 (2018) 1547–1555.
[91] L.K. Prasad, H. Smyth, Drug Dev. Ind. Pharm. 42 (2016) 1019–1031.
[92] S. Bose, S. Vahabzadeh, A. Bandyopadhyay, Mater. Today 16 (2013) 496–504.
[93] N.A. Sears, D.R. Seshadri, P.S. Dhavalikar, E. Cosgriff-Hernandez, Tissue Eng. Part B Rev. 22 (2016) 298–310.
[94] M.S. Sulkin, E. Widder, C. Shao, K.M. Holzem, C. Gloschat, S.R. Gutbrod, I.R. Efimov, American Journal of Physiology-Heart and Circulatory Physiology 305 (2013) H1569–H1573.
[95] M.J. Lerman, J. Lembong, G. Gillen, J.P. Fisher, Appl. Phys. Rev. 5 (2018) 041109.
[96] C. Yu, W. Zhu, B. Sun, D. Mei, M. Gou, S. Chen, Appl. Phys. Rev. 5 (2018) 041107.
[97] C.L. Ventola, Pharmacy and Therapeutics 39 (2014) 704–711.
[98] J.K. Placone, A.J. Engler, Adv. Healthc. Mater. 7 (2018) 1701161.
[99] U. Ghosh, S. Ning, Y. Wang, Y.L. Kong, Adv. Healthc. Mater. 7 (2018) 1800417.
[100] T.S. Shimizu, N. Le Novère, M.D. Levin, A.J. Beavil, B.J. Sutton, D. Bray, Nat. Cell Biol. 2 (2000) 792–796.
[101] P.J. Kitson, S. Glatzel, W. Chen, C.-G. Lin, Y.-F. Song, L. Cronin, Nat. Protoc. 11 (2016) 920–936.
[102] M.D. Symes, P.J. Kitson, J. Yan, C.J. Richmond, G.J.T. Cooper, R.W. Bowman, T. Vilbrandt, L. Cronin, Nat. Chem. 4 (2012) 349–354.
[103] A.J. Capel, R.P. Rimington, M.P. Lewis, S.D.R. Christie, Nat. Rev. Chem. 2 (2018) 422–436.
[104] J.M. Pearce, Science 337 (2012) 1303–1304.
[105] F. Kotz, P. Risch, D. Helmer, B.E. Rapp, Adv. Mater. 31 (2019) 1805982.
[106] A. Zolfagharian, A.Z. Kouzani, S.Y. Khoo, A.A.A. Moghadam, I. Gibson, A. Kaynak, Sens. Actuators A Phys. 250 (2016) 258–272.
[107] C. de Marco, S. Pané, B.J. Nelson, Sci. Robot. 3 (2018) eaau0449.
[108] P. Sarobol, A. Cook, P.G. Clem, D. Keicher, D. Hirschfeld, A.C. Hall, N.S. Bell, Annu. Rev. Mater. Res. 46 (2016) 41–62.
[109] G. Hu, J. Kang, L.W.T. Ng, X. Zhu, R.C.T. Howe, C.G. Jones, M.C. Hersam, T. Hasan, Chem. Soc. Rev. 47 (2018) 3265–3300.
[110] J. Li, W. Tang, Q. Wang, W. Sun, Q. Zhang, X. Guo, X. Wang, F. Yan, Mater. Sci. Eng. R Rep. 127 (2018) 1–36.
[111] D. Chimene, K.K. Lennox, R.R. Kaunas, A.K. Gaharwar, Ann. Biomed. Eng. 44 (2016) 2090–2102.
[112] T.D. Ngo, A. Kashani, G. Imbalzano, K.T.Q. Nguyen, D. Hui, Compos. Part B Eng. 143 (2018) 172–196.
[113] S. Bose, D. Ke, H. Sahasrabudhe, A. Bandyopadhyay, Prog. Mater. Sci. 93 (2018) 45–111.
[114] M. Guvendiren, J. Molde, R.M.D. Soares, J. Kohn, ACS Biomater. Sci. Eng. 2 (2016) 1679–1693.
[115] J.-Y. Lee, J. An, C.K. Chua, Appl. Mater. Today 7 (2017) 120–133.
[116] E.J. Adam, L.R. Alexandra, N.S. Ramille, Biomed. Mater. 11 (2016) 014102.
[117] W. Zhu, X. Ma, M. Gou, D. Mei, K. Zhang, S. Chen, Curr. Opin. Biotechnol. 40 (2016) 103–112.
[118] H.N. Chia, B.M. Wu, J. Biol. Eng. 9 (2015) 4.

[119] A. Marrella, T.Y. Lee, D.H. Lee, S. Karuthedom, D. Syla, A. Chawla, A. Khademhosseini, H.L. Jang, Mater. Today 21 (2018) 362–376.
[120] E.J. Lee, B.K. Huh, S.N. Kim, J.Y. Lee, C.G. Park, A.G. Mikos, Y.B. Choy, Prog. Mater. Sci. 89 (2017) 392–410.
[121] C. Loebel, J.A. Burdick, Cell Stem Cell 22 (2018) 325–339.
[122] B.A. Aguado, J.C. Grim, A.M. Rosales, J.J. Watson-Capps, K.S. Anseth, Sci. Transl. Med. 10 (2018) eaam8645.
[123] C.D. Spicer, E.T. Pashuck, M.M. Stevens, Chem. Rev. 118 (2018) 7702–7743.
[124] S. Pina, M. Oliveira Joaquim, L. Reis Rui, Adv. Mater. 27 (2015) 1143–1169.
[125] Y. Li, Y. Xiao, C. Liu, Chem. Rev. 117 (2017) 4376–4421.
[126] M.P. Lutolf, P.M. Gilbert, H.M. Blau, Nature 462 (2009) 433–441.
[127] A. Higuchi, Q.-D. Ling, Y. Chang, S.-T. Hsu, A. Umezawa, Chem. Rev. 113 (2013) 3297–3328.
[128] M.P. Lutolf, J.A. Hubbell, Nat. Biotechnol. 23 (2005) 47–55.
[129] E.T. Pashuck, M.M. Stevens, Sci. Transl. Med. 4 (2012) 160sr164.
[130] M.M. Stevens, Mater. Today 11 (2008) 18–25.
[131] M.W. Tibbitt, C.B. Rodell, J.A. Burdick, K.S. Anseth, Proc. Natl. Acad. Sci. 112 (2015) 14444–14451.
[132] O. Bas, E.M. De-Juan-Pardo, I. Catelas, D.W. Hutmacher, Adv. Drug Deliv. Rev. 132 (2018) 214–234.
[133] M. Bohner, Mater. Today 13 (2010) 24–30.
[134] U. Sharma, D. Concagh, L. Core, Y. Kuang, C. You, Q. Pham, G. Zugates, R. Busold, S. Webber, J. Merlo, R. Langer, George M. Whitesides, M. Palasis, Nat. Mater. 17 (2018) 96–103.
[135] L.L. Hench, J.M. Polak, Science 295 (2002) 1014–1017.
[136] N.A. Peppas, R. Langer, Science 263 (1994) 1715–1720.
[137] J.A. Hubbell, BioTechnology 13 (1995) 565.
[138] J.P. Vacanti, R. Langer, Lancet 354 (1999) S32–S34.
[139] D.W. Hutmacher, Biomaterials 21 (2000) 2529–2543.
[140] A. Ovsianikov, A. Khademhosseini, V. Mironov, Trends Biotechnol. 36 (2018) 348–357.
[141] F.J. O'Brien, Mater. Today 14 (2011) 88–95.
[142] L.G. Griffith, G. Naughton, Science 295 (2002) 1009–1014.
[143] K. Rezwan, Q.Z. Chen, J.J. Blaker, A.R. Boccaccini, Biomaterials 27 (2006) 3413–3431.
[144] S. Wu, X. Liu, K.W.K. Yeung, C. Liu, X. Yang, Mater. Sci. Eng. R Rep. 80 (2014) 1–36.
[145] S.J. Hollister, Nat. Mater. 4 (2005) 518–524.
[146] S. Yang, K.-F. Leong, Z. Du, C.-K. Chua, Tissue Eng. 7 (2001) 679–689.
[147] S. Bose, M. Roy, A. Bandyopadhyay, Trends Biotechnol. 30 (2012) 546–554.
[148] K.F. Leong, C.M. Cheah, C.K. Chua, Biomaterials 24 (2003) 2363–2378.
[149] D.W. Hutmacher, M. Sittinger, M.V. Risbud, Trends Biotechnol. 22 (2004) 354–362.
[150] A. Kumar, S. Mandal, S. Barui, R. Vasireddi, U. Gbureck, M. Gelinsky, B. Basu, Mater. Sci. Eng. R Rep. 103 (2016) 1–39.
[151] S.M. Bittner, J.L. Guo, A. Melchiorri, A.G. Mikos, Mater. Today 21 (2018) 861–874.
[152] A.-V. Do, B. Khorsand, S.M. Geary, A.K. Salem, Adv. Healthc. Mater. 4 (2015) 1742–1762.
[153] J. Groll, J.A. Burdick, D.W. Cho, B. Derby, M. Gelinsky, S.C. Heilshorn, T. Jüngst, J. Malda, V.A. Mironov, K. Nakayama, A. Ovsianikov, W. Sun, S. Takeuchi, J.J. Yoo, T.B.F. Woodfield, Biofabrication 11 (2019) 013001.
[154] A. Skardal, A. Atala, Ann. Biomed. Eng. 43 (2015) 730–746.
[155] Z. Yue, X. Liu, P.T. Coates, G.G. Wallace, Curr. Opin. Organ Transplant. 21 (2016) 467–475.
[156] T.-S. Jang, H.-D. Jung, H.M. Pan, W.T. Han, S. Chen, J. Song, Int. J. Bioprinting 4 (2018) 126.
[157] T. Jungst, W. Smolan, K. Schacht, T. Scheibel, J. Groll, Chem. Rev. 116 (2016) 1496–1539.
[158] J.-F. Xing, M.-L. Zheng, X.-M. Duan, Chem. Soc. Rev. 44 (2015) 5031–5039.
[159] M.W. Tibbitt, K.S. Anseth, Biotechnol. Bioeng. 103 (2009) 655–663.
[160] J.A. Rowley, G. Madlambayan, D.J. Mooney, Biomaterials 20 (1999) 45–53.
[161] H. Geckil, F. Xu, X. Zhang, S. Moon, U. Demirci, Nanomedicine 5 (2010) 469–484.
[162] A.M. Rosales, K.S. Anseth, Nat. Rev. Mater. 1 (2016) 15012.
[163] J. Leijten, J. Seo, K. Yue, G. Trujillo-de Santiago, A. Tamayol, G.U. Ruiz-Esparza, S.R. Shin, R. Sharifi, I. Noshadi, M.M. Álvarez, Y.S. Zhang, A. Khademhosseini, Mater. Sci. Eng. R Rep. 119 (2017) 1–35.
[164] N. Annabi, A. Tamayol, J.A. Uquillas, M. Akbari, L.E. Bertassoni, C. Cha, G. Camci-Unal, M.R. Dokmeci, N.A. Peppas, A. Khademhosseini, Adv. Mater. 26 (2014) 85–124.
[165] B.V. Slaughter, S.S. Khurshid, O.Z. Fisher, A. Khademhosseini, N.A. Peppas, Adv. Mater. 21 (2009) 3307–3329.
[166] E. Jabbari, J. Leijten, Q. Xu, A. Khademhosseini, Mater. Today 19 (2016) 190–196.
[167] Y.S. Zhang, A. Khademhosseini, Science 356 (2017) eaaf3627.
[168] K.Y. Lee, D.J. Mooney, Chem. Rev. 101 (2001) 1869–1880.
[169] N.A. Peppas, J.Z. Hilt, A. Khademhosseini, R. Langer, Adv. Mater. 18 (2006) 1345–1360.
[170] A. Singh, N.A. Peppas, Adv. Mater. 26 (2014) 6530–6541.
[171] G. Camci-Unal, N. Annabi, M.R. Dokmeci, R. Liao, A. Khademhosseini, NPG Asia Mater. 6 (2014) e99.
[172] A.S. Hoffman, Adv. Drug Deliv. Rev. 64 (2012) 18–23.
[173] K.T. Nguyen, J.L. West, Biomaterials 23 (2002) 4307–4314.
[174] J. Thiele, Y. Ma, S.M.C. Bruekers, S. Ma, W.T.S. Huck, Adv. Mater. 26 (2014) 125–148.
[175] J.L. Drury, D.J. Mooney, Biomaterials 24 (2003) 4337–4351.
[176] J.A. Burdick, W.L. Murphy, Nat. Commun. 3 (2012) 1269.
[177] D. Seliktar, Science 336 (2012) 1124–1128.
[178] A.M. Kloxin, C.J. Kloxin, C.N. Bowman, K.S. Anseth, Adv. Mater. 22 (2010) 3484–3494.
[179] T.E. Brown, K.S. Anseth, Chem. Soc. Rev. 46 (2017) 6532–6552.
[180] P.M. Kharkar, K.L. Kiick, A.M. Kloxin, Chem. Soc. Rev. 42 (2013) 7335–7372.
[181] H.W. Ooi, S. Hafeez, C.A. van Blitterswijk, L. Moroni, M.B. Baker, Mater. Horiz. 4 (2017) 1020–1040.
[182] R.L. Youngblood, N.F. Truong, T. Segura, L.D. Shea, Mol. Ther. 26 (2018) 2087–2106.
[183] J. Jia, D.J. Richards, S. Pollard, Y. Tan, J. Rodriguez, R.P. Visconti, T.C. Trusk, M.J. Yost, H. Yao, R.R. Markwald, Y. Mei, Acta Biomater. 10 (2014) 4323–4331.
[184] B.J. Klotz, D. Gawlitta, A.J.W.P. Rosenberg, J. Malda, F.P.W. Melchels, Trends Biotechnol. 34 (2016) 394–407.
[185] J. Malda, J. Visser, F.P. Melchels, T. Jüngst, W.E. Hennink, W.J.A. Dhert, J. Groll, D.W. Hutmacher, Adv. Mater. 25 (2013) 5011–5028.
[186] T. Billiet, M. Vandenhaute, J. Schelfhout, S. Van Vlierberghe, P. Dubruel, Biomaterials 33 (2012) 6020–6041.
[187] J.M. Lee, W.Y. Yeong, Adv. Healthc. Mater. 5 (2016) 2856–2865.
[188] M.M. Stanton, J. Samitier, S. Sánchez, Lab Chip 15 (2015) 3111–3115.
[189] P.S. Gungor-Ozkerim, I. Inci, Y.S. Zhang, A. Khademhosseini, M.R. Dokmeci, Biomater. Sci. 6 (2018) 915–946.
[190] K. Hölzl, S. Lin, L. Tytgat, S. Van Vlierberghe, L. Gu, A. Ovsianikov, Biofabrication 8 (2016) 032002.
[191] S. Abdulghani, P.G. Morouço, J. Mater. Sci. Mater. Med. 30 (2019) 20.
[192] S. Miao, N. Castro, M. Nowicki, L. Xia, H. Cui, X. Zhou, W. Zhu, S.-j. Lee, K. Sarkar, G. Vozzi, Y. Tabata, J. Fisher, L.G. Zhang, Mater. Today 20 (2017) 577–591.
[193] Z.X. Khoo, J.E.M. Teoh, Y. Liu, C.K. Chua, S. Yang, J. An, K.F. Leong, W.Y. Yeong, Virtual Phys. Prototyp. 10 (2015) 103–122.
[194] S.K. Leist, J. Zhou, Virtual Phys. Prototyp. 11 (2016) 249–262.
[195] L. Yi-Chen, Z. Yu Shrike, A. Ali, S. Su Ryon, K. Ali, Biofabrication 9 (2017) 012001.
[196] N. Ashammakhi, S. Ahadian, F. Zengjie, K. Suthiwanich, F. Lorestani, G. Orive, S. Ostrovidov, A. Khademhosseini, Biotechnol. J. 13 (2018) 1800148.
[197] P. Morouço, W. Lattanzi, N. Alves, Front. Bioeng. Biotechnol. 5 (2017) 61.
[198] B. Gao, Q. Yang, X. Zhao, G. Jin, Y. Ma, F. Xu, Trends Biotechnol. 34 (2016) 746–756.
[199] L. Ionov, Adv. Healthc. Mater. 7 (2018) 1800412.
[200] A. Panwar, L.P. Tan, Molecules 21 (2016) 685.
[201] A.L. Rutz, P.L. Lewis, R.N. Shah, MRS Bull. 42 (2017) 563–570.
[202] S.E. Critchley, D.J. Kelly, J. 3d Print. Med. 1 (2017) 269–290.
[203] N.R. Schiele, D.T. Corr, Y. Huang, N.A. Raof, Y. Xie, D.B. Chrisey, Biofabrication 2 (2010) 032001.
[204] J.P. Kruth, X. Wang, T. Laoui, L. Froyen, Assem. Autom. 23 (2003) 357–371.
[205] S. Seyed Farid Seyed, G. Samira, M. Mehdi, Y. Hooman, M. Hendrik Simon Cornelis, K. Nahrizul Adib, O. Noor Azuan Abu, Sci. Technol. Adv. Mater. 16 (2015) 033502.
[206] F.P.W. Melchels, J. Feijen, D.W. Grijpma, Biomaterials 31 (2010) 6121–6130.
[207] R.J. Mondschein, A. Kanitkar, C.B. Williams, S.S. Verbridge, T.E. Long, Biomaterials 140 (2017) 170–188.
[208] J. Zhang, P. Xiao, Polym. Chem. 9 (2018) 1530–1540.
[209] R.J. Narayan, A. Doraiswamy, D.B. Chrisey, B.N. Chichkov, Mater. Today 13 (2010) 42–48.
[210] R. Liska, M. Schuster, R. Inführ, C. Turecek, C. Fritscher, B. Seidl, V. Schmidt, L. Kuna, A. Haase, F. Varga, H. Lichtenegger, J. Stampfl, J. Coat. Technol. Res. 4 (2007) 505–510.
[211] J. Torgersen, X.-H. Qin, Z. Li, A. Ovsianikov, R. Liska, J. Stampfl, Adv. Funct. Mater. 23 (2013) 4542–4554.
[212] M. Rumi, J.W. Perry, Adv. Opt. Photonics 2 (2010) 451–518.
[213] K.-S. Lee, R.H. Kim, D.-Y. Yang, S.H. Park, Prog. Polym. Sci. 33 (2008) 631–681.
[214] M. Pawlicki, H.A. Collins, R.G. Denning, H.L. Anderson, Angew. Chemie Int. Ed. 48 (2009) 3244–3266.
[215] M. Farsari, B.N. Chichkov, Nat. Photonics 3 (2009) 450–452.
[216] J. Bohandy, B.F. Kim, F.J. Adrian, J. Appl. Phys. 60 (1986) 1538–1539.
[217] P. Serra, A. Piqué, Adv. Mater. Technol. 4 (2019) 1800099.
[218] P. Delaporte, A.-P. Alloncle, Opt. Laser Technol. 78 (2016) 33–41.
[219] B. Guillotin, F. Guillemot, Trends Biotechnol. 29 (2011) 183–190.
[220] B.R. Ringeisen, C.M. Othon, J.A. Barron, D. Young, B.J. Spargo, Biotechnol. J. 1 (2006) 930–948.
[221] A. Ovsianikov, M. Gruene, M. Pflaum, L. Koch, F. Maiorana, M. Wilhelmi, A. Haverich, B. Chichkov, Biofabrication 2 (2010) 014104.
[222] J.A. Lewis, Adv. Funct. Mater. 16 (2006) 2193–2204.
[223] I. Zein, D.W. Hutmacher, K.C. Tan, S.H. Teoh, Biomaterials 23 (2002) 1169–1185.
[224] D. Brown Toby, D. Dalton Paul, W. Hutmacher Dietmar, Adv. Mater. 23 (2011) 5651–5657.
[225] R. Landers, R. Mülhaupt, Macromol. Mater. Eng. 282 (2000) 17–21.
[226] I.T. Ozbolat, M. Hospodiuk, Biomaterials 76 (2016) 321–343.
[227] A. Shapira, N. Noor, M. Asulin, T. Dvir, Appl. Phys. Rev. 5 (2018) 041112.
[228] P. Calvert, Chem. Mater. 13 (2001) 3299–3305.
[229] E. Sachs, M. Cima, P. Williams, D. Brancazio, J. Cornie, J. Eng. Ind. 114 (1992) 481–488.
[230] M.M. Mohebi, J.R.G. Evans, J. Comb. Chem. 4 (2002) 267–274.
[231] M. Singh, H.M. Haverinen, P. Dhagat, G.E. Jabbour, Adv. Mater. 22 (2010) 673–685.
[232] B. Derby, Annu. Rev. Mater. Res. 40 (2010) 395–414.
[233] B. Derby, J. Mater. Chem. 18 (2008) 5717–5721.
[234] T. Boland, T. Xu, B. Damon, X. Cui, Biotechnol. J. 1 (2006) 910–917.
[235] B.-J. de Gans, P.C. Duineveld, U.S. Schubert, Adv. Mater. 16 (2004) 203–213.

[236] D. Kuckling, K.-F. Arndt, S. Richter, Synthesis of Hydrogels, in: G. Gerlach, K.-F. Arndt (Eds.), Hydrogel Sensors and Actuators: Engineering and Technology, vol 6, Springer, Berlin Heidelberg, Berlin, Heidelberg, 2010, pp. 15–67.
[237] Y. Li, J. Rodrigues, H. Tomas, Chem. Soc. Rev. 41 (2012) 2193–2221.
[238] M. Hamidi, A. Azadi, P. Rafiei, Adv. Drug Deliv. Rev. 60 (2008) 1638–1649.
[239] P. Gupta, K. Vermani, S. Garg, Drug Discov. Today 7 (2002) 569–579.
[240] J. Li, D.J. Mooney, Nat. Rev. Mater. 1 (2016) 16071.
[241] M.E. Wechsler, R.E. Stephenson, A.C. Murphy, H.F. Oldenkamp, A. Singh, N.A. Peppas, Biomed. Microdevices 21 (2019) 31.
[242] Q. Huang, Y. Zou, M.C. Arno, S. Chen, T. Wang, J. Gao, A.P. Dove, J. Du, Chem. Soc. Rev. 46 (2017) 6255–6275.
[243] S.Y. Chin, Y.C. Poh, A.-C. Kohler, J.T. Compton, L.L. Hsu, K.M. Lau, S. Kim, B.W. Lee, F.Y. Lee, S.K. Sia, Sci. Robot. 2 (2017) eaah6451.
[244] M. Irimia-Vladu, Chem. Soc. Rev. 43 (2014) 588–610.
[245] H.R. Culver, J.R. Clegg, N.A. Peppas, Acc. Chem. Res. 50 (2017) 170–178.
[246] I.Y. Jung, J.S. Kim, B.R. Choi, K. Lee, H. Lee, Adv. Healthc. Mater. 6 (2017) 1601475.
[247] N.A. Peppas, D.S. Van Blarcom, J. Control. Release 240 (2016) 142–150.
[248] M. Guenther, G. Gerlach, Hydrogels for Chemical Sensors, in: G. Gerlach, K.-F. Arndt (Eds.), Hydrogel Sensors and Actuators: Engineering and Technology, vol 6, Springer, Berlin Heidelberg, Berlin, Heidelberg, 2010, pp. 165–195.
[249] D.T. Eddington, D.J. Beebe, Adv. Drug Deliv. Rev. 56 (2004) 199–210.
[250] L. Dong, H. Jiang, Soft Matter 3 (2007) 1223–1230.
[251] A. Richter, Hydrogels for Actuators, in: G. Gerlach, K.-F. Arndt (Eds.), Hydrogel Sensors and Actuators: Engineering and Technology, vol 6, Springer, Berlin Heidelberg, Berlin, Heidelberg, 2010, pp. 221–248.
[252] A.C. Kumar, H. Erothu, Synthetic Polymer Hydrogels, Biomedical Applications of Polymeric Materials and Composites, Wiley-VCH Verlag GmbH & Co. KGaA, 2016, pp. 141–162.
[253] H.G. Schild, Prog. Polym. Sci. 17 (1992) 163–249.
[254] W.E. Hennink, C.F. van Nostrum, Adv. Drug Deliv. Rev. 54 (2002) 13–36.
[255] Y. Cao, M.L. Mei, Q.-L. Li, E.C.M. Lo, C.H. Chu, ACS Appl. Mater. Interfaces 6 (2014) 410–420.
[256] J.E. Eldridge, J.D. Ferry, J. Phys. Chem. 58 (1954) 992–995.
[257] T. Funami, M. Hiroe, S. Noda, I. Asai, S. Ikeda, K. Nishinari, Food Hydrocoll. 21 (2007) 617–629.
[258] K.E. Crompton, J.D. Goud, R.V. Bellamkonda, T.R. Gengenbach, D.I. Finkelstein, M.K. Horne, J.S. Forsythe, Biomaterials 28 (2007) 441–449.
[259] Z. He, J.L. Santos, H. Tian, H. Huang, Y. Hu, L. Liu, K.W. Leong, Y. Chen, H.-Q. Mao, Biomaterials 130 (2017) 28–41.
[260] P. De Vos, B. De Haan, R. Van Schilfgaarde, Biomaterials 18 (1997) 273–278.
[261] A.D. Augst, H.J. Kong, D.J. Mooney, Macromol. Biosci. 6 (2006) 623–633.
[262] N.A. Peppas, Die Makromol. Chemie 176 (1975) 3433–3440.
[263] P. Giannouli, E.R. Morris, Food Hydrocoll. 17 (2003) 495–501.
[264] S. Argin-Soysal, P. Kofinas, Y.M. Lo, Food Hydrocoll. 23 (2009) 202–209.
[265] M. Takigami, N. Nagasawa, Y. Maehara, S. Takigami, M. Tamada, Preparation of CMC Acid Gel and Its Applications, Gums and Stabilisers for the Food Industry 15, The Royal Society of Chemistry, 2009, pp. 427–434.
[266] A.B. Lugao, S.M. Malmonge, Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions With Materials and Atoms 185 (2001), pp. 37–42.
[267] R. Francis, N. Joy, A. Sivadas, Relevance of Natural Degradable Polymers in the Biomedical Field, Biomedical Applications of Polymeric Materials and Composites, Wiley-VCH Verlag GmbH & Co. KGaA, 2016, pp. 303–360.
[268] B. Balakrishnan, R. Banerjee, Chem. Rev. 111 (2011) 4453–4474.
[269] U. Freudenberg, Y. Liang, K.L. Kiick, C. Werner, Adv. Mater. 28 (2016) 8861–8891.
[270] S. Arnott, A. Fulmer, W.E. Scott, I.C.M. Dea, R. Moorhouse, D.A. Rees, J. Mol. Biol. 90 (1974) 269–284.
[271] K.Y. Lee, D.J. Mooney, Prog. Polym. Sci. 37 (2012) 106–126.
[272] K.I. Draget, G. Skjåk-Bræk, O. Smidsrød, Int. J. Biol. Macromol. 21 (1997) 47–55.
[273] M. Di Rosa, J. Pharm. Pharmacol. 24 (1972) 89–102.
[274] M. Rinaudo, Polym. Int. 57 (2008) 397–430.
[275] J.A. Grover, Methylcellulose and Its Derivatives, Industrial Gums, third edition, Academic Press, London, 1993, pp. 475–504.
[276] T. Bourtoom, Int. Food Res. J. 15 (2008) 237–248.
[277] M.N.V.R. Kumar, R.A.A. Muzzarelli, C. Muzzarelli, H. Sashiwa, A.J. Domb, Chem. Rev. 104 (2004) 6017–6084.
[278] M. Rinaudo, Prog. Polym. Sci. 31 (2006) 603–632.
[279] A.N. De Belder, Dextran, Industrial Gums, third edition, Academic Press, London, 1993, pp. 399–425.
[280] M. Milas, X. Shi, M. Rinaudo, Biopolymers 30 (1990) 451–464.
[281] K.S. Kang, D.J. Pettitt, Xanthan, gellan, welan, and rhamsan, Industrial Gums, third edition, Academic Press, London, 1993, pp. 341–397.
[282] B.M. Yapo, Carbohydr. Polym. 86 (2011) 373–385.
[283] C. Rolin, Pectin, Industrial Gums, third edition, Academic Press, London, 1993, pp. 257–293.
[284] Y. Tsujisaka, M. Mitsuhashi, Pullulan, Industrial Gums, third edition, Academic Press, London, 1993, pp. 447–460.
[285] V.D. Prajapati, G.K. Jani, S.M. Khanda, Carbohydr. Polym. 95 (2013) 540–549.
[286] A.D. Baldwin, K.L. Kiick, Pept. Sci. 94 (2010) 128–140.
[287] F.N. Lamari, N.K. Karamanos, Structure of Chondroitin Sulfate, Advances in Pharmacology vol 53, Academic Press, 2006, pp. 33–48.
[288] J.M. Trowbridge, R.L. Gallo, Glycobiology 12 (2002) 117R–125R.
[289] N. Volpi, Carbohydr. Polym. 82 (2010) 233–239.
[290] D.L. Rabenstein, Nat. Prod. Rep. 19 (2002) 312–331.
[291] I. Capila, R.J. Linhardt, Angew. Chemie Int. Ed. 41 (2002) 390–412.
[292] B.F. Chong, L.M. Blank, R. McLaughlin, L.K. Nielsen, Appl. Microbiol. Biotechnol. 66 (2005) 341–351.
[293] J. Necas, L. Bartosikova, P. Brauner, J. Kolar, Vet. Med. 53 (2008) 397–411.
[294] J.L. Funderburgh, Glycobiology 10 (2000) 951–958.
[295] K. Meyer, A. Linker, E.A. Davidson, B. Weissmann, J. Biol. Chem. 205 (1953) 611.
[296] T. Peters Jr, The Albumin Molecule: Its Structure and Chemical Properties, All About Albumin, Academic Press, San Diego, 1995, pp. 9–11.
[297] T. Peters, Serum Albumin, in: C.B. Anfinsen, J.T. Edsall, F.M. Richards (Eds.), Advances in Protein Chemistry, vol 37, Academic Press, 1985, pp. 161–245.
[298] C. Holt, Structure and Stability of Bovine Casein Micelles, in: C.B. Anfinsen, F.M. Richards, J.T. Edsall, D.S. Eisenberg (Eds.), Advances in Protein Chemistry, vol 43, Academic Press, 1992, pp. 63–151.
[299] C.G. De Kruif, C. Holt, P.F. Fox, P.L.H. McSweeney (Eds.), Advanced Dairy Chemistry—1 Proteins: Part A / Part B, Springer US, Boston, MA, 2003, pp. 233–276.
[300] M.D. Shoulders, R.T. Raines, Annu. Rev. Biochem. 78 (2009) 929–958.
[301] P. Fratzl, P. Fratzl (Ed.), Collagen: Structure and Mechanics, Springer US, Boston, MA, 2008, pp. 1–13.
[302] L.B. Sandberg, N.T. Soskel, J.G. Leslie, N. Engl. J. Med. 304 (1981) 566–579.
[303] S.M. Mithieux, A.S. Weiss, Elastin, Advances in Protein Chemistry vol 70, Academic Press, 2005, pp. 437–461.
[304] M.W. Mosesson, J. Thromb. Haemost. 3 (2005) 1894–1904.
[305] R.F. Doolittle, Annu. Rev. Biochem. 53 (1984) 195–229.
[306] Gelatin, K. te Nijenhuis (Ed.), Thermoreversible Networks: Viscoelastic Properties and Structure of Gels, Springer, Berlin Heidelberg, Berlin, Heidelberg, 1997, pp. 160–193.
[307] G.F. Elliott, A.F. Huxley, T. Weis-Fogh, J. Mol. Biol. 13 (1965) 791-IN713.
[308] S.O. Andersen, T. Weis-Fogh, Resilin. A Rubberlike Protein in Arthropod Cuticle, in: J.W.L. Beament, J.E. Treherne, V.B. Wigglesworth (Eds.), Advances in Insect Physiology, vol 2, Academic Press, 1964, pp. 1–65.
[309] C. Vepari, D.L. Kaplan, Prog. Polym. Sci. 32 (2007) 991–1007.
[310] B. Kundu, R. Rajkhowa, S.C. Kundu, X. Wang, Adv. Drug Deliv. Rev. 65 (2013) 457–470.
[311] P. Zarrintaj, S. Manouchehri, Z. Ahmadi, M.R. Saeb, A.M. Urbanska, D.L. Kaplan, M. Mozafari, Carbohydr. Polym. 187 (2018) 66–84.
[312] A.P. Balgude, X. Yu, A. Szymanski, R.V. Bellamkonda, Biomaterials 22 (2001) 1077–1084.
[313] D. Velasco, E. Tumarkin, E. Kumacheva, Small 8 (2012) 1633–1642.
[314] V. Normand, D.L. Lootens, E. Amici, K.P. Plucknett, P. Aymard, Biomacromolecules 1 (2000) 730–738.
[315] J.-X. Hu, J.-B. Ran, S. Chen, P. Jiang, X.-Y. Shen, H. Tong, Biomacromolecules 17 (2016) 2437–2447.
[316] A. Forget, J. Christensen, S. Lüdeke, E. Kohler, S. Tobias, M. Matloubi, R. Thomann, V.P. Shastri, Proc. Natl. Acad. Sci. 110 (2013) 12887–12892.
[317] I.D. Paepe, H. Declercq, M. Cornelissen, E. Schacht, Polym. Int. 51 (2002) 867–870.
[318] Y. Luo, M.S. Shoichet, Nat. Mater. 3 (2004) 249–253.
[319] X. Yu, G.P. Dillon, R.V. Bellamkonda, Tissue Eng. 5 (1999) 291–304.
[320] Y. Luo, M.S. Shoichet, Biomacromolecules 5 (2004) 2315–2323.
[321] G.P. Dillon, Y. Xiaojun, A. Sridharan, J.P. Ranieri, R.V. Bellamkonda, J. Biomater. Sci. Polym. Ed. 9 (1998) 1049–1069.
[322] J.T. Connelly, A.J. García, M.E. Levenston, J. Cell. Physiol. 217 (2008) 145–154.
[323] E. Schuh, S. Hofmann, K. Stok, H. Notbohm, R. Müller, N. Rotter, J. Biomed. Mater. Res. A. 100A (2012) 38–47.
[324] R. Meena, K. Prasad, A.K. Siddhanta, J. Appl. Polym. Sci. 104 (2007) 290–296.
[325] Y. Ling, J. Rubin, Y. Deng, C. Huang, U. Demirci, J.M. Karp, A. Khademhosseini, Lab Chip 7 (2007) 756–762.
[326] W.H. Wong, D.J. Mooney, A. Atala, D.J. Mooney (Eds.), Synthetic Biodegradable Polymer Scaffolds, Birkhäuser Boston, Boston, MA, 1997, pp. 51–82.
[327] J.T. Oliveira, R.L. Reis, J. Tissue Eng. Regen. Med. 5 (2011) 421–436.
[328] N.C. Hunt, L.M. Grover, Biotechnol. Lett. 32 (2010) 733–742.
[329] H.A. Awad, M. Quinn Wickham, H.A. Leddy, J.M. Gimble, F. Guilak, Biomaterials 25 (2004) 3211–3222.
[330] R.L. Mauck, X. Yuan, R.S. Tuan, Osteoarthr. Cartil. 14 (2006) 179–189.
[331] M.D. Buschmann, Y.A. Gluzband, A.J. Grodzinsky, E.B. Hunziker, J. Cell. Sci. 108 (1995) 1497.
[332] R.L. Mauck, M.A. Soltz, C.C.B. Wang, D.D. Wong, P.-H.G. Chao, W.B. Valhmu, C.T. Hung, G.A. Ateshian, J. Biomech. Eng. 122 (2000) 252–260.
[333] M.D. Buschmann, Y.A. Gluzband, A.J. Grodzinsky, J.H. Kimura, E.B. Hunziker, J. Orthop. Res. 10 (1992) 745–758.
[334] D.A. Lee, D.L. Bader, J. Orthop. Res. 15 (1997) 181–188.
[335] B. Rahfoth, J. Weisser, F. Sternkopf, T. Aigner, K. von der Mark, R. Bräuer, Osteoarthr. Cartil. 6 (1998) 50–65.
[336] K.W. Ng, G.A. Ateshian, C.T. Hung, Tissue Eng. Part A 15 (2009) 2315–2324.
[337] M. Ahearne, Y. Yang, A.J. El Haj, K.Y. Then, K.-K. Liu, J. R. Soc. Interface 2 (2005) 455.
[338] J. Liu, L. Li, Eur. J. Pharm. Sci. 25 (2005) 237–244.
[339] N. Wang, X.S. Wu, Pharm. Dev. Technol. 2 (1997) 135–142.
[340] Y. Aizawa, R. Wylie, M. Shoichet, Adv. Mater. 22 (2010) 4831–4835.
[341] D. Zhang, D.B. Das, C.D. Rielly, J. Pharm. Sci. 103 (2014) 613–627.
[342] N.J. Meilander-Lin, P.J. Cheung, D.L. Wilson, R.V. Bellamkonda, Tissue Eng. 11 (2005) 546–555.
[343] N.J. Meilander, M.K. Pasumarthy, T.H. Kowalczyk, M.J. Cooper, R.V. Bellamkonda, J. Control. Release 88 (2003) 321–331.
[344] X. Guojie, Y. Fuqiang, W. Huayu, H. Xuefeng, Z. Li, Z. Jinming, J. Tissue Eng. 5 (2014) 2041731413520438.

[345] N. Mokarram, K. Dymanus, A. Srinivasan, J.G. Lyon, J. Tipton, J. Chu, A.W. English, R.V. Bellamkonda, Proc. Natl. Acad. Sci. 114 (2017) E5077–E5084.
[346] J.L. Drury, R.G. Dennis, D.J. Mooney, Biomaterials 25 (2004) 3187–3199.
[347] M. George, T.E. Abraham, J. Control. Release 114 (2006) 1–14.
[348] K.I. Draget, B.T. Stokke, Y. Yuguchi, H. Urakawa, K. Kajiwara, Biomacromolecules 4 (2003) 1661–1668.
[349] O. Smidsrød, G. Skjaˆk-Brˆk, Trends Biotechnol. 8 (1990) 71–78.
[350] Ý.A. Mørch, I. Donati, B.L. Strand, Biomacromolecules 7 (2006) 1471–1480.
[351] Y. Si, L. Wang, X. Wang, N. Tang, J. Yu, B. Ding, Adv. Mater. 29 (2017) 1700339.
[352] H.-J. Kong, K.Y. Lee, D.J. Mooney, Polymer 43 (2002) 6239–6246.
[353] Z.-Y. Wang, Q.-Z. Zhang, M. Konno, S. Saito, Biopolymers 34 (1994) 737–746.
[354] J.-Y. Sun, X. Zhao, W.R.K. Illeperuma, O. Chaudhuri, K.H. Oh, D.J. Mooney, J.J. Vlassak, Z. Suo, Nature 489 (2012) 133–136.
[355] H. Yi, S. Forsythe, Y. He, Q. Liu, X. Geng, S. Wei, G. Li, A. Atala, A. Skardal, Y. Zhang, Acta Biomater. 62 (2017) 222–233.
[356] K.H. Bouhadir, K.Y. Lee, E. Alsberg, K.L. Damm, K.W. Anderson, D.J. Mooney, Biotechnol. Prog. 17 (2001) 945–950.
[357] T. Boontheekul, H.-J. Kong, D.J. Mooney, Biomaterials 26 (2005) 2455–2465.
[358] S. Reakasame, A.R. Boccaccini, Biomacromolecules 19 (2018) 3–21.
[359] M.E. Klontzas, S. Reakasame, R. Silva, J.C.F. Morais, S. Vernardis, R.J. MacFarlane, M. Heliotis, E. Tsiridis, N. Panoskaltsis, A.R. Boccaccini, A. Mantalaris, Acta Biomater. 88 (2019) 224–240.
[360] H.J. Kong, J. Liu, K. Riddle, T. Matsumoto, K. Leach, D.J. Mooney, Nat. Mater. 4 (2005) 460–464.
[361] S. Kim, J.-M. Moon, J.S. Choi, W.K. Cho, S.M. Kang, Adv. Funct. Mater. 26 (2016) 4099–4105.
[362] C.J. Kastrup, M. Nahrendorf, J.L. Figueiredo, H. Lee, S. Kambhampati, T. Lee, S.-W. Cho, R. Gorbatov, Y. Iwamoto, T.T. Dang, P. Dutta, J.H. Yeon, H. Cheng, C.D. Pritchard, A.J. Vegas, C.D. Siegel, S. MacDougall, M. Okonkwo, A. Thai, J.R. Stone, A.J. Coury, R. Weissleder, R. Langer, D.G. Anderson, Proc. Natl. Acad. Sci. 109 (2012) 21444–21449.
[363] C. Lee, J. Shin, J.S. Lee, E. Byun, J.H. Ryu, S.H. Um, D.-I. Kim, H. Lee, S.-W. Cho, Biomacromolecules 14 (2013) 2004–2013.
[364] S.H. Hong, M. Shin, J. Lee, J.H. Ryu, S. Lee, J.W. Yang, W.D. Kim, H. Lee, Adv. Healthc. Mater. 5 (2016) 75–79.
[365] N. Huebsch, P.R. Arany, A.S. Mao, D. Shvartsman, O.A. Ali, S.A. Bencherif, J. Rivera-Feliciano, D.J. Mooney, Nat. Mater. 9 (2010) 518–526.
[366] R.M. Desai, S.T. Koshy, S.A. Hilderbrand, D.J. Mooney, N.S. Joshi, Biomaterials 50 (2015) 30–37.
[367] E.J. De Leon-Peralta, Y. Brudno, B.J. Kwee, D.J. Mooney, Faseb J. 30 (2016) 1177.1110.
[368] I. Freeman, A. Kedem, S. Cohen, Biomaterials 29 (2008) 3260–3268.
[369] G.T. Grant, E.R. Morris, D.A. Rees, P.J.C. Smith, D. Thom, FEBS Lett. 32 (1973) 195–198.
[370] C.K. Kuo, P.X. Ma, Biomaterials 22 (2001) 511–521.
[371] X. Zhao, N. Huebsch, D.J. Mooney, Z. Suo, J. Appl. Phys. 107 (2010) 063509.
[372] P. Eiselt, K.Y. Lee, D.J. Mooney, Macromolecules 32 (1999) 5561–5566.
[373] K.Y. Lee, J.A. Rowley, P. Eiselt, E.M. Moy, K.H. Bouhadir, D.J. Mooney, Macromolecules 33 (2000) 4291–4294.
[374] K.A. Smeds, M.W. Grinstaff, J. Biomed. Mater. Res. 54 (2001) 115–121.
[375] M.Z. Lu, H.L. Lan, F.F. Wang, S.J. Chang, Y.J. Wang, Biotechnol. Bioeng. 70 (2000) 479–483.
[376] S. Zhao, M. Cao, H. Li, L. Li, W. Xu, Carbohydr. Res. 345 (2010) 425–431.
[377] K.Y. Lee, H.J. Kong, R.G. Larson, D.J. Mooney, Adv. Mater. 15 (2003) 1828–1832.
[378] J. Venkatesan, I. Bhatnagar, P. Manivasagan, K.-H. Kang, S.-K. Kim, Int. J. Biol. Macromol. 72 (2015) 269–281.
[379] T. Agarwal, P. Kabiraj, G.H. Narayana, S. Kulanthaivel, U. Kasiviswanathan, K. Pal, S. Giri, T.K. Maiti, I. Banerjee, ACS Appl. Mater. Interfaces 8 (2016) 32132–32145.
[380] E. Alsberg, K.W. Anderson, A. Albeiruti, R.T. Franceschi, D.J. Mooney, J. Dent. Res. 80 (2001) 2025–2029.
[381] A. Moshaverinia, S. Ansari, C. Chen, X. Xu, K. Akiyama, M.L. Snead, H.H. Zadeh, S. Shi, Biomaterials 34 (2013) 6572–6579.
[382] Y.-S. Hwang, J. Cho, F. Tay, J.Y.Y. Heng, R. Ho, S.G. Kazarian, D.R. Williams, A.R. Boccaccini, J.M. Polak, A. Mantalaris, Biomaterials 30 (2009) 499–507.
[383] H.J. Hauselmann, R.J. Fernandes, S.S. Mok, T.M. Schmid, J.A. Block, M.B. Aydelotte, K.E. Kuettner, E.J. Thonar, J. Cell. Sci. 107 (1994) 17.
[384] H.K. Heywood, P.K. Sembi, D.A. Lee, D.L. Bader, Tissue Eng. 10 (2004) 1467–1479.
[385] Z. Xu, M.T. Lam, B.H.A. Rehm, M.F. Moradali (Eds.), Alginates and Their Biomedical Applications, Springer Singapore, Singapore, 2018, pp. 185–212.
[386] E. Ruvinov, S. Cohen, Adv. Drug Deliv. Rev. 96 (2016) 54–76.
[387] M.I. Günther, N. Weidner, R. Müller, A. Blesch, Acta Biomater. 27 (2015) 140–150.
[388] K. Pawar, P. Prang, R. Müller, M. Caioni, U. Bogdahn, W. Kunz, N. Weidner, Acta Biomater. 27 (2015) 131–139.
[389] A. Bozza, E.E. Coates, T. Incitti, K.M. Ferlin, A. Messina, E. Menna, Y. Bozzi, J.P. Fisher, S. Casarosa, Biomaterials 35 (2014) 4636–4645.
[390] A. Martinsen, G. Skjåk-Bræk, O. Smidsrød, Biotechnol. Bioeng. 33 (1989) 79–89.
[391] M. Kierstan, C. Bucke, Biotechnol. Bioeng. 19 (1977) 387–397.
[392] D. Pipeleers, B. Keymeulen, Trends Endocrinol. Metab. 27 (2016) 247–248.
[393] B.J. Kim, T. Park, H.C. Moon, S.-Y. Park, D. Hong, E.H. Ko, J.Y. Kim, J.W. Hong, S.W. Han, Y.-G. Kim, I.S. Choi, Angew. Chemie Int. Ed. 53 (2014) 14443–14446.
[394] S. Utech, R. Prodanovic, A.S. Mao, R. Ostafe, D.J. Mooney, D.A. Weitz, Adv. Healthc. Mater. 4 (2015) 1628–1633.
[395] M. Clark, B.H.A. Rehm, M.F. Moradali (Eds.), Alginates and Their Biomedical Applications, Springer, Singapore, Singapore, 2018, pp. 213–222.
[396] H.H. Tønnesen, J. Karlsen, Drug Dev. Ind. Pharm. 28 (2002) 621–630.
[397] K.Y. Lee, M.C. Peters, D.J. Mooney, Adv. Mater. 13 (2001) 837–839.
[398] N. Gogoi, D. Chowdhury, J. Mater. Chem. B 2 (2014) 4089–4099.
[399] S.S. Ho, N.L. Vollmer, M.I. Refaat, O. Jeon, E. Alsberg, M.A. Lee, J.K. Leach, Adv. Healthc. Mater. 5 (2016) 2501–2509.
[400] W.R. Gombotz, S.F. Wee, Adv. Drug Deliv. Rev. 64 (2012) 194–205.
[401] C. Wu, W. Fan, M. Gelinsky, Y. Xiao, J. Chang, T. Friis, G. Cuniberti, J. R. Soc. Interface 8 (2011) 1804.
[402] K.Y. Lee, M.C. Peters, K.W. Anderson, D.J. Mooney, Nature 408 (2000) 998–1000.
[403] O. Guillaume, A. Daly, K. Lennon, J. Gansau, S.F. Buckley, C.T. Buckley, Acta Biomater. 10 (2014) 1985–1995.
[404] S. Orr, I. Strominger, E. Eremenko, E. Vinogradov, E. Ruvinov, A. Monsonego, S. Cohen, Acta Biomater. 45 (2016) 196–209.
[405] S. Jose, M.L. Hughbanks, B.Y.K. Binder, G.C. Ingavle, J.K. Leach, Acta Biomater. 10 (2014) 1955–1964.
[406] S.J. Bidarra, C.C. Barrias, P.L. Granja, Acta Biomater. 10 (2014) 1646–1662.
[407] Y. Wang, J. Zhou, L. Qiu, X. Wang, L. Chen, T. Liu, W. Di, Biomaterials 35 (2014) 4297–4309.
[408] A. Khurana, C. Godugu, B.H.A. Rehm, M.F. Moradali (Eds.), Alginates and Their Biomedical Applications, Springer, Singapore, Singapore, 2018, pp. 157–183.
[409] C. Fischbach, H.J. Kong, S.X. Hsiong, M.B. Evangelista, W. Yuen, D.J. Mooney, Proc. Natl. Acad. Sci. 106 (2009) 399–404.
[410] A.J. Vegas, O. Veiseh, M. Gürtler, J.R. Millman, F.W. Pagliuca, A.R. Bader, J.C. Doloff, J. Li, M. Chen, K. Olejnik, H.H. Tam, S. Jhunjhunwala, E. Langan, S. Aresta-Dasilva, S. Gandham, J.J. McGarrigle, M.A. Bochenek, J. Hollister-Lock, J. Oberholzer, D.L. Greiner, G.C. Weir, D.A. Melton, R. Langer, D.G. Anderson, Nat. Med. 22 (2016) 306–311.
[411] A.J. Vegas, O. Veiseh, J.C. Doloff, M. Ma, H.H. Tam, K. Bratlie, J. Li, A.R. Bader, E. Langan, K. Olejnik, P. Fenton, J.W. Kang, J. Hollister-Locke, M.A. Bochenek, A. Chiu, S. Siebert, K. Tang, S. Jhunjhunwala, S. Aresta-Dasilva, N. Dholakia, R. Thakrar, T. Vietti, M. Chen, J. Cohen, K. Siniakowicz, M. Qi, J. McGarrigle, A.C. Graham, S. Lyle, D.M. Harlan, D.L. Greiner, J. Oberholzer, G.C. Weir, R. Langer, D.G. Anderson, Nat. Biotechnol. 34 (2016) 345–352.
[412] M. Leijs, E. Villafuertes, J. Haeck, W. Koevoet, B. Fernandez-Gutierrez, M. Hoogduijn, J. Verhaar, M. Bernsen, G. van Buul, G. van Osch, Eur. Cell. Mater. 33 (2017) 43.
[413] S. Jain, T.-H. Tran, M. Amiji, Biomaterials 61 (2015) 162–177.
[414] V.L. Campo, D.F. Kawano, D.Bd. Silva, I. Carvalho, Carbohydr. Polym. 77 (2009) 167–180.
[415] G.H. Therkelsen, Carrageenan, Industrial Gums, third edition, Academic Press, London, 1993, pp. 145–180.
[416] C. Rochas, M. Rinaudo, S. Landry, Carbohydr. Polym. 10 (1989) 115–127.
[417] C. Rochas, M. Rinaudo, S. Landry, Carbohydr. Polym. 12 (1990) 255–266.
[418] M. Watase, K. Nishinari, Rheol. Acta 21 (1982) 318–324.
[419] M.R. Mangione, D. Giacomazza, D. Bulone, V. Martorana, G. Cavallaro, P.L. San Biagio, Biophys. Chem. 113 (2005) 129–135.
[420] I.S. Chronakis, J.-L. Doublier, L. Piculell, Int. J. Biol. Macromol. 28 (2000) 1–14.
[421] K.S. Hossain, K. Miyanaga, H. Maeda, N. Nemoto, Biomacromolecules 2 (2001) 442–449.
[422] D.A. Rees, Structure, Conformation, and Mechanism in the Formation of Polysaccharide Gels and Networks, in: M.L. Wolfrom, R.S. Tipson, D. Horton (Eds.), Advances in Carbohydrate Chemistry and Biochemistry, vol 24, Academic Press, 1969, pp. 267–332.
[423] S. Liu, L. Li, Food Hydrocoll. 61 (2016) 793–800.
[424] S.M. Mihaila, A.K. Gaharwar, R.L. Reis, A.P. Marques, M.E. Gomes, A. Khademhosseini, Adv. Healthc. Mater. 2 (2013) 895–907.
[425] M. Ciancia, M.D. Noseda, M.C. Matulewicz, A.S. Cerezo, Carbohydr. Polym. 20 (1993) 95–98.
[426] A.G. Viana, M.D. Noseda, M.E.R. Duarte, A.S. Cerezo, Carbohydr. Polym. 58 (2004) 455–460.
[427] D.A. Navarro, C.A. Stortz, Carbohydr. Polym. 62 (2005) 187–191.
[428] H. Yuan, W. Zhang, X. Li, X. Lü, N. Li, X. Gao, J. Song, Carbohydr. Res. 340 (2005) 685–692.
[429] H. Yuan, J. Song, W. Zhang, X. Li, N. Li, X. Gao, Bioorg. Med. Chem. Lett. 16 (2006) 1329–1334.
[430] T. Yamada, A. Ogamo, T. Saito, H. Uchiyama, Y. Nakagawa, Carbohydr. Polym. 41 (2000) 115–120.
[431] Y.-P. Jiang, X.-K. Guo, Carbohydr. Polym. 61 (2005) 441–445.
[432] M. Haijin, J. Xiaolu, G. Huashi, J. Appl. Phycol. 15 (2003) 297–303.
[433] F.C. Recuenco, K. Kobayashi, A. Ishiwa, Y. Enomoto-Rogers, N.G.V. Fundador, T. Sugi, H. Takemae, T. Iwanaga, F. Murakoshi, H. Gong, A. Inomata, T. Horimoto, T. Iwata, K. Kato, Sci. Rep. 4 (2014) 4723.
[434] R. Meena, K. Prasad, A.K. Siddhanta, Int. J. Biol. Macromol. 41 (2007) 94–101.
[435] C. Rochas, M. Rinaudo, Biopolymers 23 (1984) 735–745.
[436] I.Y. Kim, R. Iwatsuki, K. Kikuta, Y. Morita, T. Miyazaki, C. Ohtsuki, Mater. Sci. Eng. C 31 (2011) 1472–1476.
[437] M.R. Mangione, D. Giacomazza, D. Bulone, V. Martorana, P.L. San Biagio, Biophys. Chem. 104 (2003) 95–105.
[438] E.G. Popa, P.P. Carvalho, A.F. Dias, T.C. Santos, V.E. Santo, A.P. Marques, C.A. Viegas, I.R. Dias, M.E. Gomes, R.L. Reis, J. Biomed. Mater. Res. A. 102 (2014) 4087–4097.
[439] A.W. Charles, A.R. Edwin, W.N. George, Proc. Soc. Exp. Biol. Med. 111 (1962) 544–547.
[440] C. Rochas, A. Heyraud, Polym. Bull. 5 (1981) 81–86.
[441] V.E. Santo, A.M. Frias, M. Carida, R. Cancedda, M.E. Gomes, J.F. Mano, R.L. Reis,

Biomacromolecules 10 (2009) 1392–1401.
[442] E. Popa, R. Reis, M. Gomes, Biotechnol. Appl. Biochem. 59 (2012) 132–141.
[443] E.G. Popa, S.G. Caridade, J.F. Mano, R.L. Reis, M.E. Gomes, J. Tissue Eng. Regen. Med. 9 (2015) 550–563.
[444] M.R. Pedro, E.S. Vítor, E.G. Manuela, L.R. Rui, F.M. João, J. Bioact. Compat. Polym. 26 (2011) 493–507.
[445] L. Li, R. Ni, Y. Shao, S. Mao, Carbohydr. Polym. 103 (2014) 1–11.
[446] D. Guzman-Villanueva, I.M. El-Sherbiny, D. Herrera-Ruiz, H.D.C. Smyth, Biomed Res. Int. 2013 (2013) 724763.
[447] K. Makino, R. Idenuma, T. Murakami, H. Ohshima, Colloids Surf. B Biointerfaces 20 (2001) 355–359.
[448] C. Valenta, K. Schultz, J. Control. Release 95 (2004) 257–265.
[449] T. Coviello, P. Matricardi, C. Marianecci, F. Alhaique, J. Control. Release 119 (2007) 5–24.
[450] J. Liu, X. Zhan, J. Wan, Y. Wang, C. Wang, Carbohydr. Polym. 121 (2015) 27–36.
[451] A.L. Daniel-da-Silva, L. Ferreira, A.M. Gil, T. Trindade, J. Colloid Interface Sci. 355 (2011) 512–517.
[452] D. Klemm, F. Kramer, S. Moritz, T. Lindström, M. Ankerfors, D. Gray, A. Dorris, Angew. Chemie Int. Ed. 50 (2011) 5438–5466.
[453] M.N. Belgacem, A. Gandini, Production, Chemistry and Properties of Cellulose-based Materials, Biopolymers – New Materials for Sustainable Films and Coatings, John Wiley & Sons, Ltd, 2011, pp. 151–178.
[454] M. Bhattacharya, M.M. Malinen, P. Lauren, Y.-R. Lou, S.W. Kuisma, L. Kanninen, M. Lille, A. Corlu, C. GuGuen-Guillouzo, O. Ikkala, A. Laukkanen, A. Urtti, M. Yliperttula, J. Control. Release 164 (2012) 291–298.
[455] K.J. Edgar, C.M. Buchanan, J.S. Debenham, P.A. Rundquist, B.D. Seiler, M.C. Shelton, D. Tindall, Prog. Polym. Sci. 26 (2001) 1605–1688.
[456] A. Sannino, C. Demitri, M. Madaghiele, Materials 2 (2009) 353–373.
[457] C. Chang, L. Zhang, Carbohydr. Polym. 84 (2011) 40–53.
[458] J.R. Lott, J.W. McAllister, S.A. Arvidson, F.S. Bates, T.P. Lodge, Biomacromolecules 14 (2013) 2484–2488.
[459] S. Thirumala, J. Gimble, R. Devireddy, Cells 2 (2013) 460.
[460] L.E. Bromberg, E.S. Ron, Adv. Drug Deliv. Rev. 31 (1998) 197–221.
[461] Y. Xu, L. Li, P. Zheng, Y.C. Lam, X. Hu, Langmuir 20 (2004) 6134–6138.
[462] S.C. Joshi, Y.C. Lam, J. Appl. Polym. Sci. 101 (2006) 1620–1629.
[463] P. Zheng, L. Li, X. Hu, X. Zhao, J. Polym. Sci. Part B: Polym. Phys. 42 (2004) 1849–1860.
[464] E. Heymann, Transactions of the Faraday Society 31 (1935) 846–864.
[465] M.K. Bain, B. Bhowmick, D. Maity, D. Mondal, M.M.R. Mollick, D. Rana, D. Chattopadhyay, Int. J. Biol. Macromol. 51 (2012) 831–836.
[466] C.-C. Huang, Z.-X. Liao, D.-Y. Chen, C.-W. Hsiao, Y. Chang, H.-W. Sung, Adv. Healthc. Mater. 3 (2014) 1133–1148.
[467] C.-H. Chen, C.-C. Tsai, W. Chen, F.-L. Mi, H.-F. Liang, S.-C. Chen, H.-W. Sung, Biomacromolecules 7 (2006) 736–743.
[468] M.C. Tate, D.A. Shear, S.W. Hoffman, D.G. Stein, M.C. LaPlaca, Biomaterials 22 (2001) 1113–1123.
[469] M.-J. Yang, C.-H. Chen, P.-J. Lin, C.-H. Huang, W. Chen, H.-W. Sung, Biomacromolecules 8 (2007) 2746–2752.
[470] T. Saito, S. Kimura, Y. Nishiyama, A. Isogai, Biomacromolecules 8 (2007) 2485–2491.
[471] H. Valo, S. Arola, P. Laaksonen, M. Torkkeli, L. Peltonen, M.B. Linder, R. Serimaa, S. Kuga, J. Hirvonen, T. Laaksonen, Eur. J. Pharm. Sci. 50 (2013) 69–77.
[472] S.S. Stalling, S.O. Akintoye, S.B. Nicoll, Acta Biomater. 5 (2009) 1911–1918.
[473] L. Li, P. Thangamathesvaran, C. Yue, K. Tam, X. Hu, Y. Lam, Langmuir 17 (2001) 8062–8068.
[474] G. Marci, G. Mele, L. Palmisano, P. Pulito, A. Sannino, Green Chem. 8 (2006) 439–444.
[475] A. Sannino, A. Esposito, A.D. Rosa, A. Cozzolino, L. Ambrosio, L. Nicolais, J. Biomed. Mater. Res. A. 67A (2003) 1016–1024.
[476] A. Sannino, M. Madaghiele, M.G. Lionetto, T. Schettino, A. Maffezzoli, J. Appl. Polym. Sci. 102 (2006) 1524–1530.
[477] C. Demitri, R. Del Sole, F. Scalera, A. Sannino, G. Vasapollo, A. Maffezzoli, L. Ambrosio, L. Nicolais, J. Appl. Polym. Sci. 110 (2008) 2453–2460.
[478] J.M. Rosiak, P. Ulański, Radiat. Phys. Chem. 55 (1999) 139–151.
[479] B. Fei, R.A. Wach, H. Mitomo, F. Yoshii, T. Kume, J. Appl. Polym. Sci. 78 (2000) 278–283.
[480] K. Kobayashi, C.-i. Huang, T.P. Lodge, Macromolecules 32 (1999) 7070–7077.
[481] M. Takahashi, M. Shimazaki, J. Yamamoto, J. Polym. Sci. Part B: Polym. Phys. 39 (2001) 91–100.
[482] J. Andersson, H. Stenhamre, H. Bäckdahl, P. Gatenholm, J. Biomed. Mater. Res. A. 94A (2010) 1124–1132.
[483] C. Vinatier, D. Magne, A. Moreau, O. Gauthier, O. Malard, C. Vignes-Colombeix, G. Daculsi, P. Weiss, J. Guicheux, J. Biomed. Mater. Res. A. 80A (2007) 66–74.
[484] C. Vinatier, D. Magne, P. Weiss, C. Trojani, N. Rochet, G.F. Carle, C. Vignes-Colombeix, C. Chadjichristos, P. Galera, G. Daculsi, J. Guicheux, Biomaterials 26 (2005) 6643–6651.
[485] M.P. Malgosia, G.B. Brian, S.S. Molly, Biomed. Mater. 7 (2012) 024101.
[486] S.E. Stabenfeldt, A.J. García, M.C. LaPlaca, J. Biomed. Mater. Res. A. 77A (2006) 718–725.
[487] S.E. Stabenfeldt, M.C. LaPlaca, Acta Biomater. 7 (2011) 4102–4108.
[488] W.-Y. Lee, Y.-H. Chang, Y.-C. Yeh, C.-H. Chen, K.M. Lin, C.-C. Huang, Y. Chang, H.-W. Sung, Biomaterials 30 (2009) 5505–5513.
[489] M.M. Pakulska, K. Vulic, R.Y. Tam, M.S. Shoichet, Adv. Mater. 27 (2015) 5002–5008.
[490] J. Siepmann, N.A. Peppas, Adv. Drug Deliv. Rev. 48 (2001) 139–157.
[491] M.M. Pakulska, K. Vulic, M.S. Shoichet, J. Control. Release 171 (2013) 11–16.
[492] C.K.S. Pillai, W. Paul, C.P. Sharma, Prog. Polym. Sci. 34 (2009) 641–678.
[493] I.-Y. Kim, S.-J. Seo, H.-S. Moon, M.-K. Yoo, I.-Y. Park, B.-C. Kim, C.-S. Cho, Biotechnol. Adv. 26 (2008) 1–21.
[494] R. Riva, H. Ragelle, A. des Rieux, N. Duhem, C. Jérôme, V. Préat, Chitosan and Chitosan Derivatives in Drug Delivery and Tissue Engineering, in: R. Jayakumar, M. Prabaharan, R.A.A. Muzzarelli (Eds.), Chitosan for Biomaterials II, vol. 244, Springer, Berlin Heidelberg, Berlin, Heidelberg, 2011, pp. 19–44.
[495] H. Sashiwa, S.-i. Aiba, Prog. Polym. Sci. 29 (2004) 887–908.
[496] T.K. Giri, A. Thakur, A. Alexander, Ajazuddin, H. Badwaik, D.K. Tripathi, Acta Pharm. Sin. B 2 (2012) 439–449.
[497] D. Depan, R.P. Singh, Surface Modification of Chitosan and Its Implications in Tissue Engineering and Drug Delivery, Surface Modification of Biopolymers, John Wiley & Sons, Inc, 2015, pp. 20–44.
[498] E. Guibal, Sep. Purif. Technol. 38 (2004) 43–74.
[499] A.J. Varma, S.V. Deshpande, J.F. Kennedy, Carbohydr. Polym. 55 (2004) 77–93.
[500] T. Chandy, C.P. Sharma, Biomater. Artif. Cells Artif. Organs 18 (1990) 1–24.
[501] M. Dash, F. Chiellini, R.M. Ottenbrite, E. Chiellini, Prog. Polym. Sci. 36 (2011) 981–1014.
[502] I.M. van der Lubben, J.C. Verhoef, G. Borchard, H.E. Junginger, Eur. J. Pharm. Sci. 14 (2001) 201–207.
[503] E.I. Rabea, M.E.T. Badawy, C.V. Stevens, G. Smagghe, W. Steurbaut, Biomacromolecules 4 (2003) 1457–1465.
[504] E. Khor, L.Y. Lim, Biomaterials 24 (2003) 2339–2349.
[505] S.V. Madihally, H.W.T. Matthew, Biomaterials 20 (1999) 1133–1142.
[506] M.N.V. Ravi Kumar, React. Funct. Polym. 46 (2000) 1–27.
[507] J. Berger, M. Reist, J.M. Mayer, O. Felt, N.A. Peppas, R. Gurny, Eur. J. Pharm. Biopharm. 57 (2004) 19–34.
[508] J. Berger, M. Reist, J.M. Mayer, O. Felt, R. Gurny, Eur. J. Pharm. Biopharm. 57 (2004) 35–52.
[509] M.M. Thanou, A.F. Kotzé, T. Scharringhausen, H.L. Lueßen, A.G. de Boer, J.C. Verhoef, H.E. Junginger, J. Control. Release 64 (2000) 15–25.
[510] A.B. Sieval, M. Thanou, A.F. Kotze´, J.C. Verhoef, J. Brussee, H.E. Junginger, Carbohydr. Polym. 36 (1998) 157–165.
[511] K. Aiedeh, M.O. Taha, Eur. J. Pharm. Sci. 13 (2001) 159–168.
[512] C.E. Kast, A. Bernkop-Schnürch, Biomaterials 22 (2001) 2345–2352.
[513] P. Esmaeilzadeh, A. Köwitsch, A. Liedmann, M. Menzel, B. Fuhrmann, G. Schmidt, J. Klehm, T. Groth, ACS Appl. Mater. Interfaces 10 (2018) 8507–8518.
[514] P. Esmaeilzadeh, A. Köwitsch, F. Heyroth, G. Schmidt, S. Fischer, K. Richter, T. Groth, Carbohydr. Polym. 157 (2017) 1205–1214.
[515] M.-H. Ho, D.-M. Wang, H.-J. Hsieh, H.-C. Liu, T.-Y. Hsien, J.-Y. Lai, L.-T. Hou, Biomaterials 26 (2005) 3197–3206.
[516] K. Ono, Y. Saito, H. Yura, K. Ishikawa, A. Kurita, T. Akaike, M. Ishihara, J. Biomed. Mater. Res. 49 (2000) 289–295.
[517] M. Ishihara, K. Nakanishi, K. Ono, M. Sato, M. Kikuchi, Y. Saito, H. Yura, T. Matsui, H. Hattori, M. Uenoyama, A. Kurita, Biomaterials 23 (2002) 833–840.
[518] Y. Yeo, J.A. Burdick, C.B. Highley, R. Marini, R. Langer, D.S. Kohane, J. Biomed. Mater. Res. A. 78A (2006) 668–675.
[519] M. Morimoto, H. Saimoto, H. Usui, Y. Okamoto, S. Minami, Y. Shigemasa, Biomacromolecules 2 (2001) 1133–1136.
[520] M. Yalpani, L.D. Hall, Macromolecules 17 (1984) 272–281.
[521] I.-K. Park, J. Yang, H.-J. Jeong, H.-S. Bom, I. Harada, T. Akaike, S.-I. Kim, C.-S. Cho, Biomaterials 24 (2003) 2331–2337.
[522] K. Kim, K. Kim, J.H. Ryu, H. Lee, Biomaterials 52 (2015) 161–170.
[523] X. Zhao, P. Li, B. Guo, P.X. Ma, Acta Biomater. 26 (2015) 236–248.
[524] S. Zhang, J. Chen, Y. Yu, K. Dai, J. Wang, C. Liu, ACS Biomater. Sci. Eng. 5 (2019) 1944–1955.
[525] C. Wang, Y. Yu, H. Chen, S. Zhang, J. Wang, C. Liu, J. Mater. Chem. B 7 (2019) 1882–1892.
[526] S.M. Richardson, N. Hughes, J.A. Hunt, A.J. Freemont, J.A. Hoyland, Biomaterials 29 (2008) 85–93.
[527] J. Cho, M.-C. Heuzey, A. Bégin, P.J. Carreau, Biomacromolecules 6 (2005) 3267–3275.
[528] E. Ruel-Gariépy, M. Shive, A. Bichara, M. Berrada, D. Le Garrec, A. Chenite, J.-C. Leroux, Eur. J. Pharm. Biopharm. 57 (2004) 53–63.
[529] F.-L. Mi, S.-S. Shyu, T.-B. Wong, S.-F. Jang, S.-T. Lee, K.-T. Lu, J. Appl. Polym. Sci. 74 (1999) 1093–1107.
[530] A. Ghanem, D. Skonberg, J. Appl. Polym. Sci. 84 (2002) 405–413.
[531] Z.-W. Jing, Y.-Y. Jia, N. Wan, M. Luo, M.-L. Huan, T.-B. Kang, S.-Y. Zhou, B.-L. Zhang, Biomaterials 84 (2016) 276–285.
[532] E. Mirzaei, B.A. Ramazani, S.A.M. Shafiee, M. Danaei, International Journal of Polymeric Materials and Polymeric Biomaterials 62 (2013) 605–611.
[533] T.B. Fideles, J.L. Santos, H. Tomás, G.T. Furtado, D.B. Lima, S.M. Borges, M.V. Fook, Open Access Library Journal 5 (2018) 1–13.
[534] Y. Hong, H. Song, Y. Gong, Z. Mao, C. Gao, J. Shen, Acta Biomater. 3 (2007) 23–31.
[535] Y. Hong, Z. Mao, H. Wang, C. Gao, J. Shen, J. Biomed. Mater. Res. A. 79A (2006) 913–922.
[536] Z.M. Wu, X.G. Zhang, C. Zheng, C.X. Li, S.M. Zhang, R.N. Dong, D.M. Yu, Eur. J. Pharm. Sci. 37 (2009) 198–206.
[537] J. Xu, S. Strandman, J.X.X. Zhu, J. Barralet, M. Cerruti, Biomaterials 37 (2015) 395–404.
[538] N. Bhattarai, H.R. Ramay, J. Gunn, F.A. Matsen, M. Zhang, J. Control. Release 103 (2005) 609–624.
[539] F.-C. Chang, S.L. Levengood, N. Cho, L. Chen, E. Wang, J.S. Yu, M. Zhang, Adv. Ther. 1 (2018) 1800058.
[540] W.S. Wan Ngah, C.S. Endud, R. Mayanar, React. Funct. Polym. 50 (2002)

181–190.
[541] N. Li, R. Bai, Ind. Eng. Chem. Res. 44 (2005) 6692–6700.
[542] R. De Souza, P. Zahedi, C.J. Allen, M. Piquette-Miller, Biomaterials 30 (2009) 3818–3824.
[543] F.-L. Mi, Y.-C. Tan, H.-F. Liang, H.-W. Sung, Biomaterials 23 (2002) 181–191.
[544] M. Kong, X.G. Chen, K. Xing, H.J. Park, Int. J. Food Microbiol. 144 (2010) 51–63.
[545] H.K. No, N. Young Park, S. Ho Lee, S.P. Meyers, Int. J. Food Microbiol. 74 (2002) 65–72.
[546] X. Fei Liu, Y. Lin Guan, D. Zhi Yang, Z. Li, K. De Yao, J. Appl. Polym. Sci. 79 (2001) 1324–1335.
[547] Y.-L. Chiu, S.-C. Chen, C.-J. Su, C.-W. Hsiao, Y.-M. Chen, H.-L. Chen, H.-W. Sung, Biomaterials 30 (2009) 4877–4888.
[548] C.-M. Lehr, J.A. Bouwstra, E.H. Schacht, H.E. Junginger, Int. J. Pharm. 78 (1992) 43–48.
[549] K.Y. Lee, W.S. Ha, W.H. Park, Biomaterials 16 (1995) 1211–1216.
[550] A. Di Martino, M. Sittinger, M.V. Risbud, Biomaterials 26 (2005) 5983–5990.
[551] L. Cao, J.A. Werkmeister, J. Wang, V. Glattauer, K.M. McLean, C. Liu, Biomaterials 35 (2014) 2730–2742.
[552] J.K. Francis Suh, H.W.T. Matthew, Biomaterials 21 (2000) 2589–2598.
[553] T. Hao, N. Wen, J.K. Cao, H.B. Wang, S.H. Lü, T. Liu, Q.X. Lin, C.M. Duan, C.Y. Wang, Osteoarthr. Cartil. 18 (2010) 257–265.
[554] A. Chenite, C. Chaput, D. Wang, C. Combes, M.D. Buschmann, C.D. Hoemann, J.C. Leroux, B.L. Atkinson, F. Binette, A. Selmani, Biomaterials 21 (2000) 2155–2161.
[555] M. Ishihara, K. Obara, T. Ishizuka, M. Fujita, M. Sato, K. Masuoka, Y. Saito, H. Yura, T. Matsui, H. Hattori, M. Kikuchi, A. Kurita, J. Biomed. Mater. Res. A. 64A (2003) 551–559.
[556] S. Lee, C.M. Valmikinathan, J. Byun, S. Kim, G. Lee, N. Mokarram, S.B. Pai, E. Um, R.V. Bellamkonda, Y.-s. Yoon, Biomaterials 63 (2015) 158–167.
[557] C. Meyer, L. Stenberg, F. Gonzalez-Perez, S. Wrobel, G. Ronchi, E. Udina, S. Suganuma, S. Geuna, X. Navarro, L.B. Dahlin, C. Grothe, K. Haastert-Talini, Biomaterials 76 (2016) 33–51.
[558] N.D. Leipzig, R.G. Wylie, H. Kim, M.S. Shoichet, Biomaterials 32 (2011) 57–64.
[559] W.-N. Lu, S.-H. Lü, H.-B. Wang, D.-X. Li, C.-M. Duan, Z.-Q. Liu, T. Hao, W.-J. He, B. Xu, Q. Fu, Y.C. Song, X.-H. Xie, C.-Y. Wang, Tissue Eng. Part A 15 (2008) 1437–1447.
[560] H. Wang, X. Zhang, Y. Li, Y. Ma, Y. Zhang, Z. Liu, J. Zhou, Q. Lin, Y. Wang, C. Duan, C. Wang, J. Heart Lung Transplant. 29 (2010) 881–887.
[561] Z. Liu, H. Wang, Y. Wang, Q. Lin, A. Yao, F. Cao, D. Li, J. Zhou, C. Duan, Z. Du, Y. Wang, C. Wang, Biomaterials 33 (2012) 3093–3106.
[562] Y. Yeo, W. Geng, T. Ito, D.S. Kohane, J.A. Burdick, M. Radisic, J. Biomed. Mater. Res. Part B Appl. Biomater. 81B (2007) 312–322.
[563] D. An, Y. Ji, A. Chiu, Y.-C. Lu, W. Song, L. Zhai, L. Qi, D. Luo, M. Ma, Biomaterials 37 (2015) 40–48.
[564] N. Bhattarai, J. Gunn, M. Zhang, Adv. Drug Deliv. Rev. 62 (2010) 83–99.
[565] T.-Y. Liu, S.-Y. Chen, Y.-L. Lin, D.-M. Liu, Langmuir 22 (2006) 9740–9745.
[566] J.-i. Murata, Y. Ohya, T. Ouchi, Carbohydr. Polym. 32 (1997) 105–109.
[567] M. Thanou, B.I. Florea, M. Geldof, H.E. Junginger, G. Borchard, Biomaterials 23 (2002) 153–159.
[568] R.M. Raftery, I.M. Castaño, G. Chen, B. Cavanagh, B. Quinn, C.M. Curtin, S.A. Cryan, F.J. O'Brien, Biomaterials 149 (2017) 116–127.
[569] M.P. Ribeiro, A. Espiga, D. Silva, P. Baptista, J. Henriques, C. Ferreira, J.C. Silva, J.P. Borges, E. Pires, P. Chaves, I.J. Correia, Wound Repair Regen. 17 (2009) 817–824.
[570] K. Obara, M. Ishihara, Y. Ozeki, T. Ishizuka, T. Hayashi, S. Nakamura, Y. Saito, H. Yura, T. Matsui, H. Hattori, B. Takase, M. Ishihara, M. Kikuchi, T. Maehara, J. Control. Release 110 (2005) 79–89.
[571] J. Qu, X. Zhao, P.X. Ma, B. Guo, Acta Biomater. 58 (2017) 168–180.
[572] A. Monette, C. Ceccaldi, E. Assaad, S. Lerouge, R. Lapointe, Biomaterials 75 (2016) 237–249.
[573] C. Larsen, Adv. Drug Deliv. Rev. 3 (1989) 103–154.
[574] R. Mehvar, J. Control. Release 69 (2000) 1–25.
[575] W.D.A. Maycock, C.R. Ricketts, Nature 213 (1967) 88.
[576] S.R. Van Tomme, W.E. Hennink, Expert Rev. Med. Devices 4 (2007) 147–164.
[577] C.J. De Groot, M.J.A. Van Luyn, W.N.E. Van Dijk-Wolthuis, J.A. Cadée, J.A. Plantinga, W.D. Otter, W.E. Hennink, Biomaterials 22 (2001) 1197–1203.
[578] L. Pescosolido, L. Feruglio, R. Farra, S. Fiorentino, I. Colombo, T. Coviello, P. Matricardi, W.E. Hennink, T. Vermonden, M. Grassi, Soft Matter 8 (2012) 7708–7715.
[579] L.S. Ferreira, S. Gerecht, J. Fuller, H.F. Shieh, G. Vunjak-Novakovic, R. Langer, Biomaterials 28 (2007) 2706–2717.
[580] L. Ferreira, M.H. Gil, J.S. Dordick, Biomaterials 23 (2002) 3957–3967.
[581] W.N.E. van Dijk-Wolthuis, J.A.M. Hoogeboom, M.J. van Steenbergen, S.K.Y. Tsang, W.E. Hennink, Macromolecules 30 (1997) 4639–4645.
[582] J.A. Cadée, M.J.A. van Luyn, L.A. Brouwer, J.A. Plantinga, P.B. van Wachem, C.J. de Groot, W. den Otter, W.E. Hennink, J. Biomed. Mater. Res. 50 (2000) 397–404.
[583] W.N.E. van Dijk-Wolthuis, O. Franssen, H. Talsma, M.J. van Steenbergen, J.J. Kettenes-van den Bosch, W.E. Hennink, Macromolecules 28 (1995) 6317–6322.
[584] W.N.E. van Dijk-Wolthuis, J.J. Kettenes-van den Bosch, A. van der Kerk-van Hoof, W.E. Hennink, Macromolecules 30 (1997) 3411–3413.
[585] T. Ito, Y. Yeo, C.B. Highley, E. Bellas, D.S. Kohane, Biomaterials 28 (2007) 3418–3426.
[586] S.P. Hudson, R. Langer, G.R. Fink, D.S. Kohane, Biomaterials 31 (2010) 1444–1452.
[587] L. Krentsel, F. Chaubet, A. Rebrov, J. Champion, I. Ermakov, P. Bittoun, S. Fermandjian, A. Litmanovich, N. Platé, J. Jozefonvicz, Carbohydr. Polym. 33 (1997) 63–71.
[588] A. Khademhosseini, R. Langer, Biomaterials 28 (2007) 5087–5092.
[589] R. Jin, L.S. Moreira Teixeira, P.J. Dijkstra, Z. Zhong, C.A. van Blitterswijk, M. Karperien, J. Feijen, Tissue Eng. Part A 16 (2010) 2429–2440.
[590] R. Jin, C. Hiemstra, Z. Zhong, J. Feijen, Biomaterials 28 (2007) 2791–2800.
[591] C. Hiemstra, L.J. van der Aa, Z. Zhong, P.J. Dijkstra, J. Feijen, Biomacromolecules 8 (2007) 1548–1556.
[592] B.G. De Geest, W. Van Camp, F.E. Du Prez, S.C. De Smedt, J. Demeester, W.E. Hennink, Chem. Commun. (2008) 190–192.
[593] L. Naji, J. Schiller, J. Kaufmann, F. Stallmach, J. Kärger, K. Arnold, Biophys. Chem. 104 (2003) 131–140.
[594] S.J. de Jong, S.C. De Smedt, M.W.C. Wahls, J. Demeester, J.J. Kettenes-van den Bosch, W.E. Hennink, Macromolecules 33 (2000) 3680–3686.
[595] W.E. Hennink, S.J. De Jong, G.W. Bos, T.F.J. Veldhuis, C.F. van Nostrum, Int. J. Pharm. 277 (2004) 99–104.
[596] S.J. de Jong, B. van Eerdenbrugh, C.F. van Nostrum, J.J. Kettenes-van den Bosch, W.E. Hennink, J. Control. Release 71 (2001) 261–275.
[597] S.H. Kim, C.Y. Won, C.C. Chu, Carbohydr. Polym. 40 (1999) 183–190.
[598] W.E. Hennink, O. Franssen, W.N.E. van Dijk-Wolthuis, H. Talsma, J. Control. Release 48 (1997) 107–114.
[599] L. Hovgaard, H. Brøndsted, J. Control. Release 36 (1995) 159–166.
[600] H. Brøndsted, C. Andersen, L. Hovgaard, J. Control. Release 53 (1998) 7–13.
[601] S.G. Lévesque, R.M. Lim, M.S. Shoichet, Biomaterials 26 (2005) 7436–7446.
[602] S.G. Lévesque, M.S. Shoichet, Biomaterials 27 (2006) 5277–5285.
[603] O. Ziv, R.R. Avtalion, S. Margel, J. Biomed. Mater. Res. A. 85A (2008) 1011–1021.
[604] R.J.H. Stenekes, A.E. Loebis, C.M. Fernandes, D.J.A. Crommelin, W.E. Hennink, Int. J. Pharm. 214 (2001) 17–20.
[605] E. Khalikova, P. Susi, T. Korpela, Microbiol. Mol. Biol. Rev. 69 (2005) 306–325.
[606] O. Franssen, O.P. Vos, W.E. Hennink, J. Control. Release 44 (1997) 237–245.
[607] X. Wang, Z. Li, T. Shi, P. Zhao, K. An, C. Lin, H. Liu, Mater. Sci. Eng. C 73 (2017) 21–30.
[608] G. Sun, X. Zhang, Y.-I. Shen, R. Sebastian, L.E. Dickinson, K. Fox-Talbot, M. Reinblatt, C. Steenbergen, J.W. Harmon, S. Gerecht, Proc. Natl. Acad. Sci. 108 (2011) 20976–20981.
[609] G. Sun, Y.-I. Shen, S. Kusuma, K. Fox-Talbot, C.J. Steenbergen, S. Gerecht, Biomaterials 32 (2011) 95–106.
[610] G. Sun, J.J. Mao, Nanomedicine 7 (2012) 1771–1784.
[611] Z.Q. Liu, Z. Wei, X.L. Zhu, G.Y. Huang, F. Xu, J.H. Yang, Y. Osada, M. Zrínyi, J.H. Li, Y.M. Chen, Colloids Surf. B Biointerfaces 128 (2015) 140–148.
[612] H. Basan, M. Gümüşderelioğlu, M. Tevfik Orbey, Eur. J. Pharm. Biopharm. 65 (2007) 39–46.
[613] M.A. Casadei, F. Cerreto, S. Cesa, M. Giannuzzo, M. Feeney, C. Marianecci, P. Paolicelli, Int. J. Pharm. 325 (2006) 140–146.
[614] H.-C. Chiu, G.-H. Hsiue, Y.-P. Lee, L.-W. Huang, Polymer Edition 10 (1999) 591–608.
[615] O. Franssen, L. Vandervennet, P. Roders, W.E. Hennink, J. Control. Release 60 (1999) 211–221.
[616] W.E. Hennink, H. Talsma, J.C.H. Borchert, S.C. De Smedt, J. Demeester, J. Control. Release 39 (1996) 47–55.
[617] K.H. Bae, F. Lee, K. Xu, C.T. Keng, S.Y. Tan, Y.J. Tan, Q. Chen, M. Kurisawa, Biomaterials 63 (2015) 146–157.
[618] M.P. Ribeiro, P.I. Morgado, S.P. Miguel, P. Coutinho, I.J. Correia, Mater. Sci. Eng. C 33 (2013) 2958–2966.
[619] A. Zumbuehl, L. Ferreira, D. Kuhn, A. Astashkina, L. Long, Y. Yeo, T. Iaconis, M. Ghannoum, G.R. Fink, R. Langer, D.S. Kohane, Proc. Natl. Acad. Sci. 104 (2007) 12994–12998.
[620] R. Guo, P. Chen, Y. Mo, Y. Lan, W. Xue, Y. Zhang, J. Appl. Biomater. Funct. Mater. 13 (2015) 228–233.
[621] J. Hoque, J. Haldar, ACS Appl. Mater. Interfaces 9 (2017) 15975–15985.
[622] N. Riahi, B. Liberelle, O. Henry, G. De Crescenzo, Carbohydr. Polym. 161 (2017) 219–227.
[623] L. Li, C. Wang, Q. Huang, J. Xiao, Q. Zhang, Y. Cheng, J. Mater. Chem. B 6 (2018) 2474–2480.
[624] V.D. Prajapati, G.K. Jani, B.S. Zala, T.A. Khutliwala, Carbohydr. Polym. 93 (2013) 670–678.
[625] I.B. Bajaj, S.A. Survase, P.S. Saudagar, R.S. Singhal, Food Technol. Biotechnol. 45 (2007) 341–354.
[626] P.-E. Jansson, B. Lindberg, P.A. Sandford, Carbohydr. Res. 124 (1983) 135–139.
[627] E.R. Morris, K. Nishinari, M. Rinaudo, Food Hydrocoll. 28 (2012) 373–411.
[628] H. Grasdalen, O. Smidsrød, Carbohydr. Polym. 7 (1987) 371–393.
[629] C.J. Ferris, K.J. Gilmore, G.G. Wallace, Mih. Panhuis, Soft Matter 9 (2013) 3705–3711.
[630] A.H. Bacelar, J. Silva-Correia, J.M. Oliveira, R.L. Reis, J. Mater. Chem. B 4 (2016) 6164–6174.
[631] E. Miyoshi, T. Takaya, K. Nishinari, Food Hydrocoll. 8 (1994) 505–527.
[632] E. Miyoshi, T. Takaya, K. Nishinari, Carbohydr. Polym. 30 (1996) 109–119.
[633] G. Sworn, G.R. Sanderson, W. Gibson, Food Hydrocoll. 9 (1995) 265–271.
[634] E. Miyoshi, K. Nishinari, Rheological and thermal properties near the sol-gel transition of gellan gum aqueous solutions, in: K. Nishinari (Ed.), Physical Chemistry and Industrial Application of Gellan Gum, vol 114, Springer, Berlin Heidelberg, Berlin, Heidelberg, 1999, pp. 68–82.
[635] L.R. Stevens, K.J. Gilmore, G.G. Wallace, M. in het Panhuis, Biomater. Sci. 4 (2016) 1276–1290.
[636] I. Giavasis, L.M. Harvey, B. McNeil, Crit. Rev. Biotechnol. 20 (2000) 177–211.

[637] K.M. Zia, S. Tabasum, M.F. Khan, N. Akram, N. Akhter, A. Noreen, M. Zuber, Int. J. Biol. Macromol. 109 (2018) 1068–1087.
[638] K.S. Kang, G.T. Veeder, P.J. Mirrasoul, T. Kaneko, I.W. Cottrell, Appl. Environ. Microbiol. 43 (1982) 1086.
[639] S. Vieira, A. da Silva Morais, E. Garet, J. Silva-Correia, R.L. Reis, Á. González-Fernández, J. Miguel Oliveira, Acta Biomater. 93 (2019) 74–85.
[640] D.F. Coutinho, S.V. Sant, H. Shin, J.T. Oliveira, M.E. Gomes, N.M. Neves, A. Khademhosseini, R.L. Reis, Biomaterials 31 (2010) 7494–7502.
[641] A. Vilela Carlos, C. Correia, A. da Silva Morais, C. Santos Tírcia, C. Gertrudes Ana, S. Moreira Elsa, M. Frias Ana, A. Learmonth David, P. Oliveira, M. Oliveira Joaquim, A. Sousa Rui, D. Espregueira-Mendes João, L. Reis Rui, J. Biomed. Mater. Res. A. 106 (2018) 1987–1996.
[642] J. Silva-Correia, J.M. Oliveira, S.G. Caridade, J.T. Oliveira, R.A. Sousa, J.F. Mano, R.L. Reis, J. Tissue Eng. Regen. Med. 5 (2011) e97–e107.
[643] M.B. Oliveira, C.A. Custódio, L. Gasperini, R.L. Reis, J.F. Mano, Acta Biomater. 41 (2016) 119–132.
[644] R. Tsaryk, J. Silva-Correia, J.M. Oliveira, R.E. Unger, C. Landes, C. Brochhausen, S. Ghanaati, R.L. Reis, C.J. Kirkpatrick, J. Tissue Eng. Regen. Med. 11 (2017) 637–648.
[645] Z. Xu, Z. Li, S. Jiang, K.M. Bratlie, ACS Omega 3 (2018) 6998–7007.
[646] R. Lozano, L. Stevens, B.C. Thompson, K.J. Gilmore, R. Gorkin Iii, E.M. Stewart, M. in het Panhuis, M. Romero-Ortega, G.G. Wallace, Biomaterials 67 (2015) 264–273.
[647] C.J. Ferris, L.R. Stevens, K.J. Gilmore, E. Mume, I. Greguric, D.M. Kirchmajer, G.G. Wallace, M. in het Panhuis, J. Mater. Chem. B 3 (2015) 1106–1115.
[648] Y. Gong, C. Wang, R.C. Lai, K. Su, F. Zhang, D.-a. Wang, J. Mater. Chem. 19 (2009) 1968–1977.
[649] C. Wang, Y. Gong, Y. Lin, J. Shen, D.-A. Wang, Acta Biomater. 4 (2008) 1226–1234.
[650] K. Miyamoto, K. Tsuji, T. Nakamura, M. Tokita, T. Komai, Carbohydr. Polym. 30 (1996) 161–164.
[651] A.H. Krauland, V.M. Leitner, A. Bernkop-Schnürch, J. Pharm. Sci. 92 (2003) 1234–1241.
[652] H. Du, P. Hamilton, M. Reilly, N. Ravi, Macromol. Biosci. 12 (2012) 952–961.
[653] N.A. Silva, M.J. Cooke, R.Y. Tam, N. Sousa, A.J. Salgado, R.L. Reis, M.S. Shoichet, Biomaterials 33 (2012) 6345–6354.
[654] N.A. Silva, J. Moreira, S. Ribeiro-Samy, E.D. Gomes, R.Y. Tam, M.S. Shoichet, R.L. Reis, N. Sousa, A.J. Salgado, Biochimie 95 (2013) 2314–2319.
[655] E.D. Gomes, S.S. Mendes, H. Leite-Almeida, J.M. Gimble, R.Y. Tam, M.S. Shoichet, N. Sousa, N.A. Silva, A.J. Salgado, Biomaterials 105 (2016) 38–51.
[656] S. Agnello, L. Gasperini, J.F. Mano, G. Pitarresi, F.S. Palumbo, R.L. Reis, G. Giammona, Int. J. Biol. Macromol. 98 (2017) 646–653.
[657] J.T. Oliveira, T.C. Santos, L. Martins, R. Picciochi, A.P. Marques, A.G. Castro, N.M. Neves, J.F. Mano, R.L. Reis, Tissue Eng. Part A 16 (2009) 343–353.
[658] J.E. Parraga, G.K. Zorzi, Y. Diebold, B. Seijo, A. Sanchez, Int. J. Pharm. 477 (2014) 12–20.
[659] A.M. Soto, J.T. Koivisto, J.E. Parraga, J. Silva-Correia, J.M. Oliveira, R.L. Reis, M. Kellomäki, J. Hyttinen, E. Figueiras, Langmuir 32 (2016) 5173–5182.
[660] J.T. Koivisto, T. Joki, J.E. Parraga, R. Pääkkönen, L. Ylä-Outinen, L. Salonen, I. Jönkkäri, M. Peltola, T.O. Ihalainen, S. Narkilahti, M. Kellomäki, Biomed. Mater. 12 (2017) 025014.
[661] R. López-Cebral, P. Paolicelli, V. Romero-Caamaño, B. Seijo, M.A. Casadei, A. Sanchez, J. Pharm. Sci. 102 (2013) 2632–2643.
[662] A.M. Smith, R.M. Shelton, Y. Perrie, J.J. Harris, J. Biomater. Appl. 22 (2007) 241–254.
[663] S.R. Moxon, A.M. Smith, Int. J. Biol. Macromol. 84 (2016) 79–86.
[664] M. Dentini, P. Desideri, V. Crescenzi, Y. Yuguchi, H. Urakawa, K. Kajiwara, Macromolecules 34 (2001) 1449–1453.
[665] M.-W. Lee, H.-J. Chen, S.-W. Tsao, Carbohydr. Polym. 82 (2010) 920–926.
[666] J.T. Oliveira, L. Martins, R. Picciochi, P.B. Malafaya, R.A. Sousa, N.M. Neves, J.F. Mano, R.L. Reis, J. Biomed. Mater. Res. A. 93A (2010) 852–863.
[667] J. Silva-Correia, V. Miranda-Gonçalves, A.J. Salgado, N. Sousa, J.M. Oliveira, R.M. Reis, R.L. Reis, Tissue Eng. Part A 18 (2012) 1203–1212.
[668] D.A. Silva, A. Poole-Warren Laura, J. Martens Penny, M. Panhuis, J. Appl. Polym. Sci. 130 (2013) 3374–3383.
[669] C.J. Ferris, M. in het Panhuis, Soft Matter 5 (2009) 3430–3437.
[670] L.P. da Silva, A.K. Jha, V.M. Correlo, A.P. Marques, R.L. Reis, K.E. Healy, Adv. Healthc. Mater. 7 (2018) 1700686.
[671] J.T. Oliveira, L.S. Gardel, T. Rada, L. Martins, M.E. Gomes, R.L. Reis, J. Orthop. Res. 28 (2010) 1193–1199.
[672] H.Y. Jeon, E.Y. Shin, J.H. Choi, J.E. Song, R.L. Reis, G. Khang, Macromol. Res. 26 (2018) 724–729.
[673] J.T. Oliveira, T.C. Santos, L. Martins, M.A. Silva, A.P. Marques, A.G. Castro, N.M. Neves, R.L. Reis, J. Tissue Eng. Regen. Med. 3 (2009) 493–500.
[674] L. Costa, J. Silva-Correia, J.M. Oliveira, R.L. Reis, Gellan Gum-Based Hydrogels for Osteochondral Repair, in: J.M. Oliveira, S. Pina, R.L. Reis, J. San Roman (Eds.), Osteochondral Tissue Engineering: Nanotechnology, Scaffolding-Related Developments and Translation, vol 1058, Springer International Publishing, Cham, 2018, pp. 281–304.
[675] M. Raquel, M. David, N. Dorit, S. Lucília Pereira da, R.B. Ana, B.C. João, O. Joaquim Miguel, C. Vitor Manuel, R. Rui Luis, C. Jillian, Biomed. Mater. 13 (2018) 035012.
[676] M. Lago, L. da Silva, C. Henriques, A. Carvalho, R. Reis, A. Marques, Bioengineering 5 (2018) 52.
[677] C. Carvalho, S. Wrobel, C. Meyer, C. Brandenberger, I.F. Cengiz, R. Lopez-Cebral, J. Silva-Correia, G. Ronchi, R.L. Reis, C. Grothe, J.M. Oliveira, K. Haastert-Talini, Biomater. Sci. 6 (2018) 1059–1075.
[678] D.R. Pereira, J. Silva-Correia, S.G. Caridade, J.T. Oliveira, R.A. Sousa, A.J. Salgado, J.M. Oliveira, J.F. Mano, N. Sousa, R.L. Reis, Tissue Eng. Part C Methods 17 (2011) 961–972.
[679] G. Khang, S.K. Lee, H.N. Kim, J. Silva-Correia, M.E. Gomes, C.A.A. Viegas, I.R. Dias, J.M. Oliveira, R.L. Reis, J. Tissue Eng. Regen. Med. 9 (2015) 265–275.
[680] P. Matricardi, C. Cencetti, R. Ria, F. Alhaique, T. Coviello, Molecules 14 (2009) 3376–3391.
[681] S. Miyazaki, H. Aoyama, N. Kawasaki, W. Kubo, D. Attwood, J. Control. Release 60 (1999) 287–295.
[682] T. Osmałek, A. Froelich, S. Tasarek, Int. J. Pharm. 466 (2014) 328–340.
[683] C.A. Carmona-Moran, O. Zavgorodnya, A.D. Penman, E. Kharlampieva, S.L. Bridges, R.W. Hergenrother, J.A. Singh, T.M. Wick, Int. J. Pharm. 509 (2016) 465–476.
[684] B. Newland, M. Baeger, D. Eigel, H. Newland, C. Werner, ACS Biomater. Sci. Eng. 3 (2017) 787–792.
[685] R. López-Cebral, A. Civantos, V. Ramos, B. Seijo, J.L. López-Lacomba, J.V. Sanz-Casado, A. Sanchez, Carbohydr. Polym. 167 (2017) 345–355.
[686] V. Perugini, A.L. Guildford, J. Silva-Correia, J.M. Oliveira, S.T. Meikle, R.L. Reis, M. Santin, J. Tissue Eng. Regen. Med. 12 (2018) e669–e678.
[687] R.J. Babu, S. Sathigari, M.T. Kumar, J.K. Pandit, Curr. Drug Deliv. 7 (2010) 36–43.
[688] T.E.L. Douglas, A. Dokupil, K. Reczyńska, G. Brackman, M. Krok-Borkowicz, J.K. Keppler, M. Božič, P. Van Der Voort, K. Pietryga, S.K. Samal, L. Balcaen, J. van den Bulcke, J. Van Acker, F. Vanhaecke, K. Schwarz, T. Coenye, E. Pamuła, Biomed. Mater. 11 (2016) 045015.
[689] M.A. Bonifacio, S. Cometa, A. Cochis, P. Gentile, A.M. Ferreira, B. Azzimonti, G. Procino, E. Ceci, L. Rimondini, E. De Giglio, Carbohydr. Polym. 198 (2018) 462–472.
[690] U. Posadowska, M. Brzychczy-Wloch, E. Pamula, J. Mater. Sci. Mater. Med. 27 (2015) 9.
[691] C. Schiraldi, D. Cimini, M. De Rosa, Appl. Microbiol. Biotechnol. 87 (2010) 1209–1220.
[692] P.S. Chan, J.P. Caron, G.J.M. Rosa, M.W. Orth, Osteoarthr. Cartil. 13 (2005) 387–394.
[693] T. Laabs, D. Carulli, H.M. Geller, J.W. Fawcett, Curr. Opin. Neurobiol. 15 (2005) 116–120.
[694] H.-Y. Hwang, S.K. Olson, J.D. Esko, H. Robert Horvitz, Nature 423 (2003) 439–443.
[695] C.B. Knudson, W. Knudson, Semin. Cell Dev. Biol. 12 (2001) 69–78.
[696] K. Sugahara, T. Mikami, T. Uyama, S. Mizuguchi, K. Nomura, H. Kitagawa, Curr. Opin. Struct. Biol. 13 (2003) 612–620.
[697] E. Fthenou, A. Zafiropoulos, P. Katonis, A. Tsatsakis, N.K. Karamanos, G.N. Tzanakakis, J. Cell. Biochem. 103 (2008) 1866–1876.
[698] S. Sirko, A. von Holst, A. Weber, A. Wizenmann, U. Theocharidis, M. Götz, A. Faissner, Stem Cells 28 (2010) 775–787.
[699] V. Hintze, A. Miron, S. Moeller, M. Schnabelrauch, H.P. Wiesmann, H. Worch, D. Scharnweber, Acta Biomater. 8 (2012) 2144–2152.
[700] D.-A. Wang, S. Varghese, B. Sharma, I. Strehin, S. Fermanian, J. Gorham, D.H. Fairbrother, B. Cascio, J.H. Elisseeff, Nat. Mater. 6 (2007) 385–392.
[701] S. Varghese, N.S. Hwang, A.C. Canver, P. Theprungsirikul, D.W. Lin, J. Elisseeff, Matrix Biol. 27 (2008) 12–21.
[702] Q. Li, D.-a. Wang, J.H. Elisseeff, Macromolecules 36 (2003) 2556–2562.
[703] S.J. Bryant, J.A. Arthur, K.S. Anseth, Acta Biomater. 1 (2005) 243–252.
[704] S.J. Bryant, K.A. Davis-Arehart, N. Luo, R.K. Shoemaker, J.A. Arthur, K.S. Anseth, Macromolecules 37 (2004) 6726–6733.
[705] Q. Li, C.G. Williams, D.D.N. Sun, J. Wang, K. Leong, J.H. Elisseeff, J. Biomed. Mater. Res. A. 68A (2004) 28–33.
[706] H.D. Kim, E.A. Lee, Y.-H. An, S.L. Kim, S.S. Lee, S.J. Yu, H.L. Jang, K.T. Nam, S.G. Im, N.S. Hwang, ACS Appl. Mater. Interfaces 9 (2017) 21639–21650.
[707] T.C. Suekama, J. Hu, T. Kurokawa, J.P. Gong, S.H. Gehrke, ACS Macro Lett. 2 (2013) 137–140.
[708] P.A. Levett, F.P.W. Melchels, K. Schrobback, D.W. Hutmacher, J. Malda, T.J. Klein, Acta Biomater. 10 (2014) 214–223.
[709] H. Eva, T.G.E. M, B. Kirsten, Bioinspired Biomim. Nanobiomaterials 5 (2016) 51–66.
[710] M. Lopez-Moya, P. Melgar-Lesmes, K. Kolandaivelu, J.M. de la Torre Hernández, E.R. Edelman, M. Balcells, Biomacromolecules 19 (2018) 1234–1244.
[711] C. Liu, L. Fan, J. Xing, Q. Wang, C. Lin, C. Liu, X. Deng, C. Ning, L. Zhou, L. Rong, B. Liu, Biomater. Sci. 7 (2019) 1995–2008.
[712] I. Strehin, W.M. Ambrose, O. Schein, A. Salahuddin, J. Elisseeff, J. Cataract Refract. Surg. 35 (2009) 567–576.
[713] J.M.G. Reyes, S. Herretes, A. Pirouzmanesh, D.-A. Wang, J.H. Elisseeff, A. Jun, P.J. McDonnell, R.S. Chuck, A. Behrens, Invest. Ophthalmol. Vis. Sci. 46 (2005) 1247–1250.
[714] S. Dawlee, A. Sugandhi, B. Balakrishnan, D. Labarre, A. Jayakrishnan, Biomacromolecules 6 (2005) 2040–2048.
[715] T. Bobula, R. Buffa, P. Procházková, H. Vágnerová, V. Moravcová, R. Šuláková, O. Židek, V. Velebný, Carbohydr. Polym. 136 (2016) 1002–1009.
[716] P. Esmaeilzadeh, M. Menzel, T. Groth, ACS Appl. Mater. Interfaces 10 (2018) 31168–31177.
[717] A. Conovaloff, A. Panitch, J. Neural Eng. 8 (2011) 056003.
[718] A. van der Smissen, V. Hintze, D. Scharnweber, S. Moeller, M. Schnabelrauch, A. Majok, J.C. Simon, U. Anderegg, Biomaterials 32 (2011) 8938–8946.
[719] X. Hu, D. Li, F. Zhou, C. Gao, Acta Biomater. 7 (2011) 1618–1626.
[720] K.R. Kirker, Y. Luo, J.H. Nielson, J. Shelby, G.D. Prestwich, Biomaterials 23 (2002) 3661–3671.

[721] C.Y. Pumphrey, A.M. Theus, S. Li, R.S. Parrish, R.D. Sanderson, Cancer Res. 62 (2002) 3722.
[722] X. Bai, S. Lü, Z. Cao, B. Ni, X. Wang, P. Ning, D. Ma, H. Wei, M. Liu, Carbohydr. Polym. 166 (2017) 123–130.
[723] L. Borsig, L. Wang, M.C.M. Cavalcante, L. Cardilo-Reis, P.L. Ferreira, P.A.S. Mourão, J.D. Esko, M.S.G. Pavão, J. Biol. Chem. 282 (2007) 14984–14991.
[724] J. Xi, L. Zhou, H. Dai, Colloids Surf. B Biointerfaces 100 (2012) 107–115.
[725] M.E. Gilbert, K.R. Kirker, S.D. Gray, P.D. Ward, J.G. Szakacs, G.D. Prestwich, R.R. Orlandi, Laryngoscope 114 (2004) 1406–1409.
[726] F. Ronca, L. Palmieri, P. Panicucci, G. Ronca, Osteoarthr. Cartil. 6 (1998) 14–21.
[727] M. Iovu, G. Dumais, P. du Souich, Osteoarthr. Cartil. 16 (2008) S14–S18.
[728] T.E. McAlindon, M.P. LaValley, J.P. Gulin, D.T. Felson, JAMA 283 (2000) 1469–1475.
[729] S. Suzuki, H. Saito, T. Yamagata, K. Anno, N. Seno, Y. Kawai, T. Furuhashi, J. Biol. Chem. 243 (1968) 1543–1550.
[730] M. Liu, X. Zeng, C. Ma, H. Yi, Z. Ali, X. Mou, S. Li, Y. Deng, N. He, Bone Res. 5 (2017) 17014.
[731] M. Büttner, S. Möller, M. Keller, D. Huster, J. Schiller, M. Schnabelrauch, P. Dieter, U. Hempel, J. Cell. Physiol. 228 (2013) 330–340.
[732] M.I. Betancur, H.D. Mason, M. Alvarado-Velez, P.V. Holmes, R.V. Bellamkonda, L. Karumbaiah, ACS Biomater. Sci. Eng. 3 (2017) 420–430.
[733] L. Karumbaiah, S.F. Enam, A.C. Brown, T. Saxena, M.I. Betancur, T.H. Barker, R.V. Bellamkonda, Bioconjug. Chem. 26 (2015) 2336–2349.
[734] S. Sirko, A. von Holst, A. Wizenmann, M. Götz, A. Faissner, Development 134 (2007) 2727.
[735] J.F. Piai, M.A. da Silva, A. Martins, A.B. Torres, S. Faria, R.L. Reis, E.C. Muniz, N.M. Neves, Appl. Surf. Sci. 403 (2017) 112–125.
[736] E.A. Aisenbrey, S.J. Bryant, Biomaterials 190-191 (2019) 51–62.
[737] L. Zhao, M. Liu, J. Wang, G. Zhai, Carbohydr. Polym. 133 (2015) 391–399.
[738] J.J. Lim, T.M. Hammoudi, A.M. Bratt-Leal, S.K. Hamilton, K.L. Kepple, N.C. Bloodworth, T.C. McDevitt, J.S. Temenoff, Acta Biomater. 7 (2011) 986–995.
[739] M.K. Yoo, K.Y. Cho, H.H. Song, Y.J. Choi, J.W. Kwon, M.K. Kim, J.H. Lee, W.R. Wee, C.S. Cho, Drug Dev. Ind. Pharm. 31 (2005) 455–463.
[740] R. Anouz, A. Repanas, E. Schwarz, T. Groth, Macromol. Biosci. 18 (2018) 1800283.
[741] X.H. Zou, Y.Z. Jiang, G.R. Zhang, H.M. Jin, N.T.M. Hieu, H.W. Ouyang, Acta Biomater. 5 (2009) 1588–1595.
[742] B. Casu, Structure and Biological Activity of Heparin, in: R.S. Tipson, D. Horton (Eds.), Advances in Carbohydrate Chemistry and Biochemistry, vol 43, Academic Press, 1985, pp. 51–134.
[743] B. Mulloy, M.J. Forster, C. Jones, D.B. Davies, Biochem. J. 293 (1993) 849.
[744] D. Edgar, R. Timpl, H. Thoenen, EMBO J. 3 (1984) 1463–1468.
[745] K. Holme, M. Ishihara, P.N. Shaklee, R.J. Stack, W. Liang, Z. Wei, Z. Yang, Glycobiology 4 (1994) 451–458.
[746] S.L. Rogers, J.B. McCarthy, S.L. Palm, L.T. Furcht, P.C. Letourneau, J. Neurosci. 5 (1985) 369.
[747] J. Choay, M. Petitou, J.C. Lormeau, P. Sinaÿ, B. Casu, G. Gatti, Biochem. Biophys. Res. Commun. 116 (1983) 492–499.
[748] R. Ruppert, E. Hoffmann, W. Sebald, Eur. J. Biochem. 237 (1996) 295–302.
[749] S.E. Sakiyama-Elbert, Acta Biomater. 10 (2014) 1581–1587.
[750] Y. Liang, K.L. Kiick, Acta Biomater. 10 (2014) 1588–1600.
[751] R. Langer, Nature (1998) 5–10.
[752] T. Fernández-Muiños, L. Recha-Sancho, P. López-Chicón, C. Castells-Sala, A. Mata, C.E. Semino, Acta Biomater. 16 (2015) 35–48.
[753] U. Freudenberg, A. Hermann, P.B. Welzel, K. Stirl, S.C. Schwarz, M. Grimmer, A. Zieris, W. Panyanuwat, S. Zschoche, D. Meinhold, A. Storch, C. Werner, Biomaterials 30 (2009) 5049–5060.
[754] Y. Peng, L.E. Tellier, J.S. Temenoff, Biomater. Sci. 4 (2016) 1371–1380.
[755] K. Zeng, T. Groth, K. Zhang, ChemBioChem 20 (2019) 737–746.
[756] J. You, V.K. Raghunathan, K.J. Son, D. Patel, A. Haque, C.J. Murphy, A. Revzin, ACS Appl. Mater. Interfaces 7 (2015) 12299–12308.
[757] G. Tae, Y.-J. Kim, W.-I. Choi, M. Kim, P.S. Stayton, A.S. Hoffman, Biomacromolecules 8 (2007) 1979–1986.
[758] S.S. Shah, M. Kim, K. Cahill-Thompson, G. Tae, A. Revzin, Soft Matter 7 (2011) 3133–3140.
[759] M.E. Ogle, J.R. Krieger, L.E. Tellier, J. McFaline-Figueroa, J.S. Temenoff, E.A. Botchwey, ACS Biomater. Sci. Eng. 4 (2018) 1241–1250.
[760] J.R. Krieger, M.E. Ogle, J. McFaline-Figueroa, C.E. Segar, J.S. Temenoff, E.A. Botchwey, Biomaterials 77 (2016) 280–290.
[761] M.V. Tsurkan, K.R. Levental, U. Freudenberg, C. Werner, Chem. Commun. 46 (2010) 1141–1143.
[762] G. Tae, M. Scatena, P.S. Stayton, A.S. Hoffman, J. Biomater. Sci. Polym. Ed. 17 (2006) 187–197.
[763] H. Ding, M. Zhong, Y.J. Kim, P. Pholpabu, A. Balasubramanian, C.M. Hui, H. He, H. Yang, K. Matyjaszewski, C.J. Bettinger, ACS Nano 8 (2014) 4348–4357.
[764] A. Nilasaroya, L.A. Poole-Warren, J.M. Whitelock, P. Jo Martens, Biomaterials 29 (2008) 4658–4664.
[765] O. Jeon, K. Lee, E. Alsberg, Small 14 (2018) 1800579.
[766] T. Nie, R.E. Akins, K.L. Kiick, Acta Biomater. 5 (2009) 865–875.
[767] T. Nie, A. Baldwin, N. Yamaguchi, K.L. Kiick, J. Control. Release 122 (2007) 287–296.
[768] Y.K. Joung, S.S. You, K.M. Park, D.H. Go, K.D. Park, Colloids Surf. B Biointerfaces 99 (2012) 102–107.
[769] Z. Yang, J. Wang, R. Luo, M.F. Maitz, F. Jing, H. Sun, N. Huang, Biomaterials 31 (2010) 2072–2083.
[770] M.F. Maitz, J. Zitzmann, J. Hanke, C. Renneberg, M.V. Tsurkan, C. Sperling, U. Freudenberg, C. Werner, Biomaterials 135 (2017) 53–61.
[771] M.V. Tsurkan, R. Wetzel, H.R. Pérez-Hernández, K. Chwalek, A. Kozlova, U. Freudenberg, G. Kempermann, Y. Zhang, A.F. Lasagni, C. Werner, Adv. Healthc. Mater. 4 (2015) 516–521.
[772] J.C. Krupa, A. Marie Butler, J.S. Mort, Anal. Biochem. 319 (2003) 280–286.
[773] D.S.W. Benoit, A.R. Durney, K.S. Anseth, Biomaterials 28 (2007) 66–77.
[774] D.S.W. Benoit, K.S. Anseth, Acta Biomater. 1 (2005) 461–470.
[775] M. Kim, Y. Shin, B.-H. Hong, Y.-J. Kim, J.-S. Chun, G. Tae, Y.H. Kim, Tissue Eng. Part C Methods 16 (2009) 1–10.
[776] M. Kim, S.E. Kim, S.S. Kang, Y.H. Kim, G. Tae, Biomaterials 32 (2011) 7883–7896.
[777] J. You, D.-S. Shin, D. Patel, Y. Gao, A. Revzin, Adv. Healthc. Mater. 3 (2014) 126–132.
[778] M. Kim, J.Y. Lee, C.N. Jones, A. Revzin, G. Tae, Biomaterials 31 (2010) 3596–3603.
[779] Y. Hwang, M. Goh, M. Kim, G. Tae, Biomaterials 165 (2018) 94–104.
[780] E. Foster, J. You, C. Siltanen, D. Patel, A. Haque, L. Anderson, A. Revzin, Eur. Polym. J. 72 (2015) 726–735.
[781] J.A. Beamish, L.C. Geyer, N.A. Haq-Siddiqi, K. Kottke-Marchant, R.E. Marchant, Biomaterials 30 (2009) 6286–6294.
[782] S. Nakamura, M. Ishihara, K. Obara, K. Masuoka, T. Ishizuka, Y. Kanatani, B. Takase, T. Matsui, H. Hattori, T. Sato, Y. Kariya, T. Maehara, J. Biomed. Mater. Res. A. 78A (2006) 364–371.
[783] H.M. Weber, M.V. Tsurkan, V. Magno, U. Freudenberg, C. Werner, Acta Biomater. 57 (2017) 59–69.
[784] R. Wieduwild, M. Tsurkan, K. Chwalek, P. Murawala, M. Nowak, U. Freudenberg, C. Neinhuis, C. Werner, Y. Zhang, J. Am. Chem. Soc. 135 (2013) 2919–2922.
[785] S.B. Seif-Naraghi, D. Horn, P.J. Schup-Magoffin, K.L. Christman, Acta Biomater. 8 (2012) 3695–3703.
[786] D.B. Pike, S. Cai, K.R. Pomraning, M.A. Firpo, R.J. Fisher, X.Z. Shu, G.D. Prestwich, R.A. Peattie, Biomaterials 27 (2006) 5242–5251.
[787] W.I. Choi, M. Kim, G. Tae, Y.H. Kim, Biomacromolecules 9 (2008) 1698–1704.
[788] G. Bhakta, B. Rai, Z.X.H. Lim, J.H. Hui, G.S. Stein, A.J. van Wijnen, V. Nurcombe, G.D. Prestwich, S.M. Cool, Biomaterials 33 (2012) 6113–6122.
[789] L.E. Tellier, E.A. Treviño, A.L. Brimeyer, D.S. Reece, N.J. Willett, R.E. Guldberg, J.S. Temenoff, Biomater. Sci. 6 (2018) 1159–1167.
[790] M. Goh, Y. Hwang, G. Tae, Carbohydr. Polym. 147 (2016) 251–260.
[791] U. Freudenberg, A. Zieris, K. Chwalek, M.V. Tsurkan, M.F. Maitz, P. Atallah, K.R. Levental, S.A. Eming, C. Werner, J. Control. Release 220 (2015) 79–88.
[792] N. Lohmann, L. Schirmer, P. Atallah, E. Wandel, R.A. Ferrer, C. Werner, J.C. Simon, S. Franz, U. Freudenberg, Sci. Transl. Med. 9 (2017) eaai9044.
[793] F.P. Seib, M. Tsurkan, U. Freudenberg, D.L. Kaplan, C. Werner, ACS Biomater. Sci. Eng. 2 (2016) 2287–2293.
[794] L.J. Bray, M. Binner, A. Holzheu, J. Friedrichs, U. Freudenberg, D.W. Hutmacher, C. Werner, Biomaterials 53 (2015) 609–620.
[795] T.C. Laurent, J.R. Fraser, Faseb J. 6 (1992) 2397–2404.
[796] G. Kogan, L. Šoltés, R. Stern, P. Gemeiner, Biotechnol. Lett. 29 (2007) 17–25.
[797] H. Knopf-Marques, M. Pravda, L. Wolfova, V. Velebny, P. Schaaf, N.E. Vrana, P. Lavalle, Adv. Healthc. Mater. 5 (2016) 2841–2855.
[798] K. Meyer, Physiol. Rev. 27 (1947) 335–359.
[799] S. Al-Assaf, S. Navaratnam, B.J. Parsons, G.O. Phillips, Arch. Biochem. Biophys. 411 (2003) 73–82.
[800] J.A. Burdick, G.D. Prestwich, Adv. Mater. 23 (2011) H41–H56.
[801] J.A. Burdick, C. Chung, X. Jia, M.A. Randolph, R. Langer, Biomacromolecules 6 (2005) 386–391.
[802] M.A. Serban, A. Skardal, Matrix Biol. 78–79 (2019) 337–345.
[803] M.N. Collins, C. Birkinshaw, Carbohydr. Polym. 92 (2013) 1262–1279.
[804] C. Loebel, R.L. Mauck, J.A. Burdick, Nat. Mater. 18 (2019) 883–891.
[805] E. Sideris, D.R. Griffin, Y. Ding, S. Li, W.M. Weaver, D. Di Carlo, T. Hsiai, T. Segura, ACS Biomater. Sci. Eng. 2 (2016) 2034–2041.
[806] C.B. Highley, G.D. Prestwich, J.A. Burdick, Curr. Opin. Biotechnol. 40 (2016) 35–40.
[807] S. Gerecht, J.A. Burdick, L.S. Ferreira, S.A. Townsend, R. Langer, G. Vunjak-Novakovic, Proc. Natl. Acad. Sci. 104 (2007) 11298–11303.
[808] L. Ouyang, C.B. Highley, C.B. Rodell, W. Sun, J.A. Burdick, ACS Biomater. Sci. Eng. 2 (2016) 1743–1751.
[809] C.B. Rodell, A.L. Kaminski, J.A. Burdick, Biomacromolecules 14 (2013) 4125–4134.
[810] A.M. Rosales, C.B. Rodell, M.H. Chen, M.G. Morrow, K.S. Anseth, J.A. Burdick, Bioconjug. Chem. 29 (2018) 905–913.
[811] S. Khetan, M. Guvendiren, W.R. Legant, D.M. Cohen, C.S. Chen, J.A. Burdick, Nat. Mater. 12 (2013) 458–465.
[812] C.B. Rodell, R.J. Wade, B.P. Purcell, N.N. Dusaj, J.A. Burdick, ACS Biomater. Sci. Eng. 1 (2015) 277–286.
[813] M.G. Ondeck, A.J. Engler, J. Biomech. Eng. 138 (2016) 021003-021003-021006.
[814] M. Bao, J. Xie, A. Piruska, W.T.S. Huck, Nat. Commun. 8 (2017) 1962.
[815] Y.D. Park, N. Tirelli, J.A. Hubbell, Biomaterials 24 (2003) 893–900.
[816] G.D. Prestwich, D.M. Marecak, J.F. Marecek, K.P. Vercruysse, M.R. Ziebell, J. Control. Release 53 (1998) 93–103.
[817] Z. Guo, S. Mi, W. Sun, J. Mater. Chem. B 6 (2018) 6234–6244.
[818] M. Weis, J. Shan, M. Kuhlmann, T. Jungst, J. Tessmar, J. Groll, Gels 4 (2018) 82.
[819] Q. Feng, S. Lin, K. Zhang, C. Dong, T. Wu, H. Huang, X. Yan, L. Zhang, G. Li, L. Bian, Acta Biomater. 53 (2017) 329–342.
[820] H. Tian, Z. He, C. Sun, C. Yang, P. Zhao, L. Liu, W. Leong Kam, H.-Q. Mao, Z. Liu, Y. Chen, Adv. Healthc. Mater. 7 (2018) 1800285.
[821] R. Jin, L.S. Moreira Teixeira, A. Krouwels, P.J. Dijkstra, C.A. van Blitterswijk, M. Karperien, J. Feijen, Acta Biomater. 6 (2010) 1968–1977.

[822] J.M. Townsend, B.T. Andrews, Y. Feng, J. Wang, R.J. Nudo, E. Van Kampen, S.H. Gehrke, C.J. Berkland, M.S. Detamore, Acta Biomater. 71 (2018) 148–155.
[823] O. Schmut, H. Hofmann, Graefes Arch. Clin. Exp. Ophthalmol. 218 (1982) 311–314.
[824] J. Shin, J.S. Lee, C. Lee, H.-J. Park, K. Yang, Y. Jin, J.H. Ryu, K.S. Hong, S.-H. Moon, H.-M. Chung, H.S. Yang, S.H. Um, J.-W. Oh, D.-I. Kim, H. Lee, S.-W. Cho, Adv. Funct. Mater. 25 (2015) 3814–3824.
[825] J. Baier Leach, K.A. Bivens, C.W. Patrick Jr, C.E. Schmidt, Biotechnol. Bioeng. 82 (2003) 578–589.
[826] M. Rosales Adrianne, L. Vega Sebastián, W. DelRio Frank, A. Burdick Jason, S. Anseth Kristi, Angew. Chemie Int. Ed. 56 (2017) 12132–12136.
[827] S.A. Fisher, P.N. Anandakumaran, S.C. Owen, M.S. Shoichet, Adv. Funct. Mater. 25 (2015) 7163–7172.
[828] X.Z. Shu, Y. Liu, Y. Luo, M.C. Roberts, G.D. Prestwich, Biomacromolecules 3 (2002) 1304–1311.
[829] E. Larrañeta, M. Henry, N.J. Irwin, J. Trotter, A.A. Perminova, R.F. Donnelly, Carbohydr. Polym. 181 (2018) 1194–1205.
[830] K. Tomihata, Y. Ikada, J. Biomed. Mater. Res. 37 (1997) 243–251.
[831] H.-J. Park, Y. Jin, J. Shin, K. Yang, C. Lee, H.S. Yang, S.-W. Cho, Biomacromolecules 17 (2016) 1939–1948.
[832] D.C. West, I.N. Hampson, F. Arnold, S. Kumar, Science 228 (1985) 1324–1326.
[833] J. Lam, N.F. Truong, T. Segura, Acta Biomater. 10 (2014) 1571–1580.
[834] D.D. Allison, K.J. Grande-Allen, Tissue Eng. 12 (2006) 2131–2140.
[835] F. Zamboni, S. Vieira, R.L. Reis, J. Miguel Oliveira, M.N. Collins, Prog. Mater. Sci. 97 (2018) 97–122.
[836] J. Kim, I.S. Kim, T.H. Cho, K.B. Lee, S.J. Hwang, G. Tae, I. Noh, S.H. Lee, Y. Park, K. Sun, Biomaterials 28 (2007) 1830–1837.
[837] N. Cui, J. Qian, T. Liu, N. Zhao, H. Wang, Carbohydr. Polym. 126 (2015) 192–198.
[838] L. Bian, C. Hou, E. Tous, R. Rai, R.L. Mauck, J.A. Burdick, Biomaterials 34 (2013) 413–421.
[839] L. Bian, D.Y. Zhai, E. Tous, R. Rai, R.L. Mauck, J.A. Burdick, Biomaterials 32 (2011) 6425–6434.
[840] Y.B. Park, C.W. Ha, J.A. Kim, W.J. Han, J.H. Rhim, H.J. Lee, K.J. Kim, Y.G. Park, J.Y. Chung, Osteoarthr. Cartil. 25 (2017) 570–580.
[841] C.-W. Ha, Y.-B. Park, J.-Y. Chung, Y.-G. Park, Stem Cells Transl. Med. 4 (2015) 1044–1051.
[842] M. Kim, I.E. Erickson, A.H. Huang, S.T. Garrity, R.L. Mauck, D.R. Steinberg, Tissue Eng. Part A 24 (2018) 1693–1703.
[843] I.L. Mohd Isa, S.A. Abbah, M. Kilcoyne, D. Sakai, P. Dockery, D.P. Finn, A. Pandit, Sci. Adv. 4 (2018) eaaq0597.
[844] D. Shendi, A. Dede, Y. Yin, C. Wang, C. Valmikinathan, A. Jain, J. Mater. Chem. B 4 (2016) 2803–2818.
[845] S. Wu, R. Xu, B. Duan, P. Jiang, J. Mater. Chem. B 5 (2017) 3870–3878.
[846] N. Mitrousis, R.Y. Tam, A.E.G. Baker, D. van der Kooy, M.S. Shoichet, Adv. Funct. Mater. 26 (2016) 1975–1985.
[847] R.E. Thompson, J. Pardieck, L. Smith, P. Kenny, L. Crawford, M. Shoichet, S. Sakiyama-Elbert, Biomaterials 162 (2018) 208–223.
[848] K. Zaviskova, D. Tukmachev, J. Dubisova, I. Vackova, A. Hejcl, J. Bystronova, M. Pravda, I. Scigalkova, R. Sulakova, V. Velebny, L. Wolfova, S. Kubinova, J. Biomed. Mater. Res. A. 106 (2018) 1129–1140.
[849] T. Führmann, J. Obermeyer, C.H. Tator, M.S. Shoichet, Methods 84 (2015) 60–69.
[850] L.A. Solchaga, J.E. Dennis, V.M. Goldberg, A.I. Caplan, J. Orthop. Res. 17 (1999) 205–213.
[851] F. Dosio, S. Arpicco, B. Stella, E. Fattal, Adv. Drug Deliv. Rev. 97 (2016) 204–236.
[852] Y. Zhang, F. Wang, M. Li, Z. Yu, R. Qi, J. Ding, Z. Zhang, X. Chen, Adv. Sci. 5 (2018) 1700821.
[853] Y. Luo, K.R. Kirker, G.D. Prestwich, J. Control. Release 69 (2000) 169–184.
[854] J.E. Mealy, C.B. Rodell, J.A. Burdick, J. Mater. Chem. B 3 (2015) 8010–8019.
[855] H. Kim, Y. Park, M.M. Stevens, W. Kwon, S.K. Hahn, J. Control. Release 303 (2019) 55–66.
[856] N.F. Truong, T. Segura, ACS Biomater. Sci. Eng. 4 (2018) 981–987.
[857] T. Tokatlian, C. Cam, T. Segura, Adv. Healthc. Mater. 4 (2015) 1084–1091.
[858] I. Villate-Beitia, N.F. Truong, I. Gallego, J. Zárate, G. Puras, J.L. Pedraz, T. Segura, RSC Adv. 8 (2018) 31934–31942.
[859] R.A. Peattie, D.B. Pike, B. Yu, S. Cai, X.Z. Shu, G.D. Prestwich, M.A. Firpo, R.J. Fisher, Drug Deliv. 15 (2008) 389–397.
[860] P. Bulpitt, D. Aeschlimann, J. Biomed. Mater. Res. 47 (1999) 152–169.
[861] J.L. Holloway, H. Ma, R. Rai, J.A. Burdick, J. Control. Release 191 (2014) 63–70.
[862] X. Xu, A.K. Jha, R.L. Duncan, X. Jia, Acta Biomater. 7 (2011) 3050–3059.
[863] L. Shi, Y. Zhao, Q. Xie, C. Fan, J. Hilborn, J. Dai, D.A. Ossipov, Adv. Healthc. Mater. 7 (2018) 1700973.
[864] S.V. Murphy, A. Skardal, L. Song, K. Sutton, R. Haug, D.L. Mack, J. Jackson, S. Soker, A. Atala, Stem Cells Transl. Med. 6 (2017) 2020–2032.
[865] V. Quagliariello, E. Armenia, C. Aurilio, F. Rosso, O. Clemente, G. de Sena, M. Barbarisi, A. Barbarisi, J. Cell. Physiol. 231 (2016) 1784–1795.
[866] M.C. Gómez-Guillén, B. Giménez, M.E. López-Caballero, M.P. Montero, Food Hydrocoll. 25 (2011) 1813–1827.
[867] L.S. Nair, C.T. Laurencin, Prog. Polym. Sci. 32 (2007) 762–798.
[868] K. Lin, D. Zhang, M.H. Macedo, W. Cui, B. Sarmento, G. Shen, Adv. Funct. Mater. 29 (2019) 1804943.
[869] D.R. Eyre, J.-J. Wu, Collagen Cross-Links, J. Brinckmann, H. Notbohm, P.K. Müller (Eds.), Collagen: Primer in Structure, Processing and Assembly, vol 247, Springer, Berlin Heidelberg, Berlin, Heidelberg, 2005, pp. 207–229.
[870] W.F. Harrington, P.H. Von Hippel, The Structure Of Collagen And Gelatin, in: C.B. Anfinsen, M.L. Anson, K. Bailey, J.T. Edsall (Eds.), Advances in Protein Chemistry, vol 16, Academic Press, 1962, pp. 1–138.
[871] W. Traub, K.A. Piez, The Chemistry and Structure of Collagen, in: C.B. Anfinsen, J.T. Edsall, F.M. Richards (Eds.), Advances in Protein Chemistry, vol 25, Academic Press, 1971, pp. 243–352.
[872] D.R. Eyre, M.A. Paz, P.M. Gallop, Annu. Rev. Biochem. 53 (1984) 717–748.
[873] P. Fratzl, K. Misof, I. Zizak, G. Rapp, H. Amenitsch, S. Bernstorff, J. Struct. Biol. 122 (1998) 119–122.
[874] J. Glowacki, S. Mizuno, Biopolymers 89 (2007) 338–344.
[875] C.H. Lee, A. Singla, Y. Lee, Int. J. Pharm. 221 (2001) 1–22.
[876] B. Lanfer, F.P. Seib, U. Freudenberg, D. Stamov, T. Bley, M. Bornhäuser, C. Werner, Biomaterials 30 (2009) 5950–5958.
[877] L. Gu, T. Shan, Y.-x. Ma, F.R. Tay, L. Niu, Trends Biotechnol. 37 (2019) 464–491.
[878] J.C. Tiller, G. Bonner, L.-C. Pan, A.M. Klibanov, Biotechnol. Bioeng. 73 (2001) 246–252.
[879] C. Fan, J. Shi, Y. Zhuang, L. Zhang, L. Huang, W. Yang, B. Chen, Y. Chen, Z. Xiao, H. Shen, Y. Zhao, J. Dai, Adv. Mater. 31 (2019) 1902900.
[880] S. Koutsopoulos, S. Zhang, Acta Biomater. 9 (2013) 5162–5169.
[881] G. Forgacs, S.A. Newman, B. Hinner, C.W. Maier, E. Sackmann, Biophys. J. 84 (2003) 1272–1280.
[882] K.S. Weadock, E.J. Miller, E.L. Keuffel, M.G. Dunn, J. Biomed. Mater. Res. 32 (1996) 221–226.
[883] K.S. Weadock, E.J. Miller, L.D. Bellincampi, J.P. Zawadsky, M.G. Dunn, J. Biomed. Mater. Res. 29 (1995) 1373–1379.
[884] T. Zhang, H. Chen, Y. Zhang, Y. Zan, T. Ni, M. Liu, R. Pei, Colloids Surf. B Biointerfaces 174 (2019) 528–535.
[885] W. Lee, V. Lee, S. Polio, P. Keegan, J.-H. Lee, K. Fischer, J.-K. Park, S.-S. Yoo, Biotechnol. Bioeng. 105 (2010) 1178–1186.
[886] P. Gangatirkar, S. Paquet-Fifield, A. Li, R. Rossi, P. Kaur, Nat. Protoc. 2 (2007) 178–186.
[887] P.B. van Wachem, J.A. Plantinga, M.J.B. Wissink, R. Beernink, A.A. Poot, G.H.M. Engbers, T. Beugeling, W.G. van Aken, J. Feijen, M.J.A. van Luyn, J. Biomed. Mater. Res. 55 (2001) 368–378.
[888] C. Mu, K. Zhang, W. Lin, D. Li, J. Biomed. Mater. Res. A. 101A (2013) 385–393.
[889] A. Rajaram, C.C. Chu, Journal of Biomaterials Science, Polymer Edition 1 (1989) 167–172.
[890] M.E. Nimni, D. Cheung, B. Strates, M. Kodama, K. Sheikh, J. Biomed. Mater. Res. 21 (1987) 741–771.
[891] M.G. Yeo, G.H. Kim, Biofabrication 9 (2017) 025004.
[892] J. Lee, M. Yeo, W. Kim, Y. Koo, G.H. Kim, Int. J. Biol. Macromol. 110 (2018) 497–503.
[893] N. Inoue, M. Bessho, M. Furuta, T. Kojima, S. Okuda, M. Hara, J. Biomater. Sci. Polym. Ed. 17 (2006) 837–858.
[894] X. Zhang, L. Xu, X. Huang, S. Wei, M. Zhai, J. Biomed. Mater. Res. A. 100A (2012) 2960–2969.
[895] W. Friess, Eur. J. Pharm. Biopharm. 45 (1998) 113–136.
[896] B. Chevallay, D. Herbage, Med. Biol. Eng. Comput. 38 (2000) 211–218.
[897] T. Yuan, L. Zhang, K. Li, H. Fan, Y. Fan, J. Liang, X. Zhang, J. Biomed. Mater. Res. Part B Appl. Biomater. 102 (2014) 337–344.
[898] L.A. Liotta, K. Tryggvason, S. Garbisa, I. Hart, C.M. Foltz, S. Shafie, Nature 284 (1980) 67–68.
[899] I. Metzmacher, F. Radu, M. Bause, P. Knabner, W. Friess, Eur. J. Pharm. Biopharm. 67 (2007) 349–360.
[900] L. Chen, K. Yang, H. Zhao, A. Liu, W. Tu, C. Wu, S. Chen, Z. Guo, H. Luo, J. Sun, H. Fan, ACS Biomater. Sci. Eng. 5 (2019) 1405–1415.
[901] H. Naito, M. Yoshimura, T. Mizuno, S. Takasawa, T. Tojo, S. Taniguchi, J. Biomed. Mater. Res. A. 101 (2013) 2838–2845.
[902] M.P. Lutolf, F.E. Weber, H.G. Schmoekel, J.C. Schense, T. Kohler, R. Muller, J.A. Hubbell, Nat Biotech 21 (2003) 513–518.
[903] T. Nakaji-Hirabayashi, K. Kato, H. Iwata, Bioconjug. Chem. 24 (2013) 1798–1804.
[904] D.B. Hoban, B. Newland, T.C. Moloney, L. Howard, A. Pandit, E. Dowd, Biomaterials 34 (2013) 9420–9429.
[905] S. Ulrich, R. Lars, A. Stefan, S. Sebastian, F. Florian, R. Johannes, L. Ingo, B. Thomas, R. Maximilian, N. Ulrich, Am. J. Sports Med. 39 (2011) 2558–2565.
[906] X. Chen, F. Zhang, X. He, Y. Xu, Z. Yang, L. Chen, S. Zhou, Y. Yang, Z. Zhou, W. Sheng, Y. Zeng, Injury 44 (2013) 540–549.
[907] L. Zhang, T. Yuan, L. Guo, X. Zhang, J. Biomed. Mater. Res. A. 100A (2012) 2717–2725.
[908] T. Mimura, S. Imai, N. Okumura, L. Li, K. Nishizawa, S. Araki, H. Ueba, M. Kubo, K. Mori, Y. Matsusue, J. Biomed. Mater. Res. Part B Appl. Biomater. 98B (2011) 360–368.
[909] S. Wakitani, T. Kimura, A. Hirooka, T. Ochi, M. Yoneda, N. Yasui, H. Owaki, K. Ono, Journal of Bone & Joint Surgery, British 71-B (1989) 74.
[910] Y. Zhao, Y. Xu, B. Zhang, X. Wu, F. Xu, W. Liang, X. Du, R. Li, Tissue Eng. Part C Methods 16 (2009) 653–659.
[911] A.S. Lee, M. Inayathullah, M.A. Lijkwan, X. Zhao, W. Sun, S. Park, W.X. Hong, M.B. Parekh, A.V. Malkovskiy, E. Lau, X. Qin, V.R. Pothineni, V. Sanchez-Freire, W.Y. Zhang, N.G. Kooreman, A.D. Ebert, C.K.F. Chan, P.K. Nguyen, J. Rajadas, J.C. Wu, Nat. Biomed. Eng. 2 (2018) 104–113.
[912] J.W. Park, Y.D. Kang, J.S. Kim, J.H. Lee, H.-W. Kim, Biochem. Biophys. Res. Commun. 447 (2014) 400–406.
[913] T. Xu, P. Molnar, C. Gregory, M. Das, T. Boland, J.J. Hickman, Biomaterials 30 (2009) 4377–4383.
[914] L. Huang, R. Li, W. Liu, J. Dai, Z. Du, X. Wang, J. Ma, J. Zhao, Neural Regen. Res. 9 (2014) 1371–1378.
[915] S.J. Lee, H.-J. Wang, T.-H. Kim, J.S. Choi, G. Kulkarni, J.D. Jackson, A. Atala, J.J. Yoo, Stem Cells Transl. Med. 7 (2018) 241–250.
[916] J. Huling, S.-i. Min, D.S. Kim, I.K. Ko, A. Atala, J.J. Yoo, Acta Biomater. 95 (2019)

328–336.
[917] R.G. Young, D.L. Butler, W. Weber, A.I. Caplan, S.L. Gordon, D.J. Fink, J. Orthop. Res. 16 (1998) 406–413.
[918] M. Zhong, X. Liu, D. Wei, J. Sun, L. Guo, H. Zhu, Y. Wan, H. Fan, Mater. Sci. Eng. C 101 (2019) 370–379.
[919] D. Olsen, C. Yang, M. Bodo, R. Chang, S. Leigh, J. Baez, D. Carmichael, M. Perälä, E.-R. Hämäläinen, M. Jarvinen, J. Polarek, Adv. Drug Deliv. Rev. 55 (2003) 1547–1567.
[920] E.Y. Egawa, K. Kato, M. Hiraoka, T. Nakaji-Hirabayashi, H. Iwata, Biomaterials 32 (2011) 4737–4743.
[921] S.-A. Oh, H.-Y. Lee, J.H. Lee, T.-H. Kim, J.-H. Jang, H.-W. Kim, I. Wall, Tissue Eng. Part A 18 (2011) 1087–1100.
[922] Y. Zhang, D. Zhu, Y. Wei, Y. Wu, W. Cui, L. Liuqin, G. Fan, Q. Yang, Z. Wang, Z. Xu, D. Kong, L. Zeng, Q. Zhao, Acta Biomater. 86 (2019) 223–234.
[923] W. Liu, M. Griffith, F. Li, J. Mater. Sci. Mater. Med. 19 (2008) 3365–3371.
[924] X. Yu, Q. Yuan, M. Yang, R. Liu, S. Zhu, J. Li, W. Zhang, J. You, S. Xiong, Y. Hu, Macromol. Mater. Eng. 304 (2019) 1800755.
[925] K.S. Rho, L. Jeong, G. Lee, B.-M. Seo, Y.J. Park, S.-D. Hong, S. Roh, J.J. Cho, W.H. Park, B.-M. Min, Biomaterials 27 (2006) 1452–1461.
[926] C. Curtin, J.C. Nolan, R. Conlon, L. Deneweth, C. Gallagher, Y.J. Tan, B.L. Cavanagh, A.Z. Asraf, H. Harvey, S. Miller-Delaney, J. Shohet, I. Bray, F.J. O'Brien, R.L. Stallings, O. Piskareva, Acta Biomater. 70 (2018) 84–97.
[927] D. Yip, C.H. Cho, Biochem. Biophys. Res. Commun. 433 (2013) 327–332.
[928] C.S. Szot, C.F. Buchanan, J.W. Freeman, M.N. Rylander, Tissue Eng. Part C Methods 19 (2013) 864–874.
[929] S. Young, M. Wong, Y. Tabata, A.G. Mikos, J. Control. Release 109 (2005) 256–274.
[930] J. Heino, M. Huhtala, J. Käpylä, M.S. Johnson, Int. J. Biochem. Cell Biol. 41 (2009) 341–348.
[931] P.E. Van den Steen, B. Dubois, I. Nelissen, P.M. Rudd, R.A. Dwek, G. Opdenakker, Crit. Rev. Biochem. Mol. Biol. 37 (2002) 375–536.
[932] K.B. Djagny, Z. Wang, S. Xu, Crit. Rev. Food Sci. Nutr. 41 (2001) 481–492.
[933] K. Yue, G. Trujillo-de Santiago, M.M. Alvarez, A. Tamayol, N. Annabi, A. Khademhosseini, Biomaterials 73 (2015) 254–271.
[934] J.-Y. Lai, Y.-T. Li, Biomacromolecules 11 (2010) 1387–1397.
[935] H.-W. Kang, Y. Tabata, Y. Ikada, Biomaterials 20 (1999) 1339–1344.
[936] N. Reddy, R. Reddy, Q. Jiang, Trends Biotechnol. 33 (2015) 362–369.
[937] Y.Z. Zhang, J. Venugopal, Z.M. Huang, C.T. Lim, S. Ramakrishna, Polymer 47 (2006) 2911–2917.
[938] A.K. Miri, H.G. Hosseinabadi, B. Cecen, S. Hassan, Y.S. Zhang, Acta Biomater. 77 (2018) 38–47.
[939] A.I. Van Den Bulcke, B. Bogdanov, N. De Rooze, E.H. Schacht, M. Cornelissen, H. Berghmans, Biomacromolecules 1 (2000) 31–38.
[940] J.W. Nichol, S.T. Koshy, H. Bae, C.M. Hwang, S. Yamanlar, A. Khademhosseini, Biomaterials 31 (2010) 5536–5544.
[941] A. Sheikhi, J. de Rutte, R. Haghniaz, O. Akouissi, A. Sohrabi, D. Di Carlo, A. Khademhosseini, Biomaterials 192 (2019) 560–568.
[942] D. Loessner, C. Meinert, E. Kaemmerer, L.C. Martine, K. Yue, P.A. Levett, T.J. Klein, F.P.W. Melchels, A. Khademhosseini, D.W. Hutmacher, Nat. Protoc. 11 (2016) 727–746.
[943] G.C.J. Brown, K.S. Lim, B.L. Farrugia, G.J. Hooper, T.B.F. Woodfield, Macromol. Biosci. 17 (2017) 1700158.
[944] D. Hellio, M. Djabourov, Macromol. Symp. 241 (2006) 23–27.
[945] D.M. Kirchmajer, C.A. Watson, M. Ranson, Mih. Panhuis, RSC Adv. 3 (2013) 1073–1081.
[946] A. Bigi, G. Cojazzi, S. Panzavolta, N. Roveri, K. Rubini, Biomaterials 23 (2002) 4827–4832.
[947] X. Zhao, S. Liu, L. Yildirimer, H. Zhao, R. Ding, H. Wang, W. Cui, D. Weitz, Adv. Funct. Mater. 26 (2016) 2809–2819.
[948] J.R. Choi, K.W. Yong, J.Y. Choi, A.C. Cowie, BioTechniques 66 (2019) 40–53.
[949] H.-C. Liang, W.-H. Chang, H.-F. Liang, M.-H. Lee, H.-W. Sung, J. Appl. Polym. Sci. 91 (2004) 4017–4026.
[950] H.-W. Sung, D.-M. Huang, W.-H. Chang, R.-N. Huang, J.-C. Hsu, J. Biomed. Mater. Res. 46 (1999) 520–530.
[951] H.-C. Liang, W.-H. Chang, K.-J. Lin, H.-W. Sung, J. Biomed. Mater. Res. A. 65A (2003) 271–282.
[952] A. Bigi, G. Cojazzi, S. Panzavolta, K. Rubini, N. Roveri, Biomaterials 22 (2001) 763–768.
[953] J. Su, S.C. Satchell, J.A. Wertheim, R.N. Shah, Biomaterials 201 (2019) 99–112.
[954] L. Zhang, J. Liu, X. Zheng, A. Zhang, X. Zhang, K. Tang, Carbohydr. Polym. 216 (2019) 45–53.
[955] Q. Zhao, S. Mu, Y. Long, J. Zhou, W. Chen, D. Astruc, C. Gaidau, H. Gu, Macromol. Mater. Eng. 304 (2019) 1800664.
[956] X. Zhang, M.D. Do, P. Casey, A. Sulistio, G.G. Qiao, L. Lundin, P. Lillford, S. Kosaraju, J. Agric. Food Chem. 58 (2010) 6809–6815.
[957] N. Cao, Y. Fu, J. He, Food Hydrocoll. 21 (2007) 575–584.
[958] M.C. Echave, C. Pimenta-Lopes, J.L. Pedraz, M. Mehrali, A. Dolatshahi-Pirouz, F. Ventura, G. Orive, Int. J. Pharm. 562 (2019) 151–161.
[959] C.-C. Tsai, Y.-J. Hong, R.J. Lee, N.-C. Cheng, J. Yu, J. Mater. Chem. B 7 (2019) 1064–1075.
[960] S. Gorgieva, V. Kokol, Collagen-vs. Gelatine-based Biomaterials and Their Biocompatibility: Review and Perspectives, Biomaterials Applications for Nanomedicine, IntechOpen, 2011, pp. 17–52.
[961] H. Shin, B.D. Olsen, A. Khademhosseini, Biomaterials 33 (2012) 3143–3152.
[962] A. Bigi, S. Panzavolta, K. Rubini, Biomaterials 25 (2004) 5675–5680.
[963] P. Dalev, E. Vassileva, J.E. Mark, S. Fakirov, Biotechnol. Tech. 12 (1998) 889–892.
[964] Q. Feng, J. Xu, K. Zhang, H. Yao, N. Zheng, L. Zheng, J. Wang, K. Wei, X. Xiao, L. Qin, L. Bian, ACS Cent. Sci. 5 (2019) 440–450.
[965] Q. Feng, K. Wei, S. Lin, Z. Xu, Y. Sun, P. Shi, G. Li, L. Bian, Biomaterials 101 (2016) 217–228.
[966] M. Sarem, N. Arya, M. Heizmann, A.T. Neffe, A. Barbero, T.P. Gebauer, I. Martin, A. Lendlein, V.P. Shastri, Acta Biomater. 69 (2018) 83–94.
[967] S.M. Choi, K.-M. Lee, S.B. Ryu, Y.J. Park, Y.G. Hwang, D. Baek, Y. Choi, K.H. Park, K.D. Park, J.W. Lee, Cell Death Dis. 9 (2018) 866.
[968] I. Otto, R. Levato, W. Webb, I. Khan, C. Breugem, J. Malda, Eur. Cell. Mater. 35 (2018) 132.
[969] S. Pahoff, C. Meinert, O. Bas, L. Nguyen, T.J. Klein, D.W. Hutmacher, J. Mater. Chem. B 7 (2019) 1761–1772.
[970] W. Schuurman, P.A. Levett, M.W. Pot, P.R. van Weeren, W.J.A. Dhert, D.W. Hutmacher, F.P.W. Melchels, T.J. Klein, J. Malda, Macromol. Biosci. 13 (2013) 551–561.
[971] E. Shirzaei Sani, A. Kheirkhah, D. Rana, Z. Sun, W. Foulsham, A. Sheikhi, A. Khademhosseini, R. Dana, N. Annabi, Sci. Adv. 5 (2019) eaav1281.
[972] B.W. Walker, R.P. Lara, C. Yu, E.S. Sani, W. Kimball, S. Joyce, N. Annabi, Biomaterials 207 (2019) 89–101.
[973] J.-Y. Lai, P.-K. Lin, G.-H. Hsiue, H.-Y. Cheng, S.-J. Huang, Y.-T. Li, Biomacromolecules 10 (2009) 310–319.
[974] Y. Dong, S.A.M. Rodrigues, X. Li, S.H. Kwon, N. Kosaric, S. Khong, Y. Gao, W. Wang, G.C. Gurtner, Adv. Funct. Mater. 27 (2017) 1606619.
[975] A.O. Elzoghby, J. Control. Release 172 (2013) 1075–1091.
[976] T. Tanaka, T. Matsushita, K. Nishida, K. Takayama, K. Nagai, D. Araki, T. Matsumoto, Y. Tabata, R. Kuroda, J. Tissue Eng. Regen. Med. 13 (2019) 423–432.
[977] K.W. Leong, H.Q. Mao, V.L. Truong-Le, K. Roy, S.M. Walsh, J.T. August, J. Control. Release 53 (1998) 183–193.
[978] M. Yamamoto, Y. Ikada, Y. Tabata, J. Biomater. Sci. Polym. Ed. 12 (2001) 77–88.
[979] C. Amit, S. Muralikumar, S. Janaki, M. Lakshmipathy, K.L. Therese, V. Umashankar, P. Padmanabhan, J. Narayanan, Int. J. Nanomedicine 14 (2019) 605–622.
[980] Y. Tabata, Y. Ikada, Adv. Drug Deliv. Rev. 31 (1998) 287–301.
[981] K. Yamashita, S. Tsunoda, S. Gunji, T. Murakami, T. Suzuki, Y. Tabata, Y. Sakai, Surg. Today 49 (2019) 785–794.
[982] M.T. Ngo, B.A.C. Harley, Biomaterials 198 (2019) 122–134.
[983] F.G. Omenetto, D.L. Kaplan, Science 329 (2010) 528–531.
[984] C. Holland, K. Numata, J. Rnjak-Kovacina, F.P. Seib, Adv. Healthc. Mater. 8 (2019) 1800465.
[985] G.H. Altman, F. Diaz, C. Jakuba, T. Calabro, R.L. Horan, J. Chen, H. Lu, J. Richmond, D.L. Kaplan, Biomaterials 24 (2003) 401–416.
[986] Y. Wang, H.-J. Kim, G. Vunjak-Novakovic, D.L. Kaplan, Biomaterials 27 (2006) 6064–6082.
[987] Y. Wang, J. Guo, L. Zhou, C. Ye, F.G. Omenetto, D.L. Kaplan, S. Ling, Adv. Funct. Mater. 28 (2018) 1805305.
[988] Y.-Q. Zhang, Biotechnol. Adv. 20 (2002) 91–100.
[989] W. Huang, S. Ling, C. Li, F.G. Omenetto, D.L. Kaplan, Chem. Soc. Rev. 47 (2018) 6486–6504.
[990] S.C. Kundu, B.C. Dash, R. Dash, D.L. Kaplan, Prog. Polym. Sci. 33 (2008) 998–1012.
[991] D.N. Rockwood, R.C. Preda, T. Yücel, X. Wang, M.L. Lovett, D.L. Kaplan, Nat. Protoc. 6 (2011) 1612–1631.
[992] M. Heim, D. Keerl, T. Scheibel, Angew. Chemie Int. Ed. 48 (2009) 3584–3596.
[993] O. Tokareva, M. Jacobsen, M. Buehler, J. Wong, D.L. Kaplan, Acta Biomater. 10 (2014) 1612–1626.
[994] Z. Shao, F. Vollrath, Nature 418 (2002) 741-741.
[995] A. Lazaris, S. Arcidiacono, Y. Huang, J.-F. Zhou, F. Duguay, N. Chretien, E.A. Welsh, J.W. Soares, C.N. Karatzas, Science 295 (2002) 472–476.
[996] J. Huang, C. Wong, A. George, D.L. Kaplan, Biomaterials 28 (2007) 2358–2367.
[997] M.M. Moisenovich, O. Pustovalova, J. Shackelford, T.V. Vasiljeva, T.V. Druzhinina, Y.A. Kamenchuk, V.V. Guzeev, O.S. Sokolova, V.G. Bogush, V.G. Debabov, M.P. Kirpichnikov, I.I. Agapov, Biomaterials 33 (2012) 3887–3898.
[998] M. Widhe, H. Bysell, S. Nystedt, I. Schenning, M. Malmsten, J. Johansson, A. Rising, M. Hedhammar, Biomaterials 31 (2010) 9575–9585.
[999] R. Jansson, N. Thatikonda, D. Lindberg, A. Rising, J. Johansson, P.-Å. Nygren, M. Hedhammar, Biomacromolecules 15 (2014) 1696–1706.
[1000] K. Spiess, R. Ene, C.D. Keenan, J. Senker, F. Kremer, T. Scheibel, J. Mater. Chem. 21 (2011) 13594–13604.
[1001] T.B. Aigner, E. DeSimone, T. Scheibel, Adv. Mater. 30 (2018) 1704636.
[1002] C. Wong Po Foo, D.L. Kaplan, Adv. Drug Deliv. Rev. 54 (2002) 1131–1143.
[1003] M. Widhe, U. Johansson, C.-O. Hillerdahl, M. Hedhammar, Biomaterials 34 (2013) 8223–8234.
[1004] C. Vendrely, T. Scheibel, Macromol. Biosci. 7 (2007) 401–409.
[1005] B. Kundu, N.E. Kurland, S. Bano, C. Patra, F.B. Engel, V.K. Yadavalli, S.C. Kundu, Prog. Polym. Sci. 39 (2014) 251–267.
[1006] S. Kapoor, S.C. Kundu, Acta Biomater. 31 (2016) 17–32.
[1007] H.-Y. Wang, Y.-Q. Zhang, Biotechnol. Prog. 31 (2015) 630–640.
[1008] A.R. Murphy, P.S. John, D.L. Kaplan, Biomaterials 29 (2008) 2829–2838.
[1009] I.V. Bessonov, Y.A. Rochev, A.Y. Arkhipova, M.N. Kopitsyna, D.V. Bagrov, E.A. Karpushkin, T.N. Bibikova, A.M. Moysenovich, A.S. Soldatenko, I.I. Nikishin, M.S. Kotliarova, V.G. Bogush, K.V. Shaitan, M.M. Moisenovich, Biomed. Mater. 14 (2019) 034102.
[1010] X. Wu, J. Hou, M. Li, J. Wang, D.L. Kaplan, S. Lu, Acta Biomater. 8 (2012) 2185–2192.
[1011] X. Wang, J.A. Kluge, G.G. Leisk, D.L. Kaplan, Biomaterials 29 (2008) 1054–1064.

[1012] W. Zhang, X. Wang, S. Wang, J. Zhao, L. Xu, C. Zhu, D. Zeng, J. Chen, Z. Zhang, D.L. Kaplan, X. Jiang, Biomaterials 32 (2011) 9415–9424.
[1013] T. Yucel, P. Cebe, D.L. Kaplan, Biophys. J. 97 (2009) 2044–2050.
[1014] V. Ribeiro, S. Pina, J.B. Costa, I.F. Cengiz, L. García-Fernández, Md.M. Fernández-Gutiérrez, O.C. Paiva, A.L. Oliveira, J. San Roman, J.M. Oliveira, R.L. Reis, ACS Appl. Mater. Interfaces 11 (2019) 3781–3799.
[1015] R. Carvalho Cristiana, B. Costa João, A. da Silva Morais, R. López-Cebral, J. Silva-Correia, L. Reis Rui, J.M. Oliveira, Adv. Healthc. Mater. 7 (2018) 1800186.
[1016] M. McGill, J.M. Coburn, B.P. Partlow, X. Mu, D.L. Kaplan, Acta Biomater. 63 (2017) 76–84.
[1017] B.P. Partlow, C.W. Hanna, J. Rnjak-Kovacina, J.E. Moreau, M.B. Applegate, K.A. Burke, B. Marelli, A.N. Mitropoulos, F.G. Omenetto, D.L. Kaplan, Adv. Funct. Mater. 24 (2014) 4615–4624.
[1018] S. Rammensee, D. Huemmerich, K.D. Hermanson, T. Scheibel, A.R. Bausch, Appl. Phys. A 82 (2005) 261.
[1019] Z. Wang, Y. Zhang, J. Zhang, L. Huang, J. Liu, Y. Li, G. Zhang, S.C. Kundu, L. Wang, Sci. Rep. 4 (2014) 7064.
[1020] A.R. Franco, E. Palma Kimmerling, C. Silva, F.J. Rodrigues, I.B. Leonor, R.L. Reis, D.L. Kaplan, Macromol. Biosci. 18 (2018) 1800262.
[1021] K. Schacht, T. Scheibel, Biomacromolecules 12 (2011) 2488–2495.
[1022] C. Li, T. Luo, Z. Zheng, A.R. Murphy, X. Wang, D.L. Kaplan, Acta Biomater. 11 (2015) 222–232.
[1023] L. Shi, F. Wang, W. Zhu, Z. Xu, S. Fuchs, J. Hilborn, L. Zhu, Q. Ma, Y. Wang, X. Weng, A. Ossipov Dmitri, Adv. Funct. Mater. 27 (2017) 1700591.
[1024] Y. Yan, B. Cheng, K. Chen, W. Cui, J. Qi, X. Li, L. Deng, Adv. Healthc. Mater. 8 (2019) 1801043.
[1025] M. Fini, A. Motta, P. Torricelli, G. Giavaresi, N. Nicoli Aldini, M. Tschon, R. Giardino, C. Migliaresi, Biomaterials 26 (2005) 3527–3536.
[1026] S. Sofia, M.B. McCarthy, G. Gronowicz, D.L. Kaplan, J. Biomed. Mater. Res. 54 (2001) 139–148.
[1027] T. Diab, E.M. Pritchard, B.A. Uhrig, J.D. Boerckel, D.L. Kaplan, R.E. Guldberg, J. Mech. Behav. Biomed. Mater. 11 (2012) 123–131.
[1028] E. James, E. Van Doren, C. Li, D.L. Kaplan, Tissue Eng. Part A 25 (2019) 12–23.
[1029] V.P. Ribeiro, A. da Silva Morais, F.R. Maia, R.F. Canadas, J.B. Costa, A.L. Oliveira, J.M. Oliveira, R.L. Reis, Acta Biomater. 72 (2018) 167–181.
[1030] S. Yodmuang, S.L. McNamara, A.B. Nover, B.B. Mandal, M. Agarwal, T.-A.N. Kelly, P.-h.G. Chao, C. Hung, D.L. Kaplan, G. Vunjak-Novakovic, Acta Biomater. 11 (2015) 27–36.
[1031] M. Farokhi, F. Mottaghitalab, Y. Fatahi, M.R. Saeb, P. Zarrintaj, S.C. Kundu, A. Khademhosseini, Eur. Polym. J. 115 (2019) 251–267.
[1032] P.-H.G. Chao, S. Yodmuang, X. Wang, L. Sun, D.L. Kaplan, G. Vunjak-Novakovic, J. Biomed. Mater. Res. Part B Appl. Biomater. 95B (2010) 84–90.
[1033] S. Hofmann, S. Knecht, R. Langer, D.L. Kaplan, G. Vunjak-Novakovic, H.P. Merkle, L. Meinel, Tissue Eng. 12 (2006) 2729–2738.
[1034] J.E. Brown, B.P. Partlow, A.M. Berman, M.D. House, D.L. Kaplan, Am. J. Obstet. Gynecol. 214 (2016) 118.e111-118.e119.
[1035] B.K. Bhunia, B.B. Mandal, ACS Biomater. Sci. Eng. 5 (2019) 870–886.
[1036] N.E. Davis, L.N. Beenken-Rothkopf, A. Mirsoian, N. Kojic, D.L. Kaplan, A.E. Barron, M.J. Fontaine, Biomaterials 33 (2012) 6691–6697.
[1037] N.D. Shalaly, M. Ria, U. Johansson, K. Åvall, P.-O. Berggren, M. Hedhammar, Biomaterials 90 (2016) 50–61.
[1038] A.M. Hopkins, L. De Laporte, F. Tortelli, E. Spedden, C. Staii, T.J. Atherton, J.A. Hubbell, D.L. Kaplan, Adv. Funct. Mater. 23 (2013) 5140–5149.
[1039] M. Lewicka, O. Hermanson, A.U. Rising, Biomaterials 33 (2012) 7712–7717.
[1040] X. Leng, B. Liu, B. Su, M. Liang, L. Shi, S. Li, S. Qu, X. Fu, Y. Liu, M. Yao, D.L. Kaplan, Y. Wang, X. Wang, J. Tissue Eng. Regen. Med. 11 (2017) 822–830.
[1041] K. Numata, D.L. Kaplan, Adv. Drug Deliv. Rev. 62 (2010) 1497–1508.
[1042] K. Numata, S. Yamazaki, N. Naga, Biomacromolecules 13 (2012) 1383–1389.
[1043] A. Lammel, M. Schwab, M. Hofer, G. Winter, T. Scheibel, Biomaterials 32 (2011) 2233–2240.
[1044] C. Herrera-Rincon, A.S. Golding, K.M. Moran, C. Harrison, C.J. Martyniuk, J.A. Guay, J. Zaltsman, H. Carabello, D.L. Kaplan, M. Levin, Cell Rep. 25 (2018) 1593-1609.e1597.
[1045] J.G. Hardy, A. Leal-Egaña, T.R. Scheibel, Macromol. Biosci. 13 (2013) 1431–1437.
[1046] X. Xie, L. Liu, Z. Zheng, Z. Han, M. Zhi, D.L. Kaplan, G. Li, X. Wang, Macromol. Biosci. 18 (2018) 1700384.
[1047] E.M. Pritchard, T. Valentin, B. Panilaitis, F. Omenetto, D.L. Kaplan, Adv. Funct. Mater. 23 (2013) 854–861.
[1048] N. Hassani Besheli, F. Mottaghitalab, M. Eslami, M. Gholami, S.C. Kundu, D.L. Kaplan, M. Farokhi, ACS Appl. Mater. Interfaces 9 (2017) 5128–5138.
[1049] N. Guziewicz, A. Best, B. Perez-Ramirez, D.L. Kaplan, Biomaterials 32 (2011) 2642–2650.
[1050] B. Yavuz, J.L. Morgan, C. Herrera, K. Harrington, B. Perez-Ramirez, P.J. LiWang, D.L. Kaplan, J. Control. Release 301 (2019) 1–12.
[1051] K. Numata, D.L. Kaplan, Biomacromolecules 11 (2010) 3189–3195.
[1052] K. Numata, J. Hamasaki, B. Subramanian, D.L. Kaplan, J. Control. Release 146 (2010) 136–143.
[1053] M.L. Lovett, X. Wang, T. Yucel, L. York, M. Keirstead, L. Haggerty, D.L. Kaplan, Eur. J. Pharm. Biopharm. 95 (2015) 271–278.
[1054] M. Hofer, G. Winter, J. Myschik, Biomaterials 33 (2012) 1554–1562.
[1055] M. Farokhi, F. Mottaghitalab, Y. Fatahi, A. Khademhosseini, D.L. Kaplan, Trends Biotechnol. 36 (2018) 907–922.
[1056] F.P. Seib, E.M. Pritchard, D.L. Kaplan, Adv. Funct. Mater. 23 (2013) 58–65.
[1057] V.P. Ribeiro, J. Silva-Correia, C. Gonçalves, S. Pina, H. Radhouani, T. Montonen, J. Hyttinen, A. Roy, A.L. Oliveira, R.L. Reis, J.M. Oliveira, PLoS One 13 (2018) e0194441.
[1058] B. Yavuz, J. Zeki, J. Taylor, K. Harrington, J.M. Coburn, N. Ikegaki, D.L. Kaplan, B. Chiu, J. Pharm. Sci. 108 (2019) 2748–2755.
[1059] C.M. R, M.F. Raquel, V. Sílvia, R.R. L, O.J. M, Glob. Chall. 2 (2018) 1700100.
[1060] M.C. Hacker, J. Krieghoff, A.G. Mikos, A. Atala, R. Lanza, A.G. Mikos, R. Nerem (Eds.), Principles of Regenerative Medicine, third edition, Academic Press, Boston, 2019, pp. 559–590.
[1061] I. Gibas, H. Janik, Chem. Chem. Technol. 4 (2010) 297–304.
[1062] H. Yuk, T. Zhang, S. Lin, G.A. Parada, X. Zhao, Nat. Mater. 15 (2015) 190–196.
[1063] J.-Y. Sun, C. Keplinger, G.M. Whitesides, Z. Suo, Adv. Mater. 26 (2014) 7608–7614.
[1064] P. Morelle Xavier, R. Illeperuma Widusha, K. Tian, R. Bai, Z. Suo, J. Vlassak Joost, Adv. Mater. 30 (2018) 1801541.
[1065] Y. Jiawei, B. Ruobing, S. Zhigang, Adv. Mater. 30 (2018) 1800671.
[1066] Y. Gao, K. Wu, Z. Suo, Adv. Mater. 31 (2019) 1806948.
[1067] F. Chen, D. Zhou, J. Wang, T. Li, X. Zhou, T. Gan, S. Handschuh-Wang, X. Zhou, Angew. Chemie Int. Ed. 57 (2018) 6568–6571.
[1068] B. Trappmann, J.E. Gautrot, J.T. Connelly, D.G.T. Strange, Y. Li, M.L. Oyen, M.A. Cohen Stuart, H. Boehm, B. Li, V. Vogel, J.P. Spatz, F.M. Watt, W.T.S. Huck, Nat. Mater. 11 (2012) 642–649.
[1069] C.E. Kandow, P.C. Georges, P.A. Janmey, K.A. Beningo, Methods Cell Biol. 83 (2007) 29–46. Academic Press.
[1070] D.E. Discher, P. Janmey, Y.-l. Wang, Science 310 (2005) 1139–1143.
[1071] A.J. Engler, S. Sen, H.L. Sweeney, D.E. Discher, Cell 126 (2006) 677–689.
[1072] W. de Cássia Novaes, A. Berg, Aesthetic Plast. Surg. 27 (2003) 376–380.
[1073] N.-x. Cheng, Y.-l. Wang, J.-h. Wang, X.-m. Zhang, H. Zhong, Aesthetic Plast. Surg. 26 (2002) 375–382.
[1074] L. Christensen, L. Camitz, K.E. Illigen, M. Hansen, R. Sarvaa, P.G. Conaghan, Osteoarthr. Cartil. 24 (2016) 1999–2002.
[1075] J.P. Lee, E. Kassianidou, J.I. MacDonald, M.B. Francis, S. Kumar, Biomaterials 102 (2016) 268–276.
[1076] H. Kaşgöz, S. Özgümüş, M. Orbay, Polymer 44 (2003) 1785–1793.
[1077] R.H. Pelton, J. Polym. Sci.: Polym. Chem. Ed. 22 (1984) 3955–3966.
[1078] H. Tanaka, J. Polym. Sci.: Polym. Chem. Ed. 17 (1979) 1239–1245.
[1079] H. Tanaka, L. Ödberg, J. Polym. Sci. Part A: Polym. Chem. 27 (1989) 4329–4339.
[1080] H.J. Naghash, O. Okay, J. Appl. Polym. Sci. 60 (1996) 971–979.
[1081] N. Tang, Z. Peng, R. Guo, M. An, X. Chen, X. Li, N. Yang, J. Zang, Polymers 9 (2017) 688.
[1082] L.H. Christensen, J.B. Nielsen, L. Mouritsen, M. Sørensen, G. Lose, Dermatol. Surg. 34 (2008) S68–S77.
[1083] M.J. Caulfield, X. Hao, G.G. Qiao, D.H. Solomon, Polymer 44 (2003) 1331–1337.
[1084] S.R. Caliari, J.A. Burdick, Nat. Methods 13 (2016) 405–414.
[1085] X. Liu, J. Zhang, M. Fadeev, Z. Li, V. Wulf, H. Tian, I. Willner, Chem. Sci. 10 (2019) 1008–1016.
[1086] S. Nazari Pour, S.V. Ghugare, R. Wiens, K. Gough, S. Liu, Appl. Surf. Sci. 349 (2015) 695–704.
[1087] V. Breiting, A. Aasted, A. Jørgensen, P. Opitz, A. Rosetzsky, Aesthetic Plast. Surg. 28 (2004) 45–53.
[1088] Y. Huang, M. Zhong, F. Shi, X. Liu, Z. Tang, Y. Wang, Y. Huang, H. Hou, X. Xie, C. Zhi, Angew. Chemie Int. Ed. 56 (2017) 9141–9145.
[1089] C.-C. Kim, H.-H. Lee, K.H. Oh, J.-Y. Sun, Science 353 (2016) 682–687.
[1090] A. Sidorenko, T. Krupenkin, A. Taylor, P. Fratzl, J. Aizenberg, Science 315 (2007) 487–490.
[1091] M. Bassil, J. Davenas, M. El Tahchi, Sens. Actuators B Chem. 134 (2008) 496–501.
[1092] Cdl.H. Alarcon, S. Pennadam, C. Alexander, Chem. Soc. Rev. 34 (2005) 276–285.
[1093] M. Heskins, J.E. Guillet, J. Macromol. Sci. Part A - Chem. 2 (1968) 1441–1455.
[1094] Z. Yang, M. Layani, W. Shancheng, H. Peng, K. Yujie, S. Magdassi, L. Yi, Adv. Funct. Mater. 28 (2018) 1705365.
[1095] W. Wang, X. Fan, F. Li, J. Qiu, M. Umair Malik, W. Ren, B. Ju, S. Zhang, B. Tang, Adv. Opt. Mater. 6 (2017) 1701093.
[1096] X.-H. Li, C. Liu, S.-P. Feng, N.X. Fang, Joule 3 (2019) 290–302.
[1097] K. Matsubara, M. Watanabe, Y. Takeoka, Angew. Chemie Int. Ed. 46 (2007) 1688–1692.
[1098] Q. Yu, L.M. Johnson, G.P. López, Adv. Funct. Mater. 24 (2014) 3751–3759.
[1099] G. Pan, Q. Guo, Y. Ma, H. Yang, B. Li, Angew. Chemie Int. Ed. 52 (2013) 6907–6911.
[1100] T. Sun, G. Wang, L. Feng, B. Liu, Y. Ma, L. Jiang, D. Zhu, Angew. Chemie Int. Ed. 43 (2003) 357–360.
[1101] Y. Akiyama, M. Matsuyama, M. Yamato, N. Takeda, T. Okano, Biomacromolecules 19 (2018) 4014–4022.
[1102] M.A. Akimoto, H.E. Niitsu, K. Nagase, T. Okano, H. Kanazawa, R. Yoshida, Int. J. Mol. Sci. 19 (2018) 1253.
[1103] Y. Zhao, Y. Wu, L. Wang, M. Zhang, X. Chen, M. Liu, J. Fan, J. Liu, F. Zhou, Z. Wang, Nat. Commun. 8 (2017) 2218.
[1104] Y. Zhu, Z. Cankova, M. Iwanaszko, S. Lichtor, M. Mrksich, G.A. Ameer, Proc. Natl. Acad. Sci. 115 (2018) 6816–6821.
[1105] R.A. Stile, K.E. Healy, Biomacromolecules 2 (2001) 185–194.
[1106] M. Yamato, M. Utsumi, A. Kushida, C. Konno, A. Kikuchi, T. Okano, Tissue Eng. 7 (2001) 473–480.
[1107] K. Nagase, M. Yamato, H. Kanazawa, T. Okano, Biomaterials 153 (2018) 27–48.
[1108] N. Matsuda, T. Shimizu, M. Yamato, T. Okano, Adv. Mater. 19 (2007) 3089–3099.
[1109] M. Yamato, Y. Akiyama, J. Kobayashi, J. Yang, A. Kikuchi, T. Okano, Prog. Polym. Sci. 32 (2007) 1123–1133.
[1110] R.M.P. da Silva, J.F. Mano, R.L. Reis, Trends Biotechnol. 25 (2007) 577–583.

[1111] A.K.A.S. Brun-Graeppi, C. Richard, M. Bessodes, D. Scherman, O.-W. Merten, Prog. Polym. Sci. 35 (2010) 1311–1324.
[1112] Z.M.O. Rzaev, S. Dinçer, E. Pişkin, Prog. Polym. Sci. 32 (2007) 534–595.
[1113] M. Najafi, E. Hebels, W.E. Hennink, T. Vermonden, Poly(N-isopropylacrylamide): Physicochemical Properties and Biomedical Applications, Temperature-Responsive Polymers, John Wiley & Sons Ltd., 2018, pp. 1–34.
[1114] F.L. Buchholz, Preparation Methods of Superabsorbent Polyacrylates, Superabsorbent Polymers vol 573, American Chemical Society, 1994, pp. 27–38.
[1115] Q. Wang, J.L. Mynar, M. Yoshida, E. Lee, M. Lee, K. Okuro, K. Kinbara, T. Aida, Nature 463 (2010) 339–343.
[1116] F. Chen, P.W. Tillberg, E.S. Boyden, Science 347 (2015) 543–548.
[1117] A.T. Wassie, Y. Zhao, E.S. Boyden, Nat. Methods 16 (2019) 33–41.
[1118] D. Gambarotto, F.U. Zwettler, M. Le Guennec, M. Schmidt-Cernohorska, D. Fortun, S. Borgers, J. Heine, J.-G. Schloetel, M. Reuss, M. Unser, E.S. Boyden, M. Sauer, V. Hamel, P. Guichard, Nat. Methods 16 (2019) 71–74.
[1119] R. Gao, S.M. Asano, S. Upadhyayula, I. Pisarev, D.E. Milkie, T.-L. Liu, V. Singh, A. Graves, G.H. Huynh, Y. Zhao, J. Bogovic, J. Colonell, C.M. Ott, C. Zugates, S. Tappan, A. Rodriguez, K.R. Mosaliganti, S.-H. Sheu, H.A. Pasolli, S. Pang, C.S. Xu, S.G. Megason, H. Hess, J. Lippincott-Schwartz, A. Hantman, G.M. Rubin, T. Kirchhausen, S. Saalfeld, Y. Aso, E.S. Boyden, E. Betzig, Science 363 (2019) eaau8302.
[1120] P.W. Tillberg, F. Chen, K.D. Piatkevich, Y. Zhao, C.-C. Yu, B.P. English, L. Gao, A. Martorell, H.-J. Suk, F. Yoshida, E.M. DeGennaro, D.H. Roossien, G. Gong, U. Seneviratne, S.R. Tannenbaum, R. Desimone, D. Cai, E.S. Boyden, Nat. Biotechnol. 34 (2016) 987–992.
[1121] F. Chen, A.T. Wassie, A.J. Cote, A. Sinha, S. Alon, S. Asano, E.R. Daugharthy, J.-B. Chang, A. Marblestone, G.M. Church, A. Raj, E.S. Boyden, Nat. Methods 13 (2016) 679–684.
[1122] J.-B. Chang, F. Chen, Y.-G. Yoon, E.E. Jung, H. Babcock, J.S. Kang, S. Asano, H.-J. Suk, N. Pak, P.W. Tillberg, A.T. Wassie, D. Cai, E.S. Boyden, Nat. Methods 14 (2017) 593–599.
[1123] Y. Zhao, O. Bucur, H. Irshad, F. Chen, A. Weins, A.L. Stancu, E.-Y. Oh, M. DiStasio, V. Torous, B. Glass, I.E. Stillman, S.J. Schnitt, A.H. Beck, E.S. Boyden, Nat. Biotechnol. 35 (2017) 757–764.
[1124] T.J. Chozinski, A.R. Halpern, H. Okawa, H.-J. Kim, G.J. Tremel, R.O.L. Wong, J.C. Vaughan, Nat. Methods 13 (2016) 485–488.
[1125] D. Oran, S.G. Rodriques, R. Gao, S. Asano, M.A. Skylar-Scott, F. Chen, P.W. Tillberg, A.H. Marblestone, E.S. Boyden, Science 362 (2018) 1281–1285.
[1126] A.A. D'souza, R. Shegokar, Expert Opin. Drug Deliv. 13 (2016) 1257–1275.
[1127] J.M. Harris, J. Macromol. Sci. Part C- Polym. Rev. 25 (1985) 325–373.
[1128] D.J. Gravert, K.D. Janda, Chem. Rev. 97 (1997) 489–510.
[1129] K.D. Janda, H. Han, Combinatorial Chemistry: A Liquid-Phase Approach, Methods in Enzymology vol 267, Academic Press, 1996, pp. 234–247.
[1130] J.L. West, J.A. Hubbell, React. Polym. 25 (1995) 139–147.
[1131] W.R. Gombotz, W. Guanghui, T.A. Horbett, A.S. Hoffman, J. Biomed. Mater. Res. 25 (1991) 1547–1562.
[1132] S.I. Jeon, J.H. Lee, J.D. Andrade, P.G. De Gennes, J. Colloid Interface Sci. 142 (1991) 149–158.
[1133] S.I. Jeon, J.D. Andrade, J. Colloid Interface Sci. 142 (1991) 159–166.
[1134] M. Zhang, T. Desai, M. Ferrari, Biomaterials 19 (1998) 953–960.
[1135] D.L. Hern, J.A. Hubbell, J. Biomed. Mater. Res. 39 (1998) 266–276.
[1136] F.M. Veronese, G. Pasut, Drug Discov. Today 10 (2005) 1451–1458.
[1137] T. Yamaoka, Y. Tabata, Y. Ikada, J. Pharm. Sci. 83 (1994) 601–606.
[1138] K.-S. Kim, S. Chung, I.-J. Chin, M.-N. Kim, J.-S. Yoon, J. Appl. Polym. Sci. 72 (1999) 341–348.
[1139] A. Lucke, J. Teßmar, E. Schnell, G. Schmeer, A. Göpferich, Biomaterials 21 (2000) 2361–2370.
[1140] I.O. Barcellos, V. Soldi, A.T. Nunes Pires, Polymer 39 (1998) 6073–6078.
[1141] J.K. Tessmar, A.M. Göpferich, Macromol. Biosci. 7 (2007) 23–39.
[1142] J.M. Harris, Poly (ethylene Glycol) Chemistry: Biotechnical and Biomedical Applications, Springer Science & Business Media, 2013.
[1143] J. Klier, A.B. Scranton, N.A. Peppas, Macromolecules 23 (1990) 4944–4949.
[1144] T. Cai, G. Wang, S. Thompson, M. Marquez, Z. Hu, Macromolecules 41 (2008) 9508–9512.
[1145] A.M. Kloxin, M.W. Tibbitt, K.S. Anseth, Nat. Protoc. 5 (2010) 1867–1887.
[1146] J.D. Clapper, C.A. Guymon, Macromolecules 40 (2007) 1101–1107.
[1147] S. Lu, K.S. Anseth, Macromolecules 33 (2000) 2509–2515.
[1148] A. Sannino, P.A. Netti, M. Madaghiele, V. Coccoli, A. Luciani, A. Maffezzoli, L. Nicolais, J. Biomed. Mater. Res. A. 79A (2006) 229–236.
[1149] C.N. Salinas, K.S. Anseth, J. Tissue Eng. Regen. Med. 2 (2008) 296–304.
[1150] N.C. Padmavathi, P.R. Chatterji, Macromolecules 29 (1996) 1976–1979.
[1151] H. Bodugoz, O. Güven, N.A. Peppas, J. Appl. Polym. Sci. 103 (2007) 432–441.
[1152] S.H. Foulger, P. Jiang, A.C. Lattam, D.W. Smith, J. Ballato, Langmuir 17 (2001) 6023–6026.
[1153] S. Lin-Gibson, R.L. Jones, N.R. Washburn, F. Horkay, Macromolecules 38 (2005) 2897–2902.
[1154] J.-H. Kim, J.-G. Kim, D. Kim, Y.H. Kim, J. Appl. Polym. Sci. 96 (2005) 56–61.
[1155] A. Revzin, R.J. Russell, V.K. Yadavalli, W.-G. Koh, C. Deister, D.D. Hile, M.B. Mellott, M.V. Pishko, Langmuir 17 (2001) 5440–5447.
[1156] S. Zalipsky, Bioconjug. Chem. 6 (1995) 150–165.
[1157] J. Zhu, Biomaterials 31 (2010) 4639–4656.
[1158] J.C. Grim, T.E. Brown, B.A. Aguado, D.A. Chapnick, A.L. Viert, X. Liu, K.S. Anseth, ACS Cent. Sci. 4 (2018) 909–916.
[1159] S. Tang, H. Ma, H.-C. Tu, H.-R. Wang, P.-C. Lin, K.S. Anseth, Adv. Sci. 5 (2018) 1800638.
[1160] B.P. Lee, J.L. Dalsin, P.B. Messersmith, Biomacromolecules 3 (2002) 1038–1047.
[1161] A.K. Gaharwar, P. Schexnailder, V. Kaul, O. Akkus, D. Zakharov, S. Seifert, G. Schmidt, Adv. Funct. Mater. 20 (2010) 429–436.
[1162] A.K. Gaharwar, P.J. Schexnailder, B.P. Kline, G. Schmidt, Acta Biomater. 7 (2011) 568–577.
[1163] S.J. Bryant, T.T. Chowdhury, D.A. Lee, D.L. Bader, K.S. Anseth, Ann. Biomed. Eng. 32 (2004) 407–417.
[1164] T.E. Brown, B.J. Carberry, B.T. Worrell, O.Y. Dudaryeva, M.K. McBride, C.N. Bowman, K.S. Anseth, Biomaterials 178 (2018) 496–503.
[1165] G.M. Cruise, D.S. Scharp, J.A. Hubbell, Biomaterials 19 (1998) 1287–1294.
[1166] O.S. Fenton, J.L. Andresen, M. Paolini, R. Langer, Angew. Chemie 130 (2018) 16258–16261.
[1167] M.A. Azagarsamy, K.S. Anseth, ACS Macro Lett. 2 (2013) 5–9.
[1168] B.V. Sridhar, J.R. Janczy, Ø. Hatlevik, G. Wolfson, K.S. Anseth, M.W. Tibbitt, Biomacromolecules 19 (2018) 740–747.
[1169] C.A. DeForest, K.S. Anseth, Nat. Chem. 3 (2011) 925–931.
[1170] A.S. Sawhney, C.P. Pathak, J.A. Hubbell, Biomaterials 14 (1993) 1008–1016.
[1171] A.M. Kloxin, M.W. Tibbitt, A.M. Kasko, J.A. Fairbairn, K.S. Anseth, Adv. Mater. 22 (2010) 61–66.
[1172] T.E. Brown, I.A. Marozas, K.S. Anseth, Adv. Mater. 29 (2017) 1605001.
[1173] A.M. Kloxin, A.M. Kasko, C.N. Salinas, K.S. Anseth, Science 324 (2009) 59–63.
[1174] S.P. Zustiak, J.B. Leach, Biomacromolecules 11 (2010) 1348–1357.
[1175] W.M. Han, S.E. Anderson, M. Mohiuddin, D. Barros, S.A. Nakhai, E. Shin, I.F. Amaral, A.P. Pêgo, A.J. García, Y.C. Jang, Sci. Adv. 4 (2018) eaar4008.
[1176] S.R. Peyton, C.B. Raub, V.P. Keschrumrus, A.J. Putnam, Biomaterials 27 (2006) 4881–4893.
[1177] T.E. Brown, J.S. Silver, B.T. Worrell, I.A. Marozas, F.M. Yavitt, K.A. Günay, C.N. Bowman, K.S. Anseth, J. Am. Chem. Soc. 140 (2018) 11585–11588.
[1178] F. Yang, J. Wang, J. Hou, H. Guo, C. Liu, Biomaterials 34 (2013) 1514–1528.
[1179] F. Yang, C.G. Williams, D.-a. Wang, H. Lee, P.N. Manson, J. Elisseeff, Biomaterials 26 (2005) 5991–5998.
[1180] M.C. Schneider, S. Chu, M.A. Randolph, S.J. Bryant, Acta Biomater. 93 (2019) 97–110.
[1181] N.S. Hwang, S. Varghese, P. Theprungsirikul, A. Canver, J. Elisseeff, Biomaterials 27 (2006) 6015–6023.
[1182] S.J. Bryant, K.S. Anseth, J. Biomed. Mater. Res. 59 (2002) 63–72.
[1183] B. Sharma, S. Fermanian, M. Gibson, S. Unterman, D.A. Herzka, B. Cascio, J. Coburn, A.Y. Hui, N. Marcus, G.E. Gold, J.H. Elisseeff, Sci. Transl. Med. 5 (2013) 167ra166-167ra166.
[1184] Y. Park, M.P. Lutolf, J.A. Hubbell, E.B. Hunziker, M. Wong, Tissue Eng. 10 (2004) 515–522.
[1185] M. Wathier, C.S. Johnson, T. Kim, M.W. Grinstaff, Bioconjug. Chem. 17 (2006) 873–876.
[1186] G.H. Underhill, A.A. Chen, D.R. Albrecht, S.N. Bhatia, Biomaterials 28 (2007) 256–270.
[1187] M.V. Tsurkan, P.V. Hauser, A. Zieris, R. Carvalhosa, B. Bussolati, U. Freudenberg, G. Camussi, C. Werner, J. Control. Release 167 (2013) 248–255.
[1188] P.N. Patel, A.S. Gobin, J.L. West, C.W. Patrick, Tissue Eng. 11 (2005) 1498–1505.
[1189] V.V. Rao, M.K. Vu, H. Ma, A.R. Killaars, K.S. Anseth, Bioeng. Transl. Med. 4 (2019) 51–60.
[1190] J.J. Moon, M.S. Hahn, I. Kim, B.A. Nsiah, J.L. West, Tissue Eng. Part A 15 (2008) 579–585.
[1191] W.-G. Koh, A. Revzin, M.V. Pishko, Langmuir 18 (2002) 2459–2462.
[1192] D.S.W. Benoit, M.P. Schwartz, A.R. Durney, K.S. Anseth, Nat. Mater. 7 (2008) 816–823.
[1193] K. Knop, R. Hoogenboom, D. Fischer, U.S. Schubert, Angew. Chemie Int. Ed. 49 (2010) 6288–6308.
[1194] N.A. Peppas, K.B. Keys, M. Torres-Lugo, A.M. Lowman, J. Control. Release 62 (1999) 81–87.
[1195] C.D. Pritchard, T.M. O'Shea, D.J. Siegwart, E. Calo, D.G. Anderson, F.M. Reynolds, J.A. Thomas, J.R. Slotkin, E.J. Woodard, R. Langer, Biomaterials 32 (2011) 587–597.
[1196] B.A. Badeau, M.P. Comerford, C.K. Arakawa, J.A. Shadish, C.A. DeForest, Nat. Chem. 10 (2018) 251–258.
[1197] N.B. Graham, J.M. Harris (Ed.), Poly(Ethylene Glycol) Chemistry: Biotechnical and Biomedical Applications, Springer US, Boston, MA, 1992, pp. 263–281.
[1198] L. Schirmer, P. Atallah, C. Werner, U. Freudenberg, Adv. Healthc. Mater. 5 (2016) 3157–3164.
[1199] A.A. Aimetti, A.J. Machen, K.S. Anseth, Biomaterials 30 (2009) 6048–6054.
[1200] M.B. Mellott, K. Searcy, M.V. Pishko, Biomaterials 22 (2001) 929–941.
[1201] B. Qiu, S. Stefanos, J. Ma, A. Lalloo, B.A. Perry, M.J. Leibowitz, P.J. Sinko, S. Stein, Biomaterials 24 (2003) 11–18.
[1202] P. van de Wetering, A.T. Metters, R.G. Schoenmakers, J.A. Hubbell, J. Control. Release 102 (2005) 619–627.
[1203] A. Zieris, S. Prokoph, K.R. Levental, P.B. Welzel, M. Grimmer, U. Freudenberg, C. Werner, Biomaterials 31 (2010) 7985–7994.
[1204] J.E. Leslie-Barbick, J.J. Moon, J.L. West, J. Biomater. Sci. Polym. Ed. 20 (2009) 1763–1779.
[1205] A. Song, A.A. Rane, K.L. Christman, Acta Biomater. 8 (2012) 41–50.
[1206] S. Halstenberg, A. Panitch, S. Rizzi, H. Hall, J.A. Hubbell, Biomacromolecules 3 (2002) 710–723.
[1207] R.J. Russell, M.V. Pishko, C.C. Gefrides, M.J. McShane, G.L. Coté, Anal. Chem. 71 (1999) 3126–3132.
[1208] R.J. Russell, M.V. Pishko, A.L. Simonian, J.R. Wild, Anal. Chem. 71 (1999) 4909–4912.
[1209] C.M. Hassan, N.A. Peppas, Structure and Applications of Poly(Vinyl Alcohol) Hydrogels Produced by Conventional Crosslinking or by Freezing/Thawing

Methods, Biopolymers PVA Hydrogels, Anionic Polymerisation Nanocomposites vol 153, Springer, Berlin Heidelberg, Berlin, Heidelberg, 2000, pp. 37–65.
[1210] L. Lum, J. Elisseeff, N. Ashammakhi, P. Ferretti (Eds.), Topics in Tissue Engineering, 2003, pp. 1–25.
[1211] J.K. Li, N. Wang, X.S. Wu, J. Control. Release 56 (1998) 117–126.
[1212] R.H. Schmedlen, K.S. Masters, J.L. West, Biomaterials 23 (2002) 4325–4332.
[1213] S.R. Stauffer, N.A. Peppast, Polymer 33 (1992) 3932–3936.
[1214] P.J. Willcox, D.W. Howie Jr., K. Schmidt-Rohr, D.A. Hoagland, S.P. Gido, S. Pudjijanto, L.W. Kleiner, S. Venkatraman, J. Polym. Sci. Part B: Polym. Phys. 37 (1999) 3438–3454.
[1215] R. Ricciardi, F. Auriemma, C. De Rosa, F. Lauprêtre, Macromolecules 37 (2004) 1921–1927.
[1216] C.M. Hassan, N.A. Peppas, Macromolecules 33 (2000) 2472–2479.
[1217] F. Yokoyama, I. Masada, K. Shimamura, T. Ikawa, K. Monobe, Colloid Polym. Sci. 264 (1986) 595–601.
[1218] M. Oka, T. Noguchi, P. Kumar, K. Ikeuchi, T. Yamamuro, S.H. Hyon, Y. Ikada, Clin. Mater. 6 (1990) 361–381.
[1219] S. Lin, X. Liu, J. Liu, H. Yuk, H.-C. Loh, G.A. Parada, C. Settens, J. Song, A. Masic, G.H. McKinley, X. Zhao, Sci. Adv. 5 (2019) eaau8528.
[1220] V.I. Lozinsky, Russ. Chem. Rev. 67 (1998) 573–586.
[1221] V.I. Lozinsky, F.M. Plieva, Enzyme Microb. Technol. 23 (1998) 227–242.
[1222] N.A. Peppas, S.R. Stauffer, J. Control. Release 16 (1991) 305–310.
[1223] K.R. Hixon, T. Lu, S.A. Sell, Acta Biomater. 62 (2017) 29–41.
[1224] S.-H. Hyon, W.-I. Cha, Y. Ikada, M. Kita, Y. Ogura, Y. Honda, J. Biomater. Sci. Polym. Ed. 5 (1994) 397–406.
[1225] Y. Hou, W. Xie, K. Achazi, J.L. Cuellar-Camacho, M.F. Melzig, W. Chen, R. Haag, Acta Biomater. 77 (2018) 28–37.
[1226] D. Mawad, L.A. Poole-Warren, P. Martens, L.H. Koole, T.L.B. Slots, C.S.Jv. Hooy-Corstjens, Biomacromolecules 9 (2008) 263–268.
[1227] P. Martens, T. Holland, K.S. Anseth, Polymer 43 (2002) 6093–6100.
[1228] E.H. Nafea, L.A. Poole-Warren, P.J. Martens, J. Biomater. Sci. Polym. Ed. 25 (2014) 1771–1790.
[1229] P. Martens, K.S. Anseth, Polymer 41 (2000) 7715–7722.
[1230] X.-H. Qin, X. Wang, M. Rottmar, B.J. Nelson, K. Maniura-Weber, Adv. Mater. 30 (2018) 1705564.
[1231] X.-H. Qin, K. Labuda, J. Chen, V. Hruschka, A. Khadem, R. Liska, H. Redl, P. Slezak, Adv. Funct. Mater. 25 (2015) 6606–6617.
[1232] K.S. Lim, M.H. Alves, L.A. Poole-Warren, P.J. Martens, Biomaterials 34 (2013) 7097–7105.
[1233] K.S. Lim, J.J. Roberts, M.H. Alves, L.A. Poole-Warren, P.J. Martens, J. Appl. Polym. Sci. 132 (2015).
[1234] K.S. Lim, Y. Ramaswamy, J.J. Roberts, M.-H. Alves, L.A. Poole-Warren, P.J. Martens, Macromol. Biosci. 15 (2015) 1423–1432.
[1235] Y. Zhou, S. Zhao, C. Zhang, K. Liang, J. Li, H. Yang, S. Gu, Z. Bai, D. Ye, W. Xu, Carbohydr. Polym. 184 (2018) 383–389.
[1236] P. Martens, J. Blundo, A. Nilasaroya, R.A. Odell, J. Cooper-White, L.A. Poole-Warren, Chem. Mater. 19 (2007) 2641–2648.
[1237] N.A. Peppas, E.W. Merrill, J. Biomed. Mater. Res. 11 (1977) 423–434.
[1238] Y.-N. Chen, L. Peng, T. Liu, Y. Wang, S. Shi, H. Wang, ACS Appl. Mater. Interfaces 8 (2016) 27199–27206.
[1239] B.E.B. Jensen, K. Edlund, A.N. Zelikin, Biomaterials 49 (2015) 113–124.
[1240] B.E.B. Jensen, I. Dávila, A.N. Zelikin, J. Phys. Chem. B 120 (2016) 5916–5926.
[1241] D. Mawad, P.J. Martens, R.A. Odell, L.A. Poole-Warren, Biomaterials 28 (2007) 947–955.
[1242] K. Burczak, T. Fujisato, M. Hatada, Y. Ikada, Biomaterials 15 (1994) 231–238.
[1243] M.H. Alves, C.J. Young, K. Bozzetto, L.A. Poole-Warren, P.J. Martens, Biomed. Mater. 7 (2012) 024106.
[1244] D.A. Ossipov, J. Hilborn, Macromolecules 39 (2006) 1709–1718.
[1245] J. Han, T. Lei, Q. Wu, Cellulose 20 (2013) 2947–2958.
[1246] G. Huang, H. Zhang, Y. Liu, H. Chang, H. Zhang, H. Song, D. Xu, T. Shi, Macromolecules 50 (2017) 2124–2135.
[1247] N.A. Peppas, E.W. Merrill, J. Appl. Polym. Sci. 21 (1977) 1763–1770.
[1248] N.A. Peppas, E.W. Merrill, J. Polym. Sci.: Polym. Chem. Ed. 14 (1976) 441–457.
[1249] A. Danno, J. Phys. Soc. Jpn. 13 (1958) 722–727.
[1250] N.A. Peppas, E.W. Merrill, J. Appl. Polym. Sci. 20 (1976) 1457–1465.
[1251] Y. Zhou, C. Wan, Y. Yang, H. Yang, S. Wang, Z. Dai, K. Ji, H. Jiang, X. Chen, Y. Long, Adv. Funct. Mater. 29 (2019) 1806220.
[1252] J.A. Stammen, S. Williams, D.N. Ku, R.E. Guldberg, Biomaterials 22 (2001) 799–806.
[1253] H. Zhang, H. Xia, Y. Zhao, ACS Macro Lett. 1 (2012) 1233–1236.
[1254] S.H. Hyon, W.I. Cha, Y. Ikada, Polym. Bull. 22 (1989) 119–122.
[1255] T. Noguchi, T. Yamamuro, M. Oka, P. Kumar, Y. Kotoura, S.-H. Hyonyt, Y. Ikadat, J. Appl. Biomater. 2 (1991) 101–107.
[1256] M.I. Baker, S.P. Walsh, Z. Schwartz, B.D. Boyan, J. Biomed. Mater. Res. Part B Appl. Biomater. 100B (2012) 1451–1457.
[1257] Y. Cao, D. Xiong, K. Wang, Y. Niu, J. Mech. Behav. Biomed. Mater. 68 (2017) 163–172.
[1258] L. Coluccino, R. Gottardi, F. Ayadi, A. Athanassiou, R.S. Tuan, L. Ceseracciu, ACS Biomater. Sci. Eng. 4 (2018) 1518–1527.
[1259] U.A. Aregueta-Robles, P.J. Martens, L.A. Poole-Warren, R.A. Green, Acta Biomater. 95 (2019) 269–284.
[1260] M.F.A. Cutiongco, S.H. Goh, R. Aid-Launais, C. Le Visage, H.Y. Low, E.K.F. Yim, Biomaterials 84 (2016) 184–195.
[1261] J.D. Weaver, D.N. Ku, J. Biomed. Mater. Res. Part B Appl. Biomater. 100B (2012) 103–110.
[1262] N.A. Peppas, R.E. Berner, Biomaterials 1 (1980) 158–162.
[1263] N.A. Peppas, N.K. Mongia, Eur. J. Pharm. Biopharm. 43 (1997) 51–58.
[1264] T. Li, Y. Lei, M. Guo, H. Yan, Eur. Polym. J. 101 (2018) 77–82.
[1265] J. Schulze, S. Hendrikx, M. Schulz-Siegmund, A. Aigner, Acta Biomater. 45 (2016) 210–222.
[1266] K.S.B. Masters, S.J. Leibovich, P. Belem, J.L. West, L.A. Poole-Warren, Wound Repair Regen. 10 (2002) 286–294.
[1267] S. Ramakrishna, J. Mayer, E. Wintermantel, K.W. Leong, Compos. Sci. Technol. 61 (2001) 1189–1224.
[1268] A. Sionkowska, Prog. Polym. Sci. 36 (2011) 1254–1276.
[1269] R.A. Pérez, J.-E. Won, J.C. Knowles, H.-W. Kim, Adv. Drug Deliv. Rev. 65 (2013) 471–496.
[1270] L.M. Mathieu, T.L. Mueller, P.-E. Bourban, D.P. Pioletti, R. Müller, J.-A.E. Månson, Biomaterials 27 (2006) 905–916.
[1271] S. Afewerki, A. Sheikhi, S. Kannan, S. Ahadian, A. Khademhosseini, Bioeng. Transl. Med. 4 (2019) 96–115.
[1272] A. Fernández-Ferreiro, M. González Barcia, M. Gil-Martínez, A. Vieites-Prado, I. Lema, B. Argibay, J. Blanco Méndez, M.J. Lamas, F.J. Otero-Espinar, Eur. J. Pharm. Biopharm. 94 (2015) 342–351.
[1273] Y. Luo, J.B. Kobler, J.T. Heaton, X. Jia, S.M. Zeitels, R. Langer, J. Biomed. Mater. Res. Part B Appl. Biomater. 93B (2010) 386–393.
[1274] R. Jin, L.S. Moreira Teixeira, P.J. Dijkstra, C.A. van Blitterswijk, M. Karperien, J. Feijen, J. Control. Release 152 (2011) 186–195.
[1275] M.T. Ho, C.J. Teal, M.S. Shoichet, Brain Res. Bull. 148 (2019) 46–54.
[1276] S.L. Payne, A. Tuladhar, J.M. Obermeyer, B.V. Varga, C.J. Teal, C.M. Morshead, A. Nagy, M.S. Shoichet, Biomaterials 192 (2019) 309–322.
[1277] T. Ito, Y. Yeo, C.B. Highley, E. Bellas, C.A. Benitez, D.S. Kohane, Biomaterials 28 (2007) 975–983.
[1278] B.J. Kong, A. Kim, S.N. Park, Carbohydr. Polym. 147 (2016) 473–481.
[1279] J.F. Piai, A.F. Rubira, E.C. Muniz, Acta Biomater. 5 (2009) 2601–2609.
[1280] A.R. Fajardo, M.B. Silva, L.C. Lopes, J.F. Piai, A.F. Rubira, E.C. Muniz, RSC Adv. 2 (2012) 11095–11103.
[1281] M. Tanihara, Y. Suzuki, E. Yamamoto, A. Noguchi, Y. Mizushima, J. Biomed. Mater. Res. 56 (2001) 216–221.
[1282] O. Jeon, C. Powell, L.D. Solorio, M.D. Krebs, E. Alsberg, J. Control. Release 154 (2011) 258–266.
[1283] Y. Zhu, J. Tan, H. Zhu, G. Lin, F. Yin, L. Wang, K. Song, Y. Wang, G. Zhou, W. Yi, Biomater. Sci. 5 (2017) 784–791.
[1284] H. Tan, C.R. Chu, K.A. Payne, K.G. Marra, Biomaterials 30 (2009) 2499–2506.
[1285] N. Mohan, P.V. Mohanan, A. Sabareeswaran, P. Nair, Int. J. Biol. Macromol. 104 (2017) 1936–1945.
[1286] D.G. Miranda, S.M. Malmonge, D.M. Campos, N.G. Attik, B. Grosgogeat, K. Gritsch, J. Biomed. Mater. Res. Part B Appl. Biomater. 104 (2016) 1691–1702.
[1287] L. Li, N. Wang, X. Jin, R. Deng, S. Nie, L. Sun, Q. Wu, Y. Wei, C. Gong, Biomaterials 35 (2014) 3903–3917.
[1288] Y. Deng, J. Ren, G. Chen, G. Li, X. Wu, G. Wang, G. Gu, J. Li, Sci. Rep. 7 (2017) 2699.
[1289] F. Chen, Y. Ni, B. Liu, T. Zhou, C. Yu, Y. Su, X. Zhu, X. Yu, Y. Zhou, Carbohydr. Polym. 166 (2017) 31–44.
[1290] S. Ansari, I.M. Diniz, C. Chen, T. Aghaloo, B.M. Wu, S. Shi, A. Moshaverinia, J. Mater. Sci. Mater. Med. 28 (2017) 162.
[1291] K. Ziv, H. Nuhn, Y. Ben-Haim, L.S. Sasportas, P.J. Kempen, T.P. Niedringhaus, M. Hrynyk, R. Sinclair, A.E. Barron, S.S. Gambhir, Biomaterials 35 (2014) 3736–3743.
[1292] B. Kundu, A.R. Bastos, V. Brancato, M.T. Cerqueira, J.M. Oliveira, V.M. Correlo, R.L. Reis, S.C. Kundu, ACS Appl. Mater. Interfaces 11 (2019) 14548–14559.
[1293] L.A. Reis, L.L.Y. Chiu, Y. Liang, K. Hyunh, A. Momen, M. Radisic, Acta Biomater. 8 (2012) 1022–1036.
[1294] F. Cui, G. Li, J. Huang, J. Zhang, M. Lu, W. Lu, J. Huan, Q. Huang, Drug Deliv. 18 (2011) 173–180.
[1295] B.M. Gillette, J.A. Jensen, M. Wang, J. Tchao, S.K. Sia, Adv. Mater. 22 (2010) 686–691.
[1296] B. Sarker, T. Zehnder, S.N. Rath, R.E. Horch, U. Kneser, R. Detsch, A.R. Boccaccini, ACS Biomater. Sci. Eng. 3 (2017) 1730–1737.
[1297] B. Sarker, R. Singh, T. Zehnder, T. Forgber, C. Alexiou, I. Cicha, R. Detsch, A.R. Boccaccini, J. Bioact. Compat. Polym. 32 (2016) 309–324.
[1298] B. Balakrishnan, M. Mohanty, P.R. Umashankar, A. Jayakrishnan, Biomaterials 26 (2005) 6335–6342.
[1299] M. Khamrai, S.L. Banerjee, S. Paul, S. Samanta, P.P. Kundu, Int. J. Biol. Macromol. 122 (2019) 940–953.
[1300] K. Numata, S. Yamazaki, T. Katashima, J.-A. Chuah, N. Naga, T. Sakai, Macromol. Biosci. 14 (2014) 799–806.
[1301] K. Gwon, E. Kim, G. Tae, Acta Biomater. 49 (2017) 284–295.
[1302] M.S. Bae, N.R. Ko, S.J. Lee, J.B. Lee, D.N. Heo, W. Byun, B.-J. Choi, H.B. Jeon, H.-J. Jang, J.-Y. Ahn, D.-S. Hwang, B.-Y. Jung, I.K. Kwon, Macromol. Res. 24 (2016) 829–837.
[1303] X. Zhou, J. Wang, W. Fang, Y. Tao, T. Zhao, K. Xia, C. Liang, J. Hua, F. Li, Q. Chen, Acta Biomater. 71 (2018) 496–509.
[1304] Y. Gao, B. Li, W. Kong, L. Yuan, L. Guo, C. Li, H. Fan, Y. Fan, X. Zhang, Int. J. Biol. Macromol. 118 (2018) 2014–2020.
[1305] S. Thönes, S. Rother, T. Wippold, J. Blaszkiewicz, K. Balamurugan, S. Moeller, G. Ruiz-Gómez, M. Schnabelrauch, D. Scharnweber, A. Saalbach, J. Rademann, M.T. Pisabarro, V. Hintze, U. Anderegg, Acta Biomater. 86 (2019) 135–147.
[1306] S. Kupper, I. Kłosowska-Chomiczewska, P. Szumała, Carbohydr. Polym. 175 (2017) 347–354.
[1307] R.H. Koh, Y. Jin, B.-J. Kang, N.S. Hwang, Acta Biomater. 53 (2017) 318–328.
[1308] H. Ying, J. Zhou, M. Wang, D. Su, Q. Ma, G. Lv, J. Chen, Mater. Sci. Eng. C 101

(2019) 487–498.
[1309] J. Yang, Y. Liu, L. He, Q. Wang, L. Wang, T. Yuan, Y. Xiao, Y. Fan, X. Zhang, Acta Biomater. 74 (2018) 156–167.
[1310] G.W. Omlor, A.G. Nerlich, H. Lorenz, T. Bruckner, W. Richter, M. Pfeiffer, T. Gühring, Eur. Spine J. 21 (2012) 1700–1708.
[1311] A. Matsiko, T.J. Levingstone, J.P. Gleeson, F.J. O'Brien, Adv. Healthc. Mater. 4 (2015) 1175–1179.
[1312] Y. Liu, J. Yang, Z. Luo, D. Li, J. Lu, Q. Wang, Y. Xiao, X. Zhang, J. Mater. Chem. B 7 (2019) 2845–2854.
[1313] H.C. Bowers, M.L. Fiori, J.B. Khadela, P.A. Janmey, P.A. Galie, Exp. Cell Res. 376 (2019) 49–57.
[1314] D. Zhu, H. Wang, P. Trinh, S.C. Heilshorn, F. Yang, Biomaterials 127 (2017) 132–140.
[1315] H. Wang, D. Zhu, A. Paul, L. Cai, A. Enejder, F. Yang, S.C. Heilshorn, Adv. Funct. Mater. 27 (2017) 1605609.
[1316] Y. Yu, M.J. Brouillette, D. Seol, H. Zheng, J.A. Buckwalter, J.A. Martin, Arthritis Rheumatol. 67 (2015) 1274–1285.
[1317] P.E. Mikael, H.S. Kim, S.P. Nukavarapu, J. Biomed. Mater. Res. Part B Appl. Biomater. 106 (2018) 300–309.
[1318] N.R. Raia, B.P. Partlow, M. McGill, E.P. Kimmerling, C.E. Ghezzi, D.L. Kaplan, Biomaterials 131 (2017) 58–67.
[1319] B. Tavsanli, O. Okay, Carbohydr. Polym. 208 (2019) 413–420.
[1320] J.-W. Luo, C. Liu, J.-H. Wu, L.-X. Lin, H.-M. Fan, D.-H. Zhao, Y.-Q. Zhuang, Y.-L. Sun, Mater. Sci. Eng. C 98 (2019) 628–634.
[1321] S.E.L. Vidal, K.A. Tamamoto, H. Nguyen, R.D. Abbott, D.M. Cairns, D.L. Kaplan, Biomaterials 198 (2019) 194–203.
[1322] A. Sundarakrishnan, H. Zukas, J.M. Coburn, B. Bertini, Z. Liu, I. Georgakoudi, L. Baugh, Q. Dasgupta, L.D. Black, D.L. Kaplan, ACS Biomater. Sci. Eng. 5 (2019) 2417–2429.
[1323] J.O. Buitrago, K.D. Patel, A. El-Fiqi, J.-H. Lee, B. Kundu, H.-H. Lee, H.-W. Kim, Acta Biomater. 69 (2018) 218–233.
[1324] H. Goodarzi, K. Jadidi, S. Pourmotabed, E. Sharifi, H. Aghamollaei, Int. J. Biol. Macromol. 126 (2019) 620–632.
[1325] J.R. Soucy, E. Shirzaei Sani, R. Portillo Lara, D. Diaz, F. Dias, A.S. Weiss, A.N. Koppes, R. Koppes, N. Annabi, Tissue Eng. Part A 24 (2018) 1393–1405.
[1326] W. Xiao, J. Li, X. Qu, L. Wang, Y. Tan, K. Li, H. Li, X. Yue, B. Li, X. Liao, Mater. Sci. Eng. C 99 (2019) 57–67.
[1327] X. Wenqian, L. Wenlong, S. Jing, D. Xiuli, W. Dan, F. Hongsong, J. Bioact. Compat. Polym. 27 (2012) 327–341.
[1328] W. Huang, A. Tarakanova, N. Dinjaski, Q. Wang, X. Xia, Y. Chen, J.Y. Wong, M.J. Buehler, D.L. Kaplan, Adv. Funct. Mater. 26 (2016) 4113–4123.
[1329] E.G. Roberts, N.-G. Rim, W. Huang, A. Tarakanova, J. Yeo, M.J. Buehler, D.L. Kaplan, J.Y. Wong, Macromol. Biosci. 18 (2018) 1800265.
[1330] Z. Megeed, J. Cappello, H. Ghandehari, Pharm. Res. 19 (2002) 954–959.
[1331] J.A. Gustafson, R.A. Price, K. Greish, J. Cappello, H. Ghandehari, Mol. Pharm. 7 (2010) 1050–1056.
[1332] A.A. Dinerman, J. Cappello, H. Ghandehari, S.W. Hoag, Biomaterials 23 (2002) 4203–4210.
[1333] T. Li, J. Wang, L. Zhang, J. Yang, M. Yang, D. Zhu, X. Zhou, S. Handschuh-Wang, Y. Liu, X. Zhou, J. Mater. Chem. B 5 (2017) 5726–5732.
[1334] K. Zhao, T. Chen, B. Lin, W. Cui, B. Kan, N. Yang, X. Zhou, X. Zhang, J. Wei, React. Funct. Polym. 87 (2015) 7–14.
[1335] X. Zhang, B. Lin, K. Zhao, J. Wei, J. Guo, W. Cui, S. Jiang, D. Liu, J. Li, Desalination 365 (2015) 234–241.
[1336] G. Li, G. Zhang, R. Sun, C.-P. Wong, J. Mater. Sci. 52 (2017) 8538–8545.
[1337] M.M. Fitzgerald, K. Bootsma, J.A. Berberich, J.L. Sparks, Biomacromolecules 16 (2015) 1497–1505.
[1338] F. Wang, Y. Wen, T. Bai, Mater. Sci. Eng. C 69 (2016) 268–275.
[1339] H.S. Kim, D. Kim, Y.W. Jeong, M.J. Choi, G.W. Lee, M. Thangavelu, J.E. Song, G. Khang, Int. J. Biol. Macromol. 130 (2019) 220–228.
[1340] R. Wieduwild, S. Krishnan, K. Chwalek, A. Boden, M. Nowak, D. Drechsel, C. Werner, Y. Zhang, Angew. Chemie Int. Ed. 54 (2015) 3962–3966.
[1341] S. Jung, J.H. Abel, J.L. Starger, H. Yi, Biomacromolecules 17 (2016) 2427–2436.
[1342] L.-Y. Wang, M.-J. Wang, ACS Sustain. Chem. Eng. 4 (2016) 2830–2837.
[1343] D. Zhang, W. Zhou, B. Wei, X. Wang, R. Tang, J. Nie, J. Wang, Carbohydr. Polym. 125 (2015) 189–199.
[1344] Y.-F. Tang, Y.-M. Du, X.-W. Hu, X.-W. Shi, J.F. Kennedy, Carbohydr. Polym. 67 (2007) 491–499.
[1345] M.A. Abureesh, A.A. Oladipo, M. Gazi, Int. J. Biol. Macromol. 90 (2016) 75–80.
[1346] E.A. Kamoun, E.-R.S. Kenawy, T.M. Tamer, M.A. El-Meligy, M.S. Mohy Eldin, Arab. J. Chem. 8 (2015) 38–47.
[1347] X. Jiang, N. Xiang, H. Zhang, Y. Sun, Z. Lin, L. Hou, Carbohydr. Polym. 186 (2018) 377–383.
[1348] S. Hua, H. Ma, X. Li, H. Yang, A. Wang, Int. J. Biol. Macromol. 46 (2010) 517–523.
[1349] I. Strehin, Z. Nahas, K. Arora, T. Nguyen, J. Elisseeff, Biomaterials 31 (2010) 2788–2797.
[1350] A. Zieris, K. Chwalek, S. Prokoph, K.R. Levental, P.B. Welzel, U. Freudenberg, C. Werner, J. Control. Release 156 (2011) 28–36.
[1351] P.B. Welzel, S. Prokoph, A. Zieris, M. Grimmer, S. Zschoche, U. Freudenberg, C. Werner, Polymers 3 (2011) 602–620.
[1352] A. Watarai, L. Schirmer, S. Thönes, U. Freudenberg, C. Werner, J.C. Simon, U. Anderegg, Acta Biomater. 25 (2015) 65–75.
[1353] M.V. Tsurkan, K. Chwalek, S. Prokoph, A. Zieris, K.R. Levental, U. Freudenberg, C. Werner, Adv. Mater. 25 (2013) 2606–2610.
[1354] K.M. Schultz, A.D. Baldwin, K.L. Kiick, E.M. Furst, Macromolecules 42 (2009) 5310–5316.
[1355] S. Prokoph, E. Chavakis, K.R. Levental, A. Zieris, U. Freudenberg, S. Dimmeler, C. Werner, Biomaterials 33 (2012) 4792–4800.
[1356] M. Goh, Y. Kim, K. Gwon, K. Min, Y. Hwang, G. Tae, Carbohydr. Polym. 174 (2017) 990–998.
[1357] A. Fu, K. Gwon, M. Kim, G. Tae, J.A. Kornfield, Biomacromolecules 16 (2015) 497–506.
[1358] L. Baumann, S. Prokoph, C. Gabriel, U. Freudenberg, C. Werner, A.G. Beck-Sickinger, J. Control. Release 162 (2012) 68–75.
[1359] A.D. Baldwin, K.L. Kiick, Polym. Chem. 4 (2013) 133–143.
[1360] J.J. Roberts, B.L. Farrugia, R.A. Green, J. Rnjak-Kovacina, P.J. Martens, J. Tissue Eng. 7 (2016) 2041731416677132.
[1361] E.W. Merrill, E.W. Salzman, P.S. Wong, T.P. Ashford, A.H. Brown, W.G. Austen, J. Appl. Physiol. 29 (1970) 723–730.
[1362] M.F.A. Goosen, M.V. Sefton, J. Biomed. Mater. Res. 17 (1983) 359–373.
[1363] C.H. Cholakis, W. Zingg, M.V. Sefton, J. Biomed. Mater. Res. 23 (1989) 417–441.
[1364] J.J. Roberts, P. Naudiyal, L. Jugé, L.E. Bilston, A.M. Granville, P.J. Martens, ACS Biomater. Sci. Eng. 1 (2015) 1267–1277.
[1365] X. Zheng Shu, Y. Liu, F.S. Palumbo, Y. Luo, G.D. Prestwich, Biomaterials 25 (2004) 1339–1348.
[1366] A. Ranga, M.P. Lutolf, J. Hilborn, D.A. Ossipov, Biomacromolecules 17 (2016) 1553–1560.
[1367] B.L. Ekerdt, C.M. Fuentes, Y. Lei, M.M. Adil, A. Ramasubramanian, R.A. Segalman, D.V. Schaffer, Adv. Healthc. Mater. 7 (2018) 1800225.
[1368] M. Patenaude, T. Hoare, Biomacromolecules 13 (2012) 369–378.
[1369] D. Yu, C. Sun, Z. Zheng, X. Wang, D. Chen, H. Wu, X. Wang, F. Shi, Int. J. Pharm. 503 (2016) 229–237.
[1370] X. Wang, B. Partlow, J. Liu, Z. Zheng, B. Su, Y. Wang, D.L. Kaplan, Acta Biomater. 12 (2015) 51–61.
[1371] D. Ma, G. An, M. Liang, Y. Liu, B. Zhang, Y. Wang, Mater. Sci. Eng. C 65 (2016) 221–231.
[1372] B.B. Mandal, B. Ghosh, S.C. Kundu, Int. J. Biol. Macromol. 49 (2011) 125–133.
[1373] K.S. Lim, J. Kundu, A. Reeves, L.A. Poole-Warren, S.C. Kundu, P.J. Martens, Macromol. Biosci. 12 (2012) 322–332.
[1374] J. Kundu, L.A. Poole-Warren, P. Martens, S.C. Kundu, Acta Biomater. 8 (2012) 1720–1729.
[1375] R. Yamdej, K. Pangza, T. Srichana, P. Aramwit, J. Bioact. Compat. Polym. 32 (2016) 32–44.
[1376] L. Han, J. Xu, X. Lu, D. Gan, Z. Wang, K. Wang, H. Zhang, H. Yuan, J. Weng, J. Mater. Chem. B 5 (2017) 731–741.
[1377] T. Liu, C. Jiao, X. Peng, Y.-N. Chen, Y. Chen, C. He, R. Liu, H. Wang, J. Mater. Chem. B 6 (2018) 8105–8114.
[1378] T. Jayaramudu, Y. Li, H.-U. Ko, I.R. Shishir, J. Kim, Int. J. Precis. Eng. Manuf. Technol. 3 (2016) 375–379.
[1379] Z. Yu-chong, L. Xiao-xia, L. Wei, J. Yu-kun, Z. Jia-wei, C. Tao, Macromol. Mater. Eng. 303 (2018) 1800144.
[1380] B. Suthar, H.X. Xiao, D. Klempner, K.C. Frisch, Polym. Adv. Technol. 7 (1996) 221–233.
[1381] L.H. Sperling, Polym. Eng. Sci. 25 (1985) 517–520.
[1382] H.L. Frisch, D. Klempner, K.C. Frisch, J. Polym. Sci. B 7 (1969) 775–779.
[1383] L.H. Sperling, Interpenetrating Polymer Networks: An Overview, Interpenetrating Polymer Networks vol 239, American Chemical Society, 1994, pp. 3–38.
[1384] L.H. Sperling, Interpenetrating Polymer Networks and Related Materials, Springer Science & Business Media, Boston, MA, 1981.
[1385] D. Myung, D. Waters, M. Wiseman, P.-E. Duhamel, J. Noolandi, C.N. Ta, C.W. Frank, Polym. Adv. Technol. 19 (2008) 647–657.
[1386] E.S. Dragan, Chem. Eng. J. 243 (2014) 572–590.
[1387] N. Zoratto, P. Matricardi, Semi-IPN- and IPN-Based Hydrogels, in: J.M. Oliveira, S. Pina, R.L. Reis, J. San Roman (Eds.), Osteochondral Tissue Engineering: Challenges, Current Strategies, and Technological Advances, vol 1059, Springer International Publishing, Cham, 2018, pp. 155–188.
[1388] J.P. Gong, Y. Katsuyama, T. Kurokawa, Y. Osada, Adv. Mater. 15 (2003) 1155–1158.
[1389] T. Nonoyama, S. Wada, R. Kiyama, N. Kitamura, M.T.I. Mredha, X. Zhang, T. Kurokawa, T. Nakajima, Y. Takagi, K. Yasuda, J.P. Gong, Adv. Mater. 28 (2016) 6740–6745.
[1390] C.B. Rodell, N.N. Dusaj, C.B. Highley, J.A. Burdick, Adv. Mater. 28 (2016) 8419–8424.
[1391] W. Yang, H. Furukawa, J.P. Gong, Adv. Mater. 20 (2008) 4499–4503.
[1392] Y. Zhao, T. Nakajima, J.J. Yang, T. Kurokawa, J. Liu, J. Lu, S. Mizumoto, K. Sugahara, N. Kitamura, K. Yasuda, A.U.D. Daniels, J.P. Gong, Adv. Mater. 26 (2014) 436–442.
[1393] M.C. Darnell, J.-Y. Sun, M. Mehta, C. Johnson, P.R. Arany, Z. Suo, D.J. Mooney, Biomaterials 34 (2013) 8042–8048.
[1394] J.P. Gong, Soft Matter 6 (2010) 2583–2590.
[1395] T. Matsuda, R. Kawakami, R. Namba, T. Nakajima, J.P. Gong, Science 363 (2019) 504–508.
[1396] A. Nakayama, A. Kakugo, J.P. Gong, Y. Osada, M. Takai, T. Erata, S. Kawano, Adv. Funct. Mater. 14 (2004) 1124–1128.
[1397] Q. Chen, L. Zhu, C. Zhao, Q. Wang, J. Zheng, Adv. Mater. 25 (2013) 4171–4176.
[1398] H.J. Zhang, T.L. Sun, A.K. Zhang, Y. Ikura, T. Nakajima, T. Nonoyama, T. Kurokawa, O. Ito, H. Ishitobi, J.P. Gong, Adv. Mater. 28 (2016) 4884–4890.
[1399] Y. Yang, X. Wang, F. Yang, H. Shen, D. Wu, Adv. Mater. 28 (2016) 7178–7184.
[1400] P. Fratzl, R. Weinkamer, Prog. Mater. Sci. 52 (2007) 1263–1334.
[1401] P. Fratzl, H.S. Gupta, E.P. Paschalis, P. Roschger, J. Mater. Chem. 14 (2004)

2115–2123.
[1402] H. Gao, B. Ji, I.L. Jäger, E. Arzt, P. Fratzl, Proc. Natl. Acad. Sci. 100 (2003) 5597–5600.
[1403] A.K. Gaharwar, N.A. Peppas, A. Khademhosseini, Biotechnol. Bioeng. 111 (2014) 441–453.
[1404] N.S. Satarkar, D. Biswal, J.Z. Hilt, Soft Matter 6 (2010) 2364–2371.
[1405] A. Motealleh, N.S. Kehr, Adv. Healthc. Mater. 6 (2017) 1600938.
[1406] F. Song, X. Li, Q. Wang, L. Liao, C. Zhang, J. Biomed. Nanotechnol. 11 (2015) 40–52.
[1407] N.S. Kehr, S. Atay, B. Ergün, Macromol. Biosci. 15 (2015) 445–463.
[1408] P. Schexnailder, G. Schmidt, Colloid Polym. Sci. 287 (2009) 1–11.
[1409] K. Haraguchi, Curr. Opin. Solid State Mater. Sci. 11 (2007) 47–54.
[1410] S. Merino, C. Martín, K. Kostarelos, M. Prato, E. Vázquez, ACS Nano 9 (2015) 4686–4697.
[1411] R. Messing, A.M. Schmidt, Polym. Chem. 2 (2011) 18–32.
[1412] K. Haraguchi, T. Takehisa, Adv. Mater. 14 (2002) 1120–1124.
[1413] K. Haraguchi, K. Uyama, H. Tanimoto, Macromol. Rapid Commun. 32 (2011) 1253–1258.
[1414] D.L. Taylor, M. in het Panhuis, Adv. Mater. 28 (2016) 9060–9093.
[1415] R.R. Arvizo, S. Bhattacharyya, R.A. Kudgus, K. Giri, R. Bhattacharya, P. Mukherjee, Chem. Soc. Rev. 41 (2012) 2943–2970.
[1416] E.C. Dreaden, A.M. Alkilany, X. Huang, C.J. Murphy, M.A. El-Sayed, Chem. Soc. Rev. 41 (2012) 2740–2779.
[1417] P. Thoniyot, M.J. Tan, A.A. Karim, D.J. Young, X.J. Loh, Adv. Sci. 2 (2015) 1400010.
[1418] A.M. Salgueiro, A.L. Daniel-da-Silva, S. Fateixa, T. Trindade, Carbohydr. Polym. 91 (2013) 100–109.
[1419] E. Faoucher, P. Nativo, K. Black, J.B. Claridge, M. Gass, S. Romani, A.L. Bleloch, M. Brust, Chem. Commun. (2009) 6661–6663.
[1420] A. Skardal, J. Zhang, L. McCoard, S. Oottamasathien, G.D. Prestwich, Adv. Mater. 22 (2010) 4736–4740.
[1421] D.N. Heo, W.-K. Ko, M.S. Bae, J.B. Lee, D.-W. Lee, W. Byun, C.H. Lee, E.-C. Kim, B.-Y. Jung, I.K. Kwon, J. Mater. Chem. B 2 (2014) 1584–1593.
[1422] R. Xing, K. Liu, T. Jiao, N. Zhang, K. Ma, R. Zhang, Q. Zou, G. Ma, X. Yan, Adv. Mater. 28 (2016) 3669–3676.
[1423] N. Kojic, E.M. Pritchard, H. Tao, M.A. Brenckle, J.P. Mondia, B. Panilaitis, F. Omenetto, D.L. Kaplan, Adv. Funct. Mater. 22 (2012) 3793–3798.
[1424] M.-j. Yin, Y. Zhang, Z. Yin, Q. Zheng, A.P. Zhang, Adv. Mater. Technol. 3 (2018) 1800051.
[1425] H. Qin, T. Zhang, H.-N. Li, H.-P. Cong, M. Antonietti, S.-H. Yu, Chem 3 (2017) 691–705.
[1426] C. Wang, N.T. Flynn, R. Langer, Adv. Mater. 16 (2004) 1074–1079.
[1427] Y. Lu, P. Spyra, Y. Mei, M. Ballauff, A. Pich, Macromol. Chem. Phys. 208 (2007) 254–261.
[1428] S.O. Blacklow, J. Li, B.R. Freedman, M. Zeidi, C. Chen, D.J. Mooney, Sci. Adv. 5 (2019) eaaw3963.
[1429] K.-H. Liu, T.-Y. Liu, S.-Y. Chen, D.-M. Liu, Acta Biomater. 4 (2008) 1038–1045.
[1430] M. Hasany, A. Thakur, N. Taebnia, F.B. Kadumudi, M.-A. Shahbazi, M.K. Pierchala, S. Mohanty, G. Orive, T.L. Andresen, C.B. Foldager, S. Yaghmaei, A. Arpanaei, A.K. Gaharwar, M. Mehrali, A. Dolatshahi-Pirouz, ACS Appl. Mater. Interfaces 10 (2018) 34924–34941.
[1431] Z.-K. Cui, S. Kim, J.J. Baljon, B.M. Wu, T. Aghaloo, M. Lee, Nat. Commun. 10 (2019) 3523.
[1432] A.K. Gaharwar, L.M. Cross, C.W. Peak, K. Gold, J.K. Carrow, A. Brokesh, K.A. Singh, Adv. Mater. 31 (2019) 1900332.
[1433] J.R. Xavier, T. Thakur, P. Desai, M.K. Jaiswal, N. Sears, E. Cosgriff-Hernandez, R. Kaunas, A.K. Gaharwar, ACS Nano 9 (2015) 3109–3118.
[1434] C. Yao, Z. Liu, C. Yang, W. Wang, X.-J. Ju, R. Xie, L.-Y. Chu, Adv. Funct. Mater. 25 (2015) 2980–2991.
[1435] J. Wang, L. Lin, Q. Cheng, L. Jiang, Angew. Chemie Int. Ed. 51 (2012) 4676–4680.
[1436] K. Haraguchi, H.-J. Li, Angew. Chemie Int. Ed. 44 (2005) 6500–6504.
[1437] Z. Hu, G. Chen, Adv. Mater. 26 (2014) 5950–5956.
[1438] Y.A. Shchipunov, Ty.Y. Karpenko, Langmuir 20 (2004) 3882–3887.
[1439] N. Seda Kehr, K. Riehemann, Adv. Healthc. Mater. 5 (2016) 193–197.
[1440] N.S. Kehr, E.A. Prasetyanto, K. Benson, B. Ergün, A. Galstyan, H.-J. Galla, Angew. Chemie Int. Ed. 52 (2013) 1156–1160.
[1441] R.A. Frimpong, S. Fraser, J. Zach Hilt, J. Biomed. Mater. Res. A. 80A (2007) 1–6.
[1442] N.S. Satarkar, J. Zach Hilt, Acta Biomater. 4 (2008) 11–16.
[1443] M.K. Jaiswal, J.R. Xavier, J.K. Carrow, P. Desai, D. Alge, A.K. Gaharwar, ACS Nano 10 (2016) 246–256.
[1444] C. Martín, S. Merino, J.M. González-Domínguez, R. Rauti, L. Ballerini, M. Prato, E. Vázquez, Sci. Rep. 7 (2017) 10942.
[1445] L. Wang, D. Song, X. Zhang, Z. Ding, X. Kong, Q. Lu, D.L. Kaplan, ACS Biomater. Sci. Eng. 5 (2019) 613–622.
[1446] E. Wang, M.S. Desai, S.-W. Lee, Nano Lett. 13 (2013) 2826–2830.
[1447] C.-H. Zhu, Y. Lu, J. Peng, J.-F. Chen, S.-H. Yu, Adv. Funct. Mater. 22 (2012) 4017–4022.
[1448] R. Liu, S. Liang, X.-Z. Tang, D. Yan, X. Li, Z.-Z. Yu, J. Mater. Chem. 22 (2012) 14160–14167.
[1449] S.R. Shin, B. Aghaei-Ghareh-Bolagh, T.T. Dang, S.N. Topkaya, X. Gao, S.Y. Yang, S.M. Jung, J.H. Oh, M.R. Dokmeci, X. Tang, A. Khademhosseini, Adv. Mater. 25 (2013) 6385–6391.
[1450] R.M.A. Domingues, M. Silva, P. Gershovich, S. Betta, P. Babo, S.G. Caridade, J.F. Mano, A. Motta, R.L. Reis, M.E. Gomes, Bioconjug. Chem. 26 (2015) 1571–1581.
[1451] G.K. Tummala, R. Rojas, A. Mihranyan, J. Phys. Chem. B 120 (2016) 13094–13101.
[1452] G.K. Tummala, T. Joffre, R. Rojas, C. Persson, A. Mihranyan, Soft Matter 13 (2017) 3936–3945.
[1453] A. Träger, A. Carlmark, L. Wågberg, Macromol. Mater. Eng. 303 (2018) 1700594.
[1454] D.R. Pereira, J. Silva-Correia, J.M. Oliveira, R.L. Reis, A. Pandit, M.J. Biggs, Nanomed. Nanotechnol. Biol. Med. 14 (2018) 897–908.
[1455] J. Hur, K. Im, S.W. Kim, J. Kim, D.-Y. Chung, T.-H. Kim, K.H. Jo, J.H. Hahn, Z. Bao, S. Hwang, N. Park, ACS Nano 8 (2014) 10066–10076.
[1456] M. Zhu, K. Wei, S. Lin, X. Chen, C.-C. Wu, G. Li, L. Bian, Small 14 (2018) 1702288.
[1457] Q. Feng, K. Wei, K. Zhang, B. Yang, F. Tian, G. Wang, L. Bian, NPG Asia Mater. 10 (2018) e455.
[1458] M. Nakahata, Y. Takashima, H. Yamaguchi, A. Harada, Nat. Commun. 2 (2011) 511.
[1459] H. Meng, P. Xiao, J. Gu, X. Wen, J. Xu, C. Zhao, J. Zhang, T. Chen, Chem. Commun. 50 (2014) 12277–12280.
[1460] S. Tamesue, Y. Takashima, H. Yamaguchi, S. Shinkai, A. Harada, Angew. Chemie 122 (2010) 7623–7626.
[1461] W. Deng, H. Yamaguchi, Y. Takashima, A. Harada, Angew. Chemie Int. Ed. 46 (2007) 5144–5147.
[1462] Z. Deng, Y. Guo, X. Zhao, P.X. Ma, B. Guo, Chem. Mater. 30 (2018) 1729–1742.
[1463] H. Shigemitsu, T. Fujisaku, W. Tanaka, R. Kubota, S. Minami, K. Urayama, I. Hamachi, Nat. Nanotechnol. 13 (2018) 165–172.
[1464] A. Harada, R. Kobayashi, Y. Takashima, A. Hashidzume, H. Yamaguchi, Nat. Chem. 3 (2011) 34–37.
[1465] H. Yamaguchi, Y. Kobayashi, R. Kobayashi, Y. Takashima, A. Hashidzume, A. Harada, Nat. Commun. 3 (2012) 603.
[1466] Q.-D. Hu, G.-P. Tang, P.K. Chu, Acc. Chem. Res. 47 (2014) 2017–2025.
[1467] A. Harada, Y. Takashima, H. Yamaguchi, Chem. Soc. Rev. 38 (2009) 875–882.
[1468] M. Wang, X. Zhang, L. Li, J. Wang, J. Wang, J. Ma, Z. Yuan, S.F. Lincoln, X. Guo, Macromol. Mater. Eng. 301 (2016) 191–198.
[1469] P.R.A. Chivers, D.K. Smith, Nat. Rev. Mater. 4 (2019) 463–478.
[1470] L. Voorhaar, R. Hoogenboom, Chem. Soc. Rev. 45 (2016) 4013–4031.
[1471] K. Liu, Y. Kang, Z. Wang, X. Zhang, Adv. Mater. 25 (2013) 5530–5548.
[1472] L. Saunders, P.X. Ma, Macromol. Biosci. 19 (2019) 1800313.
[1473] J. Yang, R. Bai, B. Chen, Z. Suo, Adv. Funct. Mater. 0 (2019) 1901693.
[1474] Y. Shao, H. Jia, T. Cao, D. Liu, Acc. Chem. Res. 50 (2017) 659–668.
[1475] I. Gorelikov, L.M. Field, E. Kumacheva, J. Am. Chem. Soc. 126 (2004) 15938–15939.
[1476] N. Bayat, Y. Zhang, P. Falabella, R. Menefee, J.J. Whalen, M.S. Humayun, M.E. Thompson, Sci. Transl. Med. 9 (2017) eaan3879.
[1477] J. Wu, W. Wei, L.-Y. Wang, Z.-G. Su, G.-H. Ma, Biomaterials 28 (2007) 2220–2232.
[1478] S.P. Miguel, M.P. Ribeiro, H. Brancal, P. Coutinho, I.J. Correia, Carbohydr. Polym. 111 (2014) 366–373.
[1479] X. Yang, E. Kim, Y. Liu, X.-W. Shi, G.W. Rubloff, R. Ghodssi, W.E. Bentley, Z. Pancer, G.F. Payne, Adv. Funct. Mater. 20 (2010) 1645–1652.
[1480] M. Guvendiren, J.A. Burdick, Nat. Commun. 3 (2012) 792.
[1481] A. Matsumoto, M. Tanaka, H. Matsumoto, K. Ochi, Y. Moro-oka, H. Kuwata, H. Yamada, I. Shirakawa, T. Miyazawa, H. Ishii, K. Kataoka, Y. Ogawa, Y. Miyahara, T. Suganami, Sci. Adv. 3 (2017) eaaq0723.
[1482] M.P. Lutolf, J.L. Lauer-Fields, H.G. Schmoekel, A.T. Metters, F.E. Weber, G.B. Fields, J.A. Hubbell, Proc. Natl. Acad. Sci. 100 (2003) 5413–5418.
[1483] B.P. Purcell, D. Lobb, M.B. Charati, S.M. Dorsey, R.J. Wade, K.N. Zellars, H. Doviak, S. Pettaway, C.B. Logdon, J.A. Shuman, P.D. Freels, J.H. Gorman Iii, R.C. Gorman, F.G. Spinale, J.A. Burdick, Nat. Mater. 13 (2014) 653–661.
[1484] X. Yang, G. Liu, L. Peng, J. Guo, L. Tao, J. Yuan, C. Chang, Y. Wei, L. Zhang, Adv. Funct. Mater. 27 (2017) 1703174.
[1485] H. Guo, J. Zhang, T. Xu, Z. Zhang, J. Yao, Z. Shao, Biomacromolecules 14 (2013) 2733–2738.
[1486] M.C. Koetting, J.T. Peters, S.D. Steichen, N.A. Peppas, Mater. Sci. Eng. R Rep. 93 (2015) 1–49.
[1487] I. Tomatsu, K. Peng, A. Kros, Adv. Drug Deliv. Rev. 63 (2011) 1257–1266.
[1488] L. Li, J.M. Scheiger, P.A. Levkin, Adv. Mater. 31 (2019) 1807333.
[1489] L. Klouda, A.G. Mikos, Eur. J. Pharm. Biopharm. 68 (2008) 34–45.
[1490] B. Jeong, S.W. Kim, Y.H. Bae, Adv. Drug Deliv. Rev. 54 (2002) 37–51.
[1491] J. Hu, G. Zhang, S. Liu, Chem. Soc. Rev. 41 (2012) 5933–5949.
[1492] Y. Qiu, K. Park, Adv. Drug Deliv. Rev. 53 (2001) 321–339.
[1493] Y. Lu, A.A. Aimetti, R. Langer, Z. Gu, Nat. Rev. Mater. 1 (2016) 16075.
[1494] R.O. Hynes, Science 326 (2009) 1216–1219.
[1495] C. Frantz, K.M. Stewart, V.M. Weaver, J. Cell. Sci. 123 (2010) 4195.
[1496] E. Cukierman, R. Pankov, D.R. Stevens, K.M. Yamada, Science 294 (2001) 1708–1712.
[1497] J. Lee, M.J. Cuddihy, N.A. Kotov, Tissue Eng. Part B Rev. 14 (2008) 61–86.
[1498] M. Ravi, V. Paramesh, S.R. Kaviya, E. Anuradha, F.D.P. Solomon, J. Cell. Physiol. 230 (2015) 16–26.
[1499] F. Pampaloni, E.G. Reynaud, E.H.K. Stelzer, Nat. Rev. Mol. Cell Biol. 8 (2007) 839–845.
[1500] D. Huh, G.A. Hamilton, D.E. Ingber, Trends Cell Biol. 21 (2011) 745–754.
[1501] M.F. Brizzi, G. Tarone, P. Defilippi, Curr. Opin. Cell Biol. 24 (2012) 645–651.
[1502] F.M. Watt, W.T.S. Huck, Nat. Rev. Mol. Cell Biol. 14 (2013) 467–473.
[1503] F. Gattazzo, A. Urciuolo, P. Bonaldo, Biochimica et Biophysica Acta (BBA) - General Subjects 1840 (2014) 2506–2519.
[1504] R. Langer, J.P. Vacanti, Science 260 (1993) 920–926.
[1505] V. Karageorgiou, D. Kaplan, Biomaterials 26 (2005) 5474–5491.

[1506] D.W. Hutmacher, J.T. Schantz, C.X.F. Lam, K.C. Tan, T.C. Lim, J. Tissue Eng. Regen. Med. 1 (2007) 245–260.
[1507] M.M. Stevens, J.H. George, Science 310 (2005) 1135–1138.
[1508] S. Khalil, Rapid Prototyp. J. 11 (2005) 9–17.
[1509] Q. Gao, Y. He, J.-z. Fu, A. Liu, L. Ma, Biomaterials 61 (2015) 203–215.
[1510] K. Schütz, A.-M. Placht, B. Paul, S. Brüggemeier, M. Gelinsky, A. Lode, J. Tissue Eng. Regen. Med. 11 (2017) 1574–1587.
[1511] Y. Luo, A. Lode, M. Gelinsky, Adv. Healthc. Mater. 2 (2013) 777–783.
[1512] X. Cui, K. Breitenkamp, M.G. Finn, M. Lotz, D.D. D'Lima, Tissue Eng. Part A 18 (2012) 1304–1312.
[1513] T. Billiet, E. Gevaert, T. De Schryver, M. Cornelissen, P. Dubruel, Biomaterials 35 (2014) 49–62.
[1514] L. Pescosolido, W. Schuurman, J. Malda, P. Matricardi, F. Alhaique, T. Coviello, P.R. van Weeren, W.J.A. Dhert, W.E. Hennink, T. Vermonden, Biomacromolecules 12 (2011) 1831–1838.
[1515] L.A. Hockaday, K.H. Kang, N.W. Colangelo, P.Y.C. Cheung, B. Duan, E. Malone, J. Wu, L.N. Girardi, L.J. Bonassar, H. Lipson, C.C. Chu, J.T. Butcher, Biofabrication 4 (2012) 035005.
[1516] C.B. Highley, C.B. Rodell, J.A. Burdick, Adv. Mater. 27 (2015) 5075–5079.
[1517] S. Singh, C. Prakash, S. Ramakrishna, Eur. Polym. J. 114 (2019) 234–248.
[1518] F.B. Coulter, M. Schaffner, J.A. Faber, A. Rafsanjani, R. Smith, H. Appa, P. Zilla, D. Bezuidenhout, A.R. Studart, Matter 1 (2019) 266–279.
[1519] J.C. Reichert, A. Cipitria, D.R. Epari, S. Saifzadeh, P. Krishnakanth, A. Berner, M.A. Woodruff, H. Schell, M. Mehta, M.A. Schuetz, G.N. Duda, D.W. Hutmacher, Sci. Transl. Med. 4 (2012) 141ra193.
[1520] A.E. Jakus, A.L. Rutz, S.W. Jordan, A. Kannan, S.M. Mitchell, C. Yun, K.D. Koube, S.C. Yoo, H.E. Whiteley, C.-P. Richter, R.D. Galiano, W.K. Hsu, S.R. Stock, E.L. Hsu, R.N. Shah, Sci. Transl. Med. 8 (2016) 358ra127-358ra127.
[1521] d.R. Mylène, R. Alexandre, D. Inge, C. Miguel, M. Jos, Adv. Healthc. Mater. 8 (2019) 1800418.
[1522] A. Tilman, D. Falko, K. David, V. Corina, K. Paula, L. Günter, L. Anja, G. Michael, Biofabrication 10 (2018) 045002.
[1523] L. Khoon, L. Riccardo, F.C. Pedro, C. Miguel, R.A.-O. Cesar, M.Av.D. Kim, P.W.M. Ferry, G. Debby, J.H. Gary, M. Jos, B.F.W. Tim, Biofabrication 10 (2018) 034101.
[1524] Q. Wu, D. Therriault, M.-C. Heuzey, ACS Biomater. Sci. Eng. 4 (2018) 2643–2652.
[1525] C. Xu, B. Zhang Molino, X. Wang, F. Cheng, W. Xu, P. Molino, M. Bacher, D. Su, T. Rosenau, S. Willför, G. Wallace, J. Mater. Chem. B 6 (2018) 7066–7075.
[1526] A. Abbadessa, M.M. Blokzijl, V.H.M. Mouser, P. Marica, J. Malda, W.E. Hennink, T. Vermonden, Carbohydr. Polym. 149 (2016) 163–174.
[1527] A. Lode, M. Meyer, S. Brüggemeier, B. Paul, H. Baltzer, M. Schröpfer, C. Winkelmann, F. Sonntag, M. Gelinsky, Biofabrication 8 (2016) 015015.
[1528] J. Warner, P. Soman, W. Zhu, M. Tom, S. Chen, ACS Biomater. Sci. Eng. 2 (2016) 1763–1770.
[1529] M.S. Hahn, L.J. Taite, J.J. Moon, M.C. Rowland, K.A. Ruffino, J.L. West, Biomaterials 27 (2006) 2519–2524.
[1530] M. Schuster, C. Turecek, G. Weigel, R. Saf, J. Stampfl, F. Varga, R. Liska, J. Polym. Sci. Part A: Polym. Chem. 47 (2009) 7078–7089.
[1531] W. Zhu, K.R. Tringale, S.A. Woller, S. You, S. Johnson, H. Shen, J. Schimelman, M. Whitney, J. Steinauer, W. Xu, T.L. Yaksh, Q.T. Nguyen, S. Chen, Mater. Today 21 (2018) 951–959.
[1532] G. Teng, J.G. Gregory, S.C. Joshua, P.R. Anil Kumar, S. Young-Joon, A. Anthony, J.Y. James, L. Sang Jin, Biofabrication 10 (2018) 034106.
[1533] Y. Luo, A. Lode, A.R. Akkineni, M. Gelinsky, RSC Adv. 5 (2015) 43480–43488.
[1534] T. Zhang, Y. Yan, X. Wang, Z. Xiong, F. Lin, R. Wu, R. Zhang, J. Bioact. Compat. Polym. 22 (2007) 19–29.
[1535] R. Landers, U. Hübner, R. Schmelzeisen, R. Mülhaupt, Biomaterials 23 (2002) 4437–4447.
[1536] S.E. Bakarich, R. Gorkin, R. Gately, S. Naficy, M. in het Panhuis, G.M. Spinks, Addit. Manuf. 14 (2017) 24–30.
[1537] A.R. Akkineni, T. Ahlfeld, A. Funk, A. Waske, A. Lode, M. Gelinsky, Polymers 8 (2016) 170.
[1538] P. Jiang, C. Yan, Y. Guo, X. Zhang, M. Cai, X. Jia, X. Wang, F. Zhou, Biomater. Sci. 7 (2019) 1805–1814.
[1539] Z. Wang, G. An, Y. Zhu, X. Liu, Y. Chen, H. Wu, Y. Wang, X. Shi, C. Mao, Mater. Horiz. 6 (2019) 733–742.
[1540] M. Kesti, M. Müller, J. Becher, M. Schnabelrauch, M. D'Este, D. Eglin, M. Zenobi-Wong, Acta Biomater. 11 (2015) 162–172.
[1541] F. Gao, Z. Xu, Q. Liang, H. Li, L. Peng, M. Wu, X. Zhao, X. Cui, C. Ruan, W. Liu, Adv. Sci. 6 (2019) 1900867.
[1542] C. Loebel, C.B. Rodell, M.H. Chen, J.A. Burdick, Nat. Protocols 12 (2017) 1521–1541.
[1543] F. Gao, Z. Xu, Q. Liang, B. Liu, H. Li, Y. Wu, Y. Zhang, Z. Lin, M. Wu, C. Ruan, W. Liu, Adv. Funct. Mater. 28 (2018) 1706644.
[1544] S. Hong, D. Sycks, H.F. Chan, S. Lin, G.P. Lopez, F. Guilak, K.W. Leong, X. Zhao, Adv. Mater. 27 (2015) 4035–4040.
[1545] J.L. Álvaro, S. Bapi, Z. Tobias, S. Raquel, F.M. João, R.B. Aldo, Biofabrication 8 (2016) 035005.
[1546] L. Kool, A. Bunschoten, A.H. Velders, V. Saggiomo, Beilstein J. Nanotechnol. 10 (2019) 442–447.
[1547] X. Zhai, Y. Ma, C. Hou, F. Gao, Y. Zhang, C. Ruan, H. Pan, W.W. Lu, W. Liu, ACS Biomater. Sci. Eng. 3 (2017) 1109–1118.
[1548] S. Sayyar, S. Gambhir, J. Chung, D.L. Officer, G.G. Wallace, Nanoscale 9 (2017) 2038–2050.
[1549] W. Zhu, B. Holmes, R.I. Glazer, L.G. Zhang, Nanomed. Nanotechnol. Biol. Med. 12 (2016) 69–79.
[1550] N.J. Castro, J. O'Brien, L.G. Zhang, Nanoscale 7 (2015) 14010–14022.
[1551] Y. Jin, C. Liu, W. Chai, A. Compaan, Y. Huang, ACS Appl. Mater. Interfaces 9 (2017) 17456–17465.
[1552] T. Ahlfeld, G. Cidonio, D. Kilian, S. Duin, A.R. Akkineni, J.I. Dawson, S. Yang, A. Lode, R.O.C. Oreffo, M. Gelinsky, Biofabrication 9 (2017) 034103.
[1553] A.L. Rutz, K.E. Hyland, A.E. Jakus, W.R. Burghardt, R.N. Shah, Adv. Mater. 27 (2015) 1607–1614.
[1554] K. Arcaute, B. Mann, R. Wicker, Acta Biomater. 6 (2010) 1047–1054.
[1555] A. Nadernezhad, N. Khani, G.A. Skvortsov, B. Toprakhisar, E. Bakirci, Y. Menceloglu, S. Unal, B. Koc, Sci. Rep. 6 (2016) 33178.
[1556] T. Ahlfeld, F.P. Schuster, Y. Förster, M. Quade, A.R. Akkineni, C. Rentsch, S. Rammelt, M. Gelinsky, A. Lode, Adv. Healthc. Mater. 8 (2019) 1801512.
[1557] J.R. Tumbleston, D. Shirvanyants, N. Ermoshkin, R. Januszewicz, A.R. Johnson, D. Kelly, K. Chen, R. Pinschmidt, J.P. Rolland, A. Ermoshkin, E.T. Samulski, J.M. DeSimone, Science 347 (2015) 1349–1352.
[1558] B.E. Kelly, I. Bhattacharya, H. Heidari, M. Shusteff, C.M. Spadaccini, H.K. Taylor, Science 363 (2019) 1075–1079.
[1559] J. Wang, T. Lu, M. Yang, D. Sun, Y. Xia, T. Wang, Sci. Adv. 5 (2019) eaau8769.
[1560] M.A. Skylar-Scott, J. Mueller, C.W. Visser, J.A. Lewis, Nature 575 (2019) 330–335.
[1561] S. Tibbits, Archit. Des. 84 (2014) 116–121.
[1562] R.L. Truby, J.A. Lewis, Nature 540 (2016) 371–378.
[1563] Y. Zhang, F. Zhang, Z. Yan, Q. Ma, X. Li, Y. Huang, J.A. Rogers, Nat. Rev. Mater. 2 (2017) 17019.
[1564] X. Kuang, D.J. Roach, J. Wu, C.M. Hamel, Z. Ding, T. Wang, M.L. Dunn, H.J. Qi, Adv. Funct. Mater. 29 (2019) 1805290.
[1565] S.E. Bakarich, R. Gorkin Iii, Mih. Panhuis, G.M. Spinks, Macromol. Rapid Commun. 36 (2015) 1211–1217.
[1566] A. Sydney Gladman, E.A. Matsumoto, R.G. Nuzzo, L. Mahadevan, J.A. Lewis, Nat. Mater. 15 (2016) 413–418.
[1567] S. Naficy, R. Gately, R. Gorkin Iii, H. Xin, G.M. Spinks, Macromol. Mater. Eng. 302 (2017) 1600212.
[1568] H. Yang, C. Li, M. Yang, Y. Pan, Q. Yin, J. Tang, H.J. Qi, Z. Suo, Adv. Funct. Mater. 29 (2019) 1901721.
[1569] Z. Ji, C. Yan, B. Yu, X. Zhang, M. Cai, X. Jia, X. Wang, F. Zhou, Adv. Mater. Technol. 4 (2019) 1800713.
[1570] J. Odent, S. Vanderstappen, A. Toncheva, E. Pichon, T.J. Wallin, K. Wang, R.F. Shepherd, P. Dubois, J.-M. Raquez, J. Mater. Chem. A 7 (2019) 15395–15403.
[1571] Z. Ding, C. Yuan, X. Peng, T. Wang, H.J. Qi, M.L. Dunn, Sci. Adv. 3 (2017) e1602890.
[1572] Q. Ge, H.J. Qi, M.L. Dunn, Appl. Phys. Lett. 103 (2013) 131901.
[1573] Q. Zhang, K. Zhang, G. Hu, Sci. Rep. 6 (2016) 22431.
[1574] Q. Zhang, D. Yan, K. Zhang, G. Hu, Sci. Rep. 5 (2015) 8936.
[1575] T. van Manen, S. Janbaz, A.A. Zadpoor, Mater. Today 21 (2018) 144–163.
[1576] T. van Manen, S. Janbaz, A.A. Zadpoor, Mater. Horiz. 4 (2017) 1064–1069.
[1577] M. Wehner, R.L. Truby, D.J. Fitzgerald, B. Mosadegh, G.M. Whitesides, J.A. Lewis, R.J. Wood, Nature 536 (2016) 451–455.
[1578] Q. Ge, A.H. Sakhaei, H. Lee, C.K. Dunn, N.X. Fang, M.L. Dunn, Sci. Rep. 6 (2016) 31110.
[1579] M. Hospodiuk, M. Dey, D. Sosnoski, I.T. Ozbolat, Biotechnol. Adv. 35 (2017) 217–239.
[1580] V. Mironov, T. Boland, T. Trusk, G. Forgacs, R.R. Markwald, Trends Biotechnol. 21 (2003) 157–161.
[1581] S. Khalil, W. Sun, J. Biomech. Eng. 131 (2009) 111002-111002-111008.
[1582] F. Guilak, D.M. Cohen, B.T. Estes, J.M. Gimble, W. Liedtke, C.S. Chen, Cell Stem Cell 5 (2009) 17–26.
[1583] Z. Zhang, Y. Jin, J. Yin, C. Xu, R. Xiong, K. Christensen, B.R. Ringeisen, D.B. Chrisey, Y. Huang, Appl. Phys. Rev. 5 (2018) 041304.
[1584] J. Jang, J.Y. Park, G. Gao, D.-W. Cho, Biomaterials 156 (2018) 88–106.
[1585] L. Valot, J. Martinez, A. Mehdi, G. Subra, Chem. Soc. Rev. 48 (2019) 4049–4086.
[1586] D. Chimene, R. Kaunas, A.K. Gaharwar, Advanced Materials 32 (2020) 1902026.
[1587] F. You, X. Wu, N. Zhu, M. Lei, B.F. Eames, X. Chen, ACS Biomater. Sci. Eng. 2 (2016) 1200–1210.
[1588] H.W. Ooi, C. Mota, A.T. ten Cate, A. Calore, L. Moroni, M.B. Baker, Biomacromolecules 19 (2018) 3390–3400.
[1589] E.O. Osidak, P.A. Karalkin, M.S. Osidak, V.A. Parfenov, D.E. Sivogrivov, F.D.A.S. Pereira, A.A. Gryadunova, E.V. Koudan, Y.D. Khesuani, V.A. Kasyanov, S.I. Belousov, S.V. Krasheninnikov, T.E. Grigoriev, S.N. Chvalun, E.A. Bulanova, V.A. Mironov, S.P. Domogatsky, J. Mater. Sci. Mater. Med. 30 (2019) 31.
[1590] S. Rhee, J.L. Puetzer, B.N. Mason, C.A. Reinhart-King, L.J. Bonassar, ACS Biomater. Sci. Eng. 2 (2016) 1800–1805.
[1591] N. Diamantides, L. Wang, T. Pruiksma, J. Siemiatkoski, C. Dugopolski, S. Shortkroff, S. Kennedy, L.J. Bonassar, Biofabrication 9 (2017) 034102.
[1592] R. Pereira, A. Sousa, C.C. Barrias, P. Bartolo, P.L. Granja, Mater. Horiz. 5 (2018) 1100–1111.
[1593] X. Liu, H. Yuk, S. Lin, G.A. Parada, T.-C. Tang, E. Tham, C. de la Fuente-Nunez, T.K. Lu, X. Zhao, Adv. Mater. 30 (2018) 1704821.
[1594] S. Bertlein, G. Brown, K.S. Lim, T. Jungst, T. Boeck, T. Blunk, J. Tessmar, G.J. Hooper, T.B.F. Woodfield, J. Groll, Adv. Mater. 29 (2017) 1703404.
[1595] A. Tijore, J.-M. Behr, S.A. Irvine, V. Baisane, S. Venkatraman, Biomed. Microdevices 20 (2018) 32.
[1596] W. Liu, M.A. Heinrich, Y. Zhou, A. Akpek, N. Hu, X. Liu, X. Guan, Z. Zhong, X. Jin, A. Khademhosseini, Y.S. Zhang, Adv. Healthc. Mater. 6 (2017) 1601451.
[1597] M. Tromayer, A. Dobos, P. Gruber, A. Ajami, R. Dedic, A. Ovsianikov, R. Liska,

Polym. Chem. 9 (2018) 3108–3117.
[1598] M.T. Poldervaart, B. Goversen, M. de Ruijter, A. Abbadessa, F.P.W. Melchels, F.C. Öner, W.J.A. Dhert, T. Vermonden, J. Alblas, PLoS One 12 (2017) e0177628.
[1599] K. Schacht, T. Jüngst, M. Schweinlin, A. Ewald, J. Groll, T. Scheibel, Angew. Chemie Int. Ed. 54 (2015) 2816–2820.
[1600] H. Lin, D. Zhang, P.G. Alexander, G. Yang, J. Tan, A.W.-M. Cheng, R.S. Tuan, Biomaterials 34 (2013) 331–339.
[1601] K. Arcaute, B.K. Mann, R.B. Wicker, Ann. Biomed. Eng. 34 (2006) 1429–1441.
[1602] Q. Chen, H. Chen, L. Zhu, J. Zheng, J. Mater. Chem. B 3 (2015) 3654–3676.
[1603] S.E. Bakarich, Mih. Panhuis, S. Beirne, G.G. Wallace, G.M. Spinks, J. Mater. Chem. B 1 (2013) 4939–4946.
[1604] G.R. López-Marcial, A.Y. Zeng, C. Osuna, J. Dennis, J.M. García, G.D. O'Connell, ACS Biomater. Sci. Eng. 4 (2018) 3610–3616.
[1605] T. Xu, W. Zhao, J.-M. Zhu, M.Z. Albanna, J.J. Yoo, A. Atala, Biomaterials 34 (2013) 130–139.
[1606] J.P.K. Armstrong, M. Burke, B.M. Carter, S.A. Davis, A.W. Perriman, Adv. Healthc. Mater. 5 (2016) 1724–1730.
[1607] N. Faramarzi, I.K. Yazdi, M. Nabavinia, A. Gemma, A. Fanelli, A. Caizzone, L.M. Ptaszek, I. Sinha, A. Khademhosseini, J.N. Ruskin, A. Tamayol, Adv. Healthc. Mater. 7 (2018) 1701347.
[1608] L.J. Pourchet, A. Thepot, M. Albouy, E.J. Courtial, A. Boher, L.J. Blum, C.A. Marquette, Adv. Healthc. Mater. 6 (2017) 1601101.
[1609] Q. Gu, E. Tomaskovic-Crook, G.G. Wallace, J.M. Crook, Adv. Healthc. Mater. 6 (2017) 1700175.
[1610] Q. Gu, E. Tomaskovic-Crook, R. Lozano, Y. Chen, R.M. Kapsa, Q. Zhou, G.G. Wallace, J.M. Crook, Adv. Healthc. Mater. 5 (2016) 1429–1438.
[1611] M. Neufurth, X. Wang, H.C. Schröder, Q. Feng, B. Diehl-Seifert, T. Ziebart, R. Steffen, S. Wang, W.E.G. Müller, Biomaterials 35 (2014) 8810–8819.
[1612] Y. He, F. Yang, H. Zhao, Q. Gao, B. Xia, J. Fu, Sci. Rep. 6 (2016) 29977.
[1613] B. Duan, L.A. Hockaday, K.H. Kang, J.T. Butcher, J. Biomed. Mater. Res. A. 101A (2013) 1255–1264.
[1614] N. Law, B. Doney, H. Glover, Y. Qin, Z.M. Aman, T.B. Sercombe, L.J. Liew, R.J. Dilley, B.J. Doyle, J. Mech. Behav. Biomed. Mater. 77 (2018) 389–399.
[1615] F. Ruther, T. Distler, A.R. Boccaccini, R. Detsch, J. Mater. Sci. Mater. Med. 30 (2018) 8.
[1616] M. Schaffner, P.A. Rühs, F. Coulter, S. Kilcher, A.R. Studart, Sci. Adv. 3 (2017) eaao6804.
[1617] J. Huang, S. Liu, C. Zhang, X. Wang, J. Pu, F. Ba, S. Xue, H. Ye, T. Zhao, K. Li, Y. Wang, J. Zhang, L. Wang, C. Fan, T.K. Lu, C. Zhong, Nat. Chem. Biol. 15 (2019) 34–41.
[1618] J.L. Connell, E.T. Ritschdorff, M. Whiteley, J.B. Shear, Proc. Natl. Acad. Sci. 110 (2013) 18380–18385.
[1619] K.S. Lim, B.S. Schon, N.V. Mekhileri, G.C.J. Brown, C.M. Chia, S. Prabakar, G.J. Hooper, T.B.F. Woodfield, ACS Biomater. Sci. Eng. 2 (2016) 1752–1762.
[1620] W. Zhu, X. Qu, J. Zhu, X. Ma, S. Patel, J. Liu, P. Wang, C.S.E. Lai, M. Gou, Y. Xu, K. Zhang, S. Chen, Biomaterials 124 (2017) 106–115.
[1621] H.M.M. Vivian, P.W.M. Ferry, V. Jetze, J.A.D. Wouter, G. Debby, M. Jos, Biofabrication 8 (2016) 035003.
[1622] G.-L. Ying, N. Jiang, S. Maharjan, Y.-X. Yin, R.-R. Chai, X. Cao, J.-Z. Yang, A.K. Miri, S. Hassan, Y.S. Zhang, Adv. Mater. 30 (2018) 1805460.
[1623] W. Zhu, H. Cui, B. Boualam, F. Masood, E. Flynn, R.D. Rao, Z.-Y. Zhang, L.G. Zhang, Nanotechnology 29 (2018) 185101.
[1624] Z. Wang, R. Abdulla, B. Parker, R. Samanipour, S. Ghosh, K. Kim, Biofabrication 7 (2015) 045009.
[1625] D. Wu, Y. Yu, J. Tan, L. Huang, B. Luo, L. Lu, C. Zhou, Mater. Des. 160 (2018) 486–495.
[1626] M.T. Poldervaart, H. Gremmels, K. van Deventer, J.O. Fledderus, F.C. Öner, M.C. Verhaar, W.J.A. Dhert, J. Alblas, J. Control. Release 184 (2014) 58–66.
[1627] S. Das, F. Pati, Y.-J. Choi, G. Rijal, J.-H. Shim, S.W. Kim, A.R. Ray, D.-W. Cho, S. Ghosh, Acta Biomater. 11 (2015) 233–246.
[1628] H. Kwak, S. Shin, H. Lee, J. Hyun, J. Ind. Eng. Chem. 72 (2019) 232–240.
[1629] Z. Zheng, J. Wu, M. Liu, H. Wang, C. Li, M.J. Rodriguez, G. Li, X. Wang, D.L. Kaplan, Adv. Healthc. Mater. 7 (2018) 1701026.
[1630] A.M. Pekkanen, R.J. Mondschein, C.B. Williams, T.E. Long, Biomacromolecules 18 (2017) 2669–2687.
[1631] C. Li, A. Faulkner-Jones, A.R. Dun, J. Jin, P. Chen, Y. Xing, Z. Yang, Z. Li, W. Shu, D. Liu, R.R. Duncan, Angew. Chemie Int. Ed. 54 (2015) 3957–3961.
[1632] G.M. Whitesides, B. Grzybowski, Science 295 (2002) 2418–2421.
[1633] A. Nadernezhad, O.S. Caliskan, F. Topuz, F. Afghah, B. Erman, B. Koc, ACS Applied Bio Materials 2 (2019) 796–806.
[1634] K. Markstedt, A. Mantas, I. Tournier, H. Martínez Ávila, D. Hägg, P. Gatenholm, Biomacromolecules 16 (2015) 1489–1496.
[1635] L.K. Narayanan, P. Huebner, M.B. Fisher, J.T. Spang, B. Starly, R.A. Shirwaiker, ACS Biomater. Sci. Eng. 2 (2016) 1732–1742.
[1636] E. Trampe, K. Koren, A.R. Akkineni, C. Senwitz, F. Krujatz, A. Lode, M. Gelinsky, M. Kühl, Adv. Funct. Mater. 28 (2018) 1804411.
[1637] S.A. Wilson, L.M. Cross, C.W. Peak, A.K. Gaharwar, ACS Appl. Mater. Interfaces 9 (2017) 43449–43458.
[1638] T.T. Demirtaş, G. Irmak, M. Gümüşderelioğlu, Biofabrication 9 (2017) 035003.
[1639] C.W. Peak, J. Stein, K.A. Gold, A.K. Gaharwar, Langmuir 34 (2018) 917–925.
[1640] F. Qian, C. Zhu, J.M. Knipe, S. Ruelas, J.K. Stolaroff, J.R. DeOtte, E.B. Duoss, C.M. Spadaccini, C.A. Henard, M.T. Guarnieri, S.E. Baker, Nano Lett. 19 (2019) 5829–5835.
[1641] G. Gao, A.F. Schilling, T. Yonezawa, J. Wang, G. Dai, X. Cui, Biotechnol. J. 9 (2014) 1304–1311.
[1642] C.W. Peak, K.A. Singh, Ma. Adlouni, J. Chen, A.K. Gaharwar, Adv. Healthc. Mater. 8 (2019) 1801553.
[1643] G. Cidonio, C.R. Alcala-Orozco, K.S. Lim, M. Glinka, I. Mutreja, Y.-H. Kim, J.I. Dawson, T.B.F. Woodfield, R.O.C. Oreffo, Biofabrication 11 (2019) 035027.
[1644] K. Zhu, S.R. Shin, T. van Kempen, Y.-C. Li, V. Ponraj, A. Nasajpour, S. Mandla, N. Hu, X. Liu, J. Leijten, Y.-D. Lin, M.A. Hussain, Y.S. Zhang, A. Tamayol, A. Khademhosseini, Adv. Funct. Mater. 27 (2017) 1605352.
[1645] D. Chimene, C.W. Peak, J. Gentry, J.K. Carrow, L.M. Cross, E. Mondragon, G.B.C. Cardoso, R. Kaunas, A.K. Gaharwar, ACS Appl. Mater. Interfaces 10 (2018) 9957–9968.
[1646] A. Skardal, J. Zhang, G.D. Prestwich, Biomaterials 31 (2010) 6173–6181.
[1647] W. Liu, Y.S. Zhang, M.A. Heinrich, F. De Ferrari, H.L. Jang, S.M. Bakht, M.M. Alvarez, J. Yang, Y.-C. Li, G. Trujillo-de Santiago, A.K. Miri, K. Zhu, P. Khoshakhlagh, G. Prakash, H. Cheng, X. Guan, Z. Zhong, J. Ju, G.H. Zhu, X. Jin, S.R. Shin, M.R. Dokmeci, A. Khademhosseini, Adv. Mater. 29 (2017) 1604630.
[1648] A. Blaeser, D.F. Duarte Campos, U. Puster, W. Richtering, M.M. Stevens, H. Fischer, Adv. Healthc. Mater. 5 (2016) 326–333.
[1649] T.J. Hinton, Q. Jallerat, R.N. Palchesko, J.H. Park, M.S. Grodzicki, H.-J. Shue, M.H. Ramadan, A.R. Hudson, A.W. Feinberg, Sci. Adv. 1 (2015) e1500758.
[1650] D. Joung, V. Truong, C.C. Neitzke, S.-Z. Guo, P.J. Walsh, J.R. Monat, F. Meng, S.H. Park, J.R. Dutton, A.M. Parr, M.C. McAlpine, Adv. Funct. Mater. 28 (2018) 1801850.
[1651] B. Duan, E. Kapetanovic, L.A. Hockaday, J.T. Butcher, Acta Biomater. 10 (2014) 1836–1846.
[1652] D.B. Kolesky, R.L. Truby, A.S. Gladman, T.A. Busbee, K.A. Homan, J.A. Lewis, Adv. Mater. 26 (2014) 3124–3130.
[1653] R. Levato, W.R. Webb, I.A. Otto, A. Mensinga, Y. Zhang, M. van Rijen, R. van Weeren, I.M. Khan, J. Malda, Acta Biomater. 61 (2017) 41–53.
[1654] D.B. Kolesky, K.A. Homan, M.A. Skylar-Scott, J.A. Lewis, Proc. Natl. Acad. Sci. 113 (2016) 3179–3184.
[1655] H.-W. Kang, S.J. Lee, I.K. Ko, C. Kengla, J.J. Yoo, A. Atala, Nat. Biotechnol. 34 (2016) 312–319.
[1656] X. Zhai, C. Ruan, Y. Ma, D. Cheng, M. Wu, W. Liu, X. Zhao, H. Pan, W.W. Lu, Adv. Sci. 5 (2018) 1700550.
[1657] A.K. Miri, D. Nieto, L. Iglesias, H. Goodarzi Hosseinabadi, S. Maharjan, G.U. Ruiz-Esparza, P. Khoshakhlagh, A. Manbachi, M.R. Dokmeci, S. Chen, S.R. Shin, Y.S. Zhang, A. Khademhosseini, Adv. Mater. 30 (2018) 1800242.
[1658] L. Ouyang, J.P.K. Armstrong, Q. Chen, Y. Lin, M.M. Stevens, Adv. Funct. Mater. 30 (2020) 1908349.
[1659] G. Luo, Y. Yu, Y. Yuan, X. Chen, Z. Liu, T. Kong, Adv. Mater. 31 (2019) 1904631.
[1660] B. Grigoryan, S.J. Paulsen, D.C. Corbett, D.W. Sazer, C.L. Fortin, A.J. Zaita, P.T. Greenfield, N.J. Calafat, J.P. Gounley, A.H. Ta, F. Johansson, A. Randles, J.E. Rosenkrantz, J.D. Louis-Rosenberg, P.A. Galie, K.R. Stevens, J.S. Miller, Science 364 (2019) 458–464.
[1661] A. Kirillova, R. Maxson, G. Stoychev, C.T. Gomillion, L. Ionov, Adv. Mater. 29 (2017) 1703443.
[1662] S. Miao, H. Cui, M. Nowicki, L. Xia, X. Zhou, S.-J. Lee, W. Zhu, K. Sarkar, Z. Zhang, G. Zhang Lijie, Adv. Biosyst. 2 (2018) 1800101.
[1663] K. Kuribayashi-Shigetomi, H. Onoe, S. Takeuchi, PLoS One 7 (2012) e51085.
[1664] H.-p. Lee, L. Gu, D.J. Mooney, M.E. Levenston, O. Chaudhuri, Nat. Mater. 16 (2017) 1243–1251.
[1665] X. Wang, S. Xu, S. Zhou, W. Xu, M. Leary, P. Choong, M. Qian, M. Brandt, Y.M. Xie, Biomaterials 83 (2016) 127–141.
[1666] H. Cui, W. Zhu, B. Holmes, L.G. Zhang, Adv. Sci. 3 (2016) 1600058.
[1667] N. Ashammakhi, A. Hasan, O. Kaarela, B. Byambaa, A. Sheikhi, A.K. Gaharwar, A. Khademhosseini, Adv. Healthc. Mater. 8 (2019) 1801048.
[1668] J.-Y. Rho, L. Kuhn-Spearing, P. Zioupos, Med. Eng. Phys. 20 (1998) 92–102.
[1669] D. Tang, R.S. Tare, L.-Y. Yang, D.F. Williams, K.-L. Ou, R.O.C. Oreffo, Biomaterials 83 (2016) 363–382.
[1670] U.G.K. Wegst, H. Bai, E. Saiz, A.P. Tomsia, R.O. Ritchie, Nat. Mater. 14 (2014) 23–36.
[1671] B. Byambaa, N. Annabi, K. Yue, G. Trujillo-de Santiago, M.M. Alvarez, W. Jia, M. Kazemzadeh-Narbat, S.R. Shin, A. Tamayol, A. Khademhosseini, Adv. Healthc. Mater. 6 (2017) 1700015.
[1672] H. Cui, W. Zhu, M. Nowicki, X. Zhou, A. Khademhosseini, L.G. Zhang, Adv. Healthc. Mater. 5 (2016) 2174–2181.
[1673] W. Schuurman, T.J. Klein, W.J.A. Dhert, P.R. van Weeren, D.W. Hutmacher, J. Malda, J. Tissue Eng. Regen. Med. 9 (2015) 669–678.
[1674] C. Di Bella, A. Fosang, D.M. Donati, G.G. Wallace, P.F.M. Choong, Front. Surg. 2 (2015).
[1675] K. Matti, E. Christian, P. Guglielmo, K. David, G. Daniel, B. Andreas, Z.-W. Marcy, Adv. Funct. Mater. 25 (2015) 7406–7417.
[1676] P.N. Bernal, P. Delrot, D. Loterie, Y. Li, J. Malda, C. Moser, R. Levato, Adv. Mater. 31 (2019) 1904209.
[1677] A.C. Daly, F.E. Freeman, T. Gonzalez-Fernandez, S.E. Critchley, J. Nulty, D.J. Kelly, Adv. Healthc. Mater. 6 (2017) 1700298.
[1678] L.M. Ramer, M.S. Ramer, E.J. Bradbury, Lancet Neurol. 13 (2014) 1241–1256.
[1679] J. Koffler, W. Zhu, X. Qu, O. Platoshyn, J.N. Dulin, J. Brock, L. Graham, P. Lu, J. Sakamoto, M. Marsala, S. Chen, M.H. Tuszynski, Nat. Med. 25 (2019) 263–269.
[1680] S. Ostrovidov, S. Salehi, M. Costantini, K. Suthiwanich, M. Ebrahimi, R.B. Sadeghian, T. Fujie, X. Shi, S. Cannata, C. Gargioli, A. Tamayol, M.R. Dokmeci, G. Orive, W. Swieszkowski, A. Khademhosseini, Small 15 (2019) 1805530.
[1681] N.E. Fedorovich, J. Alblas, J.R. de Wijn, W.E. Hennink, A.J. Verbout, W.J. Dhert, Tissue Eng. 13 (2007) 1905–1925.
[1682] T.K. Merceron, M. Burt, Y.-J. Seol, H.-W. Kang, S.J. Lee, J.J. Yoo, A. Atala,

Biofabrication 7 (2015) 035003.
[1683] M. Costantini, S. Testa, P. Mozetic, A. Barbetta, C. Fuoco, E. Fornetti, F. Tamiro, S. Bernardini, J. Jaroszewicz, W. Święszkowski, M. Trombetta, L. Castagnoli, D. Seliktar, P. Garstecki, G. Cesareni, S. Cannata, A. Rainer, C. Gargioli, Biomaterials 131 (2017) 98–110.
[1684] J.M. Grasman, M.J. Zayas, R.L. Page, G.D. Pins, Acta Biomater. 25 (2015) 2–15.
[1685] V. Lee, G. Singh, J.P. Trasatti, C. Bjornsson, X. Xu, T.N. Tran, S.-S. Yoo, G. Dai, P. Karande, Tissue Eng. Part C Methods 20 (2013) 473–484.
[1686] L. Koch, A. Deiwick, S. Schlie, S. Michael, M. Gruene, V. Coger, D. Zychlinski, A. Schambach, K. Reimers, P.M. Vogt, B. Chichkov, Biotechnol. Bioeng. 109 (2012) 1855–1863.
[1687] B.S. Kim, G. Gao, J.Y. Kim, D.-W. Cho, Adv. Healthc. Mater. 8 (2019) 1801019.
[1688] S. Vijayavenkataraman, W.F. Lu, J.Y.H. Fuh, Biofabrication 8 (2016) 032001.
[1689] W.L. Ng, S. Wang, W.Y. Yeong, M.W. Naing, Trends Biotechnol. 34 (2016) 689–699.
[1690] W.-C. Yan, P. Davoodi, S. Vijayavenkataraman, Y. Tian, W.C. Ng, J.Y.H. Fuh, K.S. Robinson, C.-H. Wang, Adv. Drug Deliv. Rev. 132 (2018) 270–295.
[1691] H. Bae, A.S. Puranik, R. Gauvin, F. Edalat, B. Carrillo-Conde, N.A. Peppas, A. Khademhosseini, Sci. Transl. Med. 4 (2012) 160ps123.
[1692] E.C. Novosel, C. Kleinhans, P.J. Kluger, Adv. Drug Deliv. Rev. 63 (2011) 300–311.
[1693] J.S. Miller, K.R. Stevens, M.T. Yang, B.M. Baker, D.-H.T. Nguyen, D.M. Cohen, E. Toro, A.A. Chen, P.A. Galie, X. Yu, R. Chaturvedi, S.N. Bhatia, C.S. Chen, Nat. Mater. 11 (2012) 768–774.
[1694] W. Jia, P.S. Gungor-Ozkerim, Y.S. Zhang, K. Yue, K. Zhu, W. Liu, Q. Pi, B. Byambaa, M.R. Dokmeci, S.R. Shin, A. Khademhosseini, Biomaterials 106 (2016) 58–68.
[1695] G. Gao, J.Y. Park, B.S. Kim, J. Jang, D.-W. Cho, Adv. Healthc. Mater. 7 (2018) 1801102.
[1696] D. Lei, Y. Yang, Z. Liu, B. Yang, W. Gong, S. Chen, S. Wang, L. Sun, B. Song, H. Xuan, X. Mo, B. Sun, S. Li, Q. Yang, S. Huang, S. Chen, Y. Ma, W. Liu, C. He, B. Zhu, E.M. Jeffries, F.-L. Qing, X. Ye, Q. Zhao, Z. You, Mater. Horiz. 6 (2019) 1197–1206.
[1697] T. Mirabella, J.W. MacArthur, D. Cheng, C.K. Ozaki, Y.J. Woo, M.T. Yang, C.S. Chen, Nat. Biomed. Eng. 1 (2017) 0083.
[1698] L. Shao, Q. Gao, H. Zhao, C. Xie, J. Fu, Z. Liu, M. Xiang, Y. He, Small 14 (2018) 1802187.
[1699] P. Datta, B. Ayan, I.T. Ozbolat, Acta Biomater. 51 (2017) 1–20.
[1700] B. Duan, Ann. Biomed. Eng. 45 (2017) 195–209.
[1701] M. De Vos, J. Smitz, T.K. Woodruff, Lancet 384 (2014) 1302–1310.
[1702] J.S. Jeruss, T.K. Woodruff, N. Engl. J. Med. 360 (2009) 902–911.
[1703] M.M. Laronda, A.E. Jakus, K.A. Whelan, J.A. Wertheim, R.N. Shah, T.K. Woodruff, Biomaterials 50 (2015) 20–29.
[1704] M. Rosendahl, M.T. Andersen, E. Ralfkiær, L. Kjeldsen, M.K. Andersen, C.Y. Andersen, Fertil. Steril. 94 (2010) 2186–2190.
[1705] P.K. Kreeger, J.W. Deck, T.K. Woodruff, L.D. Shea, Biomaterials 27 (2006) 714–723.
[1706] A. Shikanov, M. Xu, T.K. Woodruff, L.D. Shea, Biomaterials 30 (2009) 5476–5485.
[1707] J. Vanacker, V. Luyckx, M.-M. Dolmans, A. Des Rieux, J. Jaeger, A. Van Langendonckt, J. Donnez, C.A. Amorim, Biomaterials 33 (2012) 6079–6085.
[1708] J. Kim, A.S. Perez, J. Claflin, A. David, H. Zhou, A. Shikanov, NPJ Regen. Med. 1 (2016) 16010.
[1709] M.M. Laronda, A.L. Rutz, S. Xiao, K.A. Whelan, F.E. Duncan, E.W. Roth, T.K. Woodruff, R.N. Shah, Nat. Commun. 8 (2017) 15261.
[1710] T. Desai, L.D. Shea, Nat. Rev. Drug Discov. 16 (2016) 338–350.
[1711] A. Citro, P.T. Moser, E. Dugnani, T.K. Rajab, X. Ren, D. Evangelista-Leite, J.M. Charest, A. Peloso, B.K. Podesser, F. Manenti, S. Pellegrini, L. Piemonti, H.C. Ott, Biomaterials 199 (2019) 40–51.
[1712] X. Liu, S.-S.D. Carter, M.J. Renes, J. Kim, D.M. Rojas-Canales, D. Penko, C. Angus, S. Beirne, C.J. Drogemuller, Z. Yue, P.T. Coates, G.G. Wallace, Adv. Healthc. Mater. 8 (2019) 1801181.
[1713] G. Marchioli, L. van Gurp, P.P. van Krieken, D. Stamatialis, M. Engelse, C.A. van Blitterswijk, M.B.J. Karperien, E. de Koning, J. Alblas, L. Moroni, A.A. van Apeldoorn, Biofabrication 7 (2015) 025009.
[1714] S. Duin, K. Schütz, T. Ahlfeld, S. Lehmann, A. Lode, B. Ludwig, M. Gelinsky, Adv. Healthc. Mater. 8 (2019) 1801631.
[1715] E.J. Benjamin, M.J. Blaha, S.E. Chiuve, M. Cushman, S.R. Das, R. Deo, S.D. de Ferranti, J. Floyd, M. Fornage, C. Gillespie, C.R. Isasi, M.C. Jiménez, L.C. Jordan, S.E. Judd, D. Lackland, J.H. Lichtman, L. Lisabeth, S. Liu, C.T. Longenecker, R.H. Mackey, K. Matsushita, D. Mozaffarian, M.E. Mussolino, K. Nasir, R.W. Neumar, L. Palaniappan, D.K. Pandey, R.R. Thiagarajan, M.J. Reeves, M. Ritchey, C.J. Rodriguez, G.A. Roth, W.D. Rosamond, C. Sasson, A. Towfighi, C.W. Tsao, M.B. Turner, S.S. Virani, J.H. Voeks, J.Z. Willey, J.T. Wilkins, J.H. Wu, H.M. Alger, S.S. Wong, P. Muntner, C. American Heart Association Statistics, S. Stroke Statistics, Circulation 135 (2017) e146–e603.
[1716] Y. Zhao, N. Rafatian, N.T. Feric, B.J. Cox, R. Aschar-Sobbi, E.Y. Wang, P. Aggarwal, B. Zhang, G. Conant, K. Ronaldson-Bouchard, A. Pahnke, S. Protze, J.H. Lee, L. Davenport Huyer, D. Jekic, A. Wickeler, H.E. Naguib, G.M. Keller, G. Vunjak-Novakovic, U. Broeckel, P.H. Backx, M. Radisic, Cell 176 (2019) 913–927.
[1717] G. Vunjak-Novakovic, N. Tandon, A. Godier, R. Maidhof, A. Marsano, T.P. Martens, M. Radisic, Tissue Eng. Part B Rev. 16 (2009) 169–187.
[1718] N. Noor, A. Shapira, R. Edri, I. Gal, L. Wertheim, T. Dvir, Adv. Sci. 6 (2019) 1900344.
[1719] A. Lee, A.R. Hudson, D.J. Shiwarski, J.W. Tashman, T.J. Hinton, S. Yerneni, J.M. Bliley, P.G. Campbell, A.W. Feinberg, Science 365 (2019) 482–487.
[1720] A. Arslan-Yildiz, R.E. Assal, P. Chen, S. Guven, F. Inci, U. Demirci, Biofabrication 8 (2016) 014103.
[1721] S. Mehrotra, J.C. Moses, A. Bandyopadhyay, B.B. Mandal, ACS Applied Bio Materials 2 (2019) 1385–1405.
[1722] F. Pati, J. Gantelius, H.A. Svahn, Angew. Chemie Int. Ed. 55 (2016) 4650–4665.
[1723] A.K. Miri, A. Khalilpour, B. Cecen, S. Maharjan, S.R. Shin, A. Khademhosseini, Biomaterials 198 (2019) 204–216.
[1724] K. Fetah, P. Tebon, M.J. Goudie, J. Eichenbaum, L. Ren, N. Barros, R. Nasiri, S. Ahadian, N. Ashammakhi, M.R. Dokmeci, A. Khademhosseini, Progress in Biomedical Engineering 1 (2019) 012001.
[1725] S. Knowlton, B. Yenilmez, S. Tasoglu, Trends Biotechnol. 34 (2016) 685–688.
[1726] S. Mi, Z. Du, Y. Xu, W. Sun, J. Mater. Chem. B 6 (2018) 6191–6206.
[1727] D. Therriault, S.R. White, J.A. Lewis, Nat. Mater. 2 (2003) 265–271.
[1728] J.U. Lind, T.A. Busbee, A.D. Valentine, F.S. Pasqualini, H. Yuan, M. Yadid, S.-J. Park, A. Kotikian, A.P. Nesmith, P.H. Campbell, J.J. Vlassak, J.A. Lewis, K.K. Parker, Nat. Mater. 16 (2016) 303–308.
[1729] F. Adams, T. Qiu, A. Mark, B. Fritz, L. Kramer, D. Schlager, U. Wetterauer, A. Miernik, P. Fischer, Ann. Biomed. Eng. 45 (2017) 963–972.
[1730] S. Knowlton, C.H. Yu, F. Ersoy, S. Emadi, A. Khademhosseini, S. Tasoglu, Biofabrication 8 (2016) 025019.
[1731] J.O. Hardin, T.J. Ober, A.D. Valentine, J.A. Lewis, Adv. Mater. 27 (2015) 3279–3284.
[1732] H. Zhao, Y. Chen, L. Shao, M. Xie, J. Nie, J. Qiu, P. Zhao, H. Ramezani, J. Fu, H. Ouyang, Y. He, Small 14 (2018) 1802630.
[1733] C. Colosi, S.R. Shin, V. Manoharan, S. Massa, M. Costantini, A. Barbetta, M.R. Dokmeci, M. Dentini, A. Khademhosseini, Adv. Mater. 28 (2016) 677–684.
[1734] Q. Pi, S. Maharjan, X. Yan, X. Liu, B. Singh, A.M. van Genderen, F. Robledo-Padilla, R. Parra-Saldivar, N. Hu, W. Jia, C. Xu, J. Kang, S. Hassan, H. Cheng, X. Hou, A. Khademhosseini, Y.S. Zhang, Adv. Mater. 30 (2018) 1706913.
[1735] K.A. Homan, D.B. Kolesky, M.A. Skylar-Scott, J. Herrmann, H. Obuobi, A. Moisan, J.A. Lewis, Sci. Rep. 6 (2016) 34845.
[1736] H. Clevers, Cell 165 (2016) 1586–1597.
[1737] D.W. Hutmacher, Nat. Mater. 9 (2010) 90–93.
[1738] B. Zhang, A. Korolj, B.F.L. Lai, M. Radisic, Nat. Rev. Mater. 3 (2018) 257–278.
[1739] X. Ma, J. Liu, W. Zhu, M. Tang, N. Lawrence, C. Yu, M. Gou, S. Chen, Adv. Drug Deliv. Rev. 132 (2018) 235–251.
[1740] J. Vanderburgh, J.A. Sterling, S.A. Guelcher, Ann. Biomed. Eng. 45 (2017) 164–179.
[1741] Y. Lara, Z. Qiang, K. Shifeng, C. Chung-Wai James, C. Kyle Alexander, H. Yong, Y. Mo, Z. Xin, Biofabrication 11 (2019) 032003.
[1742] S. Eggert, D.W. Hutmacher, Biofabrication 11 (2019) 043002.
[1743] K.-H. Nam, A.S.T. Smith, S. Lone, S. Kwon, D.-H. Kim, J. Lab. Autom. 20 (2014) 201–215.
[1744] S. Knowlton, S. Tasoglu, Trends Biotechnol. 34 (2016) 681–682.
[1745] I.T. Ozbolat, W. Peng, V. Ozbolat, Drug Discov. Today 21 (2016) 1257–1271.
[1746] P. Beri, B.F. Matte, L. Fattet, D. Kim, J. Yang, A.J. Engler, Nat. Rev. Mater. 3 (2018) 418–430.
[1747] Y. Li, E. Kumacheva, Sci. Adv. 4 (2018) eaas8998.
[1748] S. Knowlton, A. Joshi, B. Yenilmez, I.T. Ozbolat, C.K. Chua, A. Khademhosseini, S. Tasoglu, Int. J. Bioprinting 2 (2016) 3–8.
[1749] J. Nie, Q. Gao, C. Xie, S. Lv, J. Qiu, Y. Liu, M. Guo, R. Guo, J. Fu, Y. He, Mater. Horiz. 7 (2020) 82–92.
[1750] X. Cao, R. Ashfaq, F. Cheng, S. Maharjan, J. Li, G. Ying, S. Hassan, H. Xiao, K. Yue, Y.S. Zhang, Adv. Funct. Mater. 29 (2019) 1807173.
[1751] H.-G. Yi, Y.H. Jeong, Y. Kim, Y.-J. Choi, H.E. Moon, S.H. Park, K.S. Kang, M. Bae, J. Jang, H. Youn, S.H. Paek, D.-W. Cho, Nat. Biomed. Eng. 3 (2019) 509–519.
[1752] T.Q. Huang, X. Qu, J. Liu, S. Chen, Biomed. Microdevices 16 (2014) 127–132.
[1753] F. Meng, C.M. Meyer, D. Joung, D.A. Vallera, M.C. McAlpine, A. Panoskaltsis-Mortari, Adv. Mater. 31 (2019) 1806899.
[1754] J. Diao, C. Zhang, D. Zhang, X. Wang, J. Zhang, C. Ma, K. Deng, T. Jiang, W. Jia, T. Xu, Biofabrication 11 (2019) 025006.
[1755] F. Rengier, A. Mehndiratta, H. von Tengg-Kobligk, C.M. Zechmann, R. Unterhinninghofen, H.U. Kauczor, F.L. Giesel, Int. J. Comput. Assist. Radiol. Surg. 5 (2010) 335–341.
[1756] H.H. Malik, A.R.J. Darwood, S. Shaunak, P. Kulatilake, A.A. El-Hilly, O. Mulki, A. Baskaradas, J. Surg. Res. 199 (2015) 512–522.
[1757] A. Hosny, J.D. Dilley, T. Kelil, M. Mathur, M.N. Dean, J.C. Weaver, B. Ripley, J. Cardiovasc. Comput. Tomogr. 13 (2019) 21–30.
[1758] D. Schmauss, C. Schmitz, A.K. Bigdeli, S. Weber, N. Gerber, A. Beiras-Fernandez, F. Schwarz, C. Becker, C. Kupatt, R. Sodian, Ann. Thorac. Surg. 93 (2012) e31–e33.
[1759] G.T. Klein, Y. Lu, M.Y. Wang, World Neurosurg. 80 (2013) 233–235.
[1760] P. Calvert, Adv. Mater. 21 (2009) 743–756.
[1761] X. Le, W. Lu, J. Zhang, T. Chen, Adv. Sci. 6 (2019) 1801584.
[1762] L. Ionov, Adv. Funct. Mater. 23 (2013) 4555–4570.
[1763] H. Yuk, S. Lin, C. Ma, M. Takaffoli, N.X. Fang, X. Zhao, Nat. Commun. 8 (2017) 14230.
[1764] R. Tognato, A.R. Armiento, V. Bonfrate, R. Levato, J. Malda, M. Alini, D. Eglin, G. Giancane, T. Serra, Adv. Funct. Mater. 29 (2019) 1804647.
[1765] X. Wang, X.-H. Qin, C. Hu, A. Terzopoulou, X.-Z. Chen, T.-Y. Huang, K. Maniura-Weber, S. Pané, B.J. Nelson, Adv. Funct. Mater. 28 (2018) 1804107.
[1766] C. Cvetkovic, R. Raman, V. Chan, B.J. Williams, M. Tolish, P. Bajaj, M.S. Sakar, H.H. Asada, M.T.A. Saif, R. Bashir, Proc. Natl. Acad. Sci. 111 (2014) 10125–10130.
[1767] Y. Morimoto, H. Onoe, S. Takeuchi, Sci. Robot. 3 (2018) eaat4440.

[1768] S.I. Rich, R.J. Wood, C. Majidi, Nat. Electron. 1 (2018) 102–112.
[1769] H. Yuk, B. Lu, X. Zhao, Chem. Soc. Rev. 48 (2019) 1642–1667.
[1770] C. Yang, Z. Suo, Nat. Rev. Mater. 3 (2018) 125–142.
[1771] S. Lin, H. Yuk, T. Zhang, G.A. Parada, H. Koo, C. Yu, X. Zhao, Adv. Mater. 28 (2016) 4497–4505.
[1772] M.A.M. Leenen, V. Arning, H. Thiem, J. Steiger, R. Anselmann, Phys. Status Solidi 206 (2009) 588–597.
[1773] A.D. Valentine, T.A. Busbee, J.W. Boley, J.R. Raney, A. Chortos, A. Kotikian, J.D. Berrigan, M.F. Durstock, J.A. Lewis, Adv. Mater. 29 (2017) 1703817.
[1774] H. Yang, W.R. Leow, X. Chen, Small Methods 2 (2018) 1700259.
[1775] M.A. Darabi, A. Khosrozadeh, R. Mbeleck, Y. Liu, Q. Chang, J. Jiang, J. Cai, Q. Wang, G. Luo, M. Xing, Adv. Mater. 29 (2017) 1700533.
[1776] K. Tian, J. Bae, S.E. Bakarich, C. Yang, R.D. Gately, G.M. Spinks, M. in het Panhuis, Z. Suo, J.J. Vlassak, Adv. Mater. 29 (2017) 1604827.
[1777] M.S. Mannoor, Z. Jiang, T. James, Y.L. Kong, K.A. Malatesta, W.O. Soboyejo, N. Verma, D.H. Gracias, M.C. McAlpine, Nano Lett. 13 (2013) 2634–2639.
[1778] M. Mehrali, S. Bagherifard, M. Akbari, A. Thakur, B. Mirani, M. Mehrali, M. Hasany, G. Orive, P. Das, J. Emneus, T.L. Andresen, A. Dolatshahi-Pirouz, Adv. Sci. 5 (2018) 1700931.

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