

## 3D bioprinting of natural materials and their AI-Enhanced printability: A review

Soumaya Grira<sup>a</sup>, Mohammad Sayem Mozumder<sup>a</sup>, Abdel-Hamid I. Mourad<sup>b,c,\*</sup>,  
Mohamad Ramadan<sup>d,e</sup>, Hadil Abu Khalifeh<sup>f</sup>, Mohammad Alkhedher<sup>g</sup>

<sup>a</sup> Department of Chemical & Petroleum Engineering, United Arab Emirates University, Al-Ain, 15551, United Arab Emirates

<sup>b</sup> Department of Mechanical and Aerospace Engineering, United Arab Emirates University, Al-Ain, 15551, United Arab Emirates

<sup>c</sup> National Water and Energy Center, United Arab Emirates University, Al-Ain, 15551, United Arab Emirates

<sup>d</sup> Lebanese International University, PO Box 146404, Beirut, Lebanon

<sup>e</sup> International University of Beirut, PO Box 146404, Beirut, Lebanon

<sup>f</sup> Chemical Engineering Department, Abu Dhabi University, 59911, Abu Dhabi, United Arab Emirates

<sup>g</sup> Mechanical and Industrial Engineering Department, Abu Dhabi University, 59911, Abu Dhabi, United Arab Emirates

### ARTICLE INFO

#### Keywords:

Bioprinting  
3D/4D printing  
Additive manufacturing  
Biofabrication  
Biomaterials  
Green bioinks  
Machine learning

### ABSTRACT

A wide array of biomaterials including proteins, polysaccharides, and other polymers are used to fabricate green 3D printing inks suitable for use in the fields of biotechnology, water treatment, food and agriculture, energy, and bioplastics. Although green materials are sustainable and have many favorable properties, each material has its own limitations. The main focus of this paper is to review the currently available 3D printing biomaterials that can be obtained from natural sources and investigate the artificial intelligence (AI) assisted approaches that can be used to enhance their printability and accelerate their development. Results reveal that above 20 natural materials can be used in 3D printing inks, and their properties vary widely making their development slow and case-dependent. To speed up this process, this study highlights the significance of AI-assisted enhancement techniques and proves that AI can be used in three main aspects of biomaterial development for 3D bioprinting; material property prediction, smart material selection, and printing parameter optimization. It discusses the AI-based systems that have the potential to accelerate the development of 3D printing biomaterials and concludes that these methods are indeed promising but should be further tested, verified, and compared before they are incorporated into the industry. The importance of this study lies in integrating advanced AI systems with sustainable natural materials to leverage their inherent properties and utilize them in various applications for a greener future.

### List of Abbreviations

| Abbreviation | Meaning                                                              | Abbreviation | Meaning                       |
|--------------|----------------------------------------------------------------------|--------------|-------------------------------|
| AI           | Artificial Intelligence                                              | HAMA         | Hyaluronic Acid Methacrylate  |
| AC           | Atelocollagen                                                        | HML          | Hierarchical Machine Learning |
| AI-HTPCSS    | Artificial Intelligence High-Throughput Computation Screening System | ILP          | Inductive logic programming   |
| AJP          | Aerosol Jet Printing                                                 | KNN          | K-Nearest Neighbors           |

(continued on next column)

### (continued)

| Abbreviation | Meaning                                    | Abbreviation | Meaning                                         |
|--------------|--------------------------------------------|--------------|-------------------------------------------------|
| ANN          | Artificial Neural Network                  | LASSO        | Least Absolute Shrinkage and Selection Operator |
| BO           | Bayesian Optimization                      | LGBM         | Light Gradient Boosting                         |
| BR           | Bayesian Regression                        | LR           | Linear Regression                               |
| CAD          | Computer-Aided Design                      | ML           | Machine Learning                                |
| CGAN         | Conditional Generative Adversarial Network | MLP          | Multi-Layer Perceptron                          |

(continued on next page)

\* Corresponding author. Department of Mechanical and Aerospace Engineering, United Arab Emirates University, Al-Ain, 15551, United Arab Emirates.  
E-mail address: [ahmourad@uaeu.ac.ae](mailto:ahmourad@uaeu.ac.ae) (A.-H.I. Mourad).

(continued)

| Abbreviation | Meaning                             | Abbreviation | Meaning                                  |
|--------------|-------------------------------------|--------------|------------------------------------------|
| CNN          | Convolutional Neural Network        | MLR          | Multiple Linear Regression               |
| dECM         | Decellularized Extracellular Matrix | OLS LR       | Ordinary Least Squares Linear Regression |
| DIW          | Direct Ink Writing                  | PBF          | Powder Bed Fusion                        |
| DL           | Deep Learning                       | PCL          | Polycaprolactone                         |
| DOD          | Drop-On-Demand Bioprinting          | PCR          | Principal Component Regression           |
| DQN          | Deep Q-Network                      | PDMS         | Polydimethylsiloxane                     |
| DT           | Decision Tree                       | PEG          | Polyethylene Glycol                      |
| FCNN         | Fully Connected Neural Network      | PHA          | Polyhydroxyalkanoates                    |
| FDM          | Fused Deposition Modeling           | PLA          | Polylactic Acid                          |
| FEM          | Finite Element Method               | PLSR         | Partial Least Squares Regression         |
| FFF          | Fused Filament Fabrication          | PPO          | Proximal Policy Optimization             |
| FRE          | Freeform Reversible Embedding       | RF           | Random Forest                            |
| GAN          | Generative Adversarial Network      | SVM          | Support Vector Machine                   |
| GBR          | Gradient Boosting Regression        | SVR          | Support Vector Regression                |
| GelMA        | Gelatin Methacrylate                | TPL          | Two-Photon Lithography                   |

(continued on next column)

(continued)

| Abbreviation | Meaning                     | Abbreviation   | Meaning                         |
|--------------|-----------------------------|----------------|---------------------------------|
| GPR          | Gaussian Process Regression | WAAM           | Wire-Arc Additive Manufacturing |
| GUI          | Graphical User Interface    | WOA            | Whale Optimization Algorithm    |
| HA           | Hyaluronic Acid             | XGBoost or XGB | Extreme Gradient Boosting       |

## 1. Introduction

The 3D printing and bioprinting industry has experienced remarkable growth over the past decade, transforming from a niche technology that is used in limited fields into a widely utilized and demanded tool across numerous industries. It finds its applications in the biomedical industry, water treatment industry, energy industry, and many more that are discussed in this review [1–4]. From medical implants and prosthetics to soilless agriculture and bio-based battery electrolytes, 3D printing is revolutionizing the way products are manufactured and designed. This rapid expansion has been driven by advancements in printing technologies and printable materials. In terms of materials, as the world becomes more conscious about sustainability and renewable resources, the 3D printing industry takes its share by moving towards

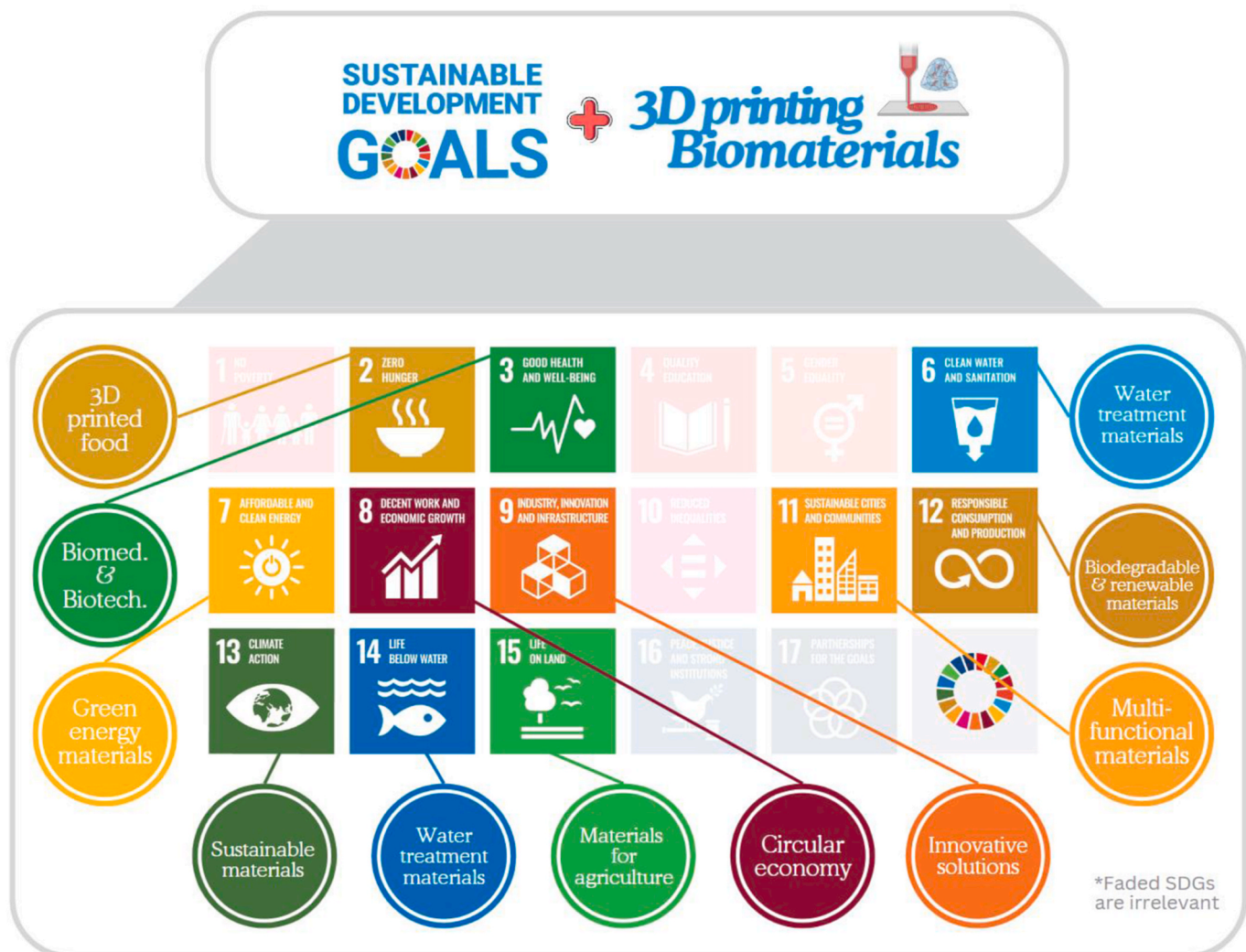


Fig. 1. Sustainable Development Goals (SDGs) that biomaterial 3D printing serves in many industries.

green materials or biomaterials that are naturally sourced (e.g., from land crops and marine sources) and preferably biodegradable. In terms of Sustainable Development Goals, the use of biomaterial 3D printing in multiple industries serves various goals, as shown in Fig. 1. To avoid confusion, the terms biomaterials and natural materials are used interchangeably to mean materials that are extracted from natural sources, which is the scope of this study. Moreover, the term bioink is used to mean a formulation that can be printed and is capable of hosting living cells, regardless of whether it is actually loaded with cells or not. Bioinks can be made of natural materials, synthetic materials, and various additives. This paper focuses only on natural materials (i.e., which can be used as base materials for bioinks and regular inks).

Biomaterials such as PLA (Polylactic acid) and natural hydrogels are being used and developed as base materials for sustainable bioinks to meet the demand of various industries that use 3D printing [1,5–7]. Although these biomaterials have various advantageous properties ranging from high biocompatibility to tunable flexibility and mechanical stability, most of them are still in their developmental stage. Generally, each material either fails on the aspect of printability, mechanical stability, or biological compatibility. On one hand, many protein-based biomaterials have attractive biological properties but lag on the mechanical stability side. Conversely, some types of polysaccharides and other polymers are mechanically and thermally stable but are not ideally biocompatible. Similarly, many hydrogels have favorable electrochemical properties and/or adsorption characteristics that are attractive as sustainable alternatives in industries such as the

energy and water treatment industries but are limited by their stability and printability. To enhance these materials, approaches such as hybrid composites, material modification and functionalization, and structure and printing parameter optimization are frequently used. A large part of these developmental approaches requires extensive amounts of experimental trial-and-error in which resources get wasted. Additionally, the progress in this field needs to be fast in order to meet the growing demand for green bioinks in various industries.

In this context, artificial intelligence (AI) has emerged as a powerful tool with the potential to address these biomaterial development challenges. Since machine learning approaches have the ability to deal with huge amounts of data and learn from it in a small amount of time, they can be used to accelerate the discovery of new biomaterials, predict their properties, and optimize their printability. Various machine learning and deep learning approaches are being explored and developed for this purpose.

Recently, as artificial intelligence is rapidly advancing in various fields, its application in different 3D printing techniques is being studied [8,9]. There are reviews in the literature that discuss the role of artificial intelligence and machine learning in advancing multiple aspects of 3D printing, such as monitoring, defect detection, parameter control, etc. [10–16]. However, original studies exploring the use of machine learning algorithms to specifically develop natural biomaterials for 3D printing and bioprinting remain scattered in the literature. Fig. 2 shows the exponential increase in publications on the use of AI in 3D printing in the last few years. It appears that the same trend (in smaller numbers) is

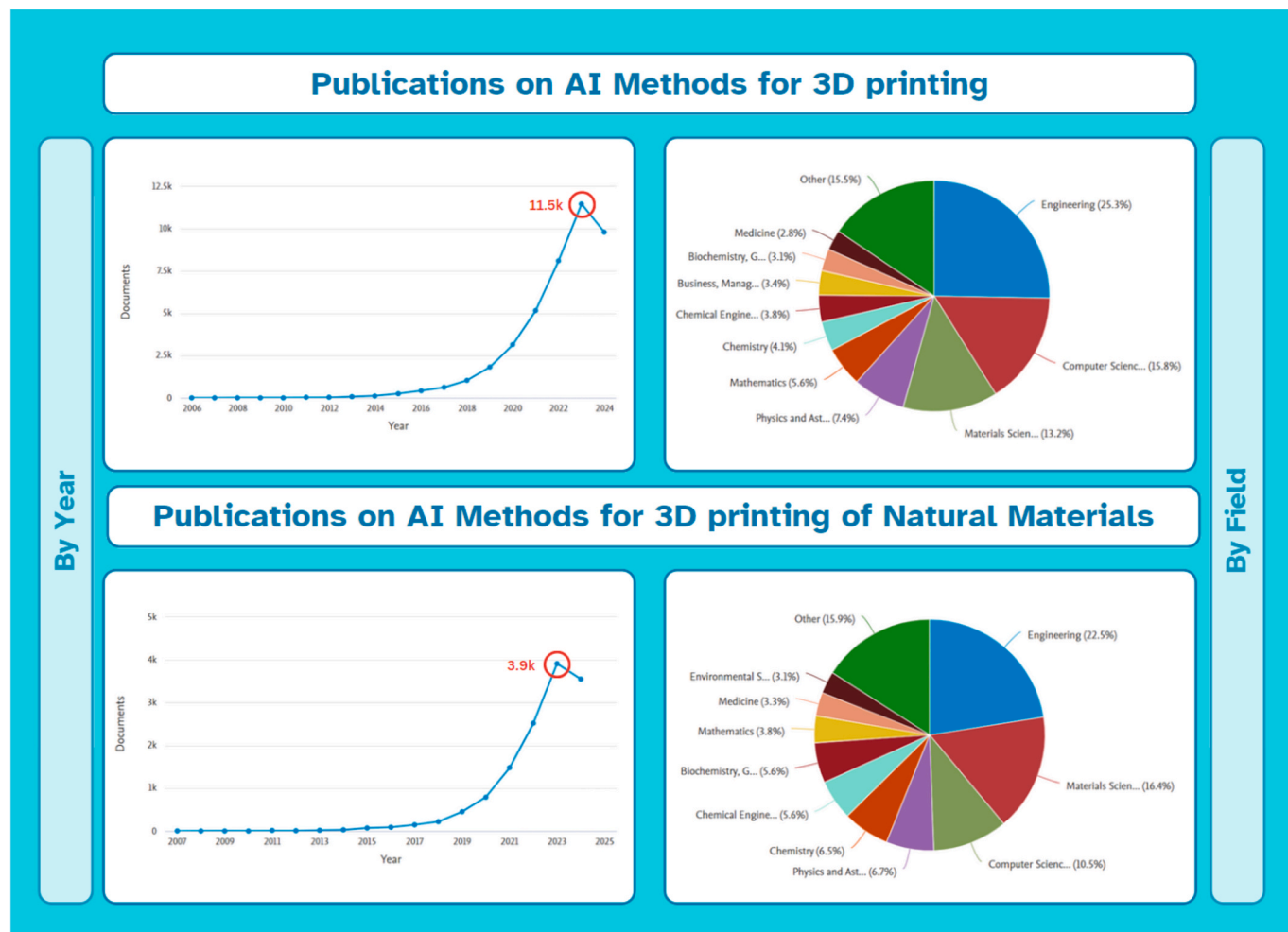


Fig. 2. The number of papers on AI methods for 3D printing in general and specifically using natural materials between 2006 and August 2024 obtained from the Scopus database.

followed when the search is limited to naturally sourced materials only. Therefore, it is crucial to pay more attention to this field to advance this type of materials in order to meet the surging demand.

This review aims to serve as a guide for natural biomaterial development using artificial intelligence for the purpose of sustainable 3D printing ink fabrication. First, it discusses and analyzes the biomaterials that have the potential to be used in this field, their properties, and applications. Then, it explains how artificial intelligence can be used to advance the printability and other properties of these materials (i.e., property prediction, smart material selection, and printing parameter optimization) according to the available literature and further interpretations. We expect this review to draw the focus of material scientists and engineers to invest in AI-assisted solutions, and attract the attention of research from various industries to advance the green solutions presented in this study.

## 2. Biomaterial-based inks for various applications

### 2.1. Biomaterials for biotechnological applications

Natural materials have various favorable properties that make them attractive for numerous applications, such as biomedical/biotechnological, water treatment, food, energy, bioplastics, and agricultural applications. Generally, applications in each field require a certain set of properties. For example, in the rapidly evolving field of biomedicine, there is an increasing demand for materials that are biocompatible with human tissues and cells. Research in this field is continuously looking for biomaterials that can be used to make scaffolds that support cell growth and maintain cell proliferation. Additionally, specific biomedical applications require particular characteristics. Fig. 3 shows the general favorable properties for biomaterials to be used in biomedical applications. For instance, tissue engineering and wound healing applications require controlled degradation and self-healing properties, drug delivery requires porosity and responsiveness to external stimuli, cosmetics and pharmaceuticals require antioxidation, anti-inflammatory, anti-aging, and other bioactive properties. There is a wide range of biomaterials that possess these properties, each in a unique combination. Among these

biomaterials, the ones suitable for making bioprinting inks are discussed in this section.

#### 2.1.1. Protein-based materials

Protein-based materials from natural sources are attractive for biomedical applications due to their minimal cytotoxicity and inherent biocompatibility with human cells. In bioprinting, fibrous proteins, globular proteins, and other protein complexes can be used to make bioinks. Fibrous proteins which are long chains of water-insoluble proteins (e.g., collagen, keratin, silk, and elastin) provide structural integrity and flexibility for the scaffold while other protein complexes support cell adhesion and proliferation. Although protein-based bioinks are highly suitable for biomedical applications, they face numerous challenges in 3D bioprinting, among which the most important limitation is their weak mechanical stability, as noticed in Table 1.

Collagen and gelatin are both protein biomaterials widely used in bioprinting due to their favorable biological properties. Collagen, abundant and supportive of cell function, faces challenges in bioprinting due to poor mechanical stability and slow gelation [23,37]. To address these, various approaches such as ink modification, sacrificial support, and hybrid materials have been explored. Modifying collagen concentration or crosslinking can improve its mechanical properties, though increasing concentration may reduce bioactivity [38–40]. Blending with materials like alginate [41] and hyaluronic acid [42] enhances stability, and techniques like hierarchical porosity [42], double crosslinking [42], and mineralization [43] further improve its suitability for bioprinting.

Gelatin, derived from collagen, shares similar biological properties but offers better mechanical stability. It is also temperature-sensitive, de-crosslinking and melting at 30–35 °C [6,24]. However, gelatin's poor rheological properties present challenges in bioprinting. To overcome this, gelatin is typically used in modified forms, such as GelMA (gelatin methacrylate), or combined with other materials like alginate [44–46], hyaluronic acid [46], and additives such as decellularized bone particles [47] and ferulic acid [48]. Efforts continue to develop gelatin-based ink formulations with tailored properties for specific biomedical applications, leveraging its biocompatibility and biodegradability.

## Properties required by biomaterials for 3D bioprinting in biomedical applications

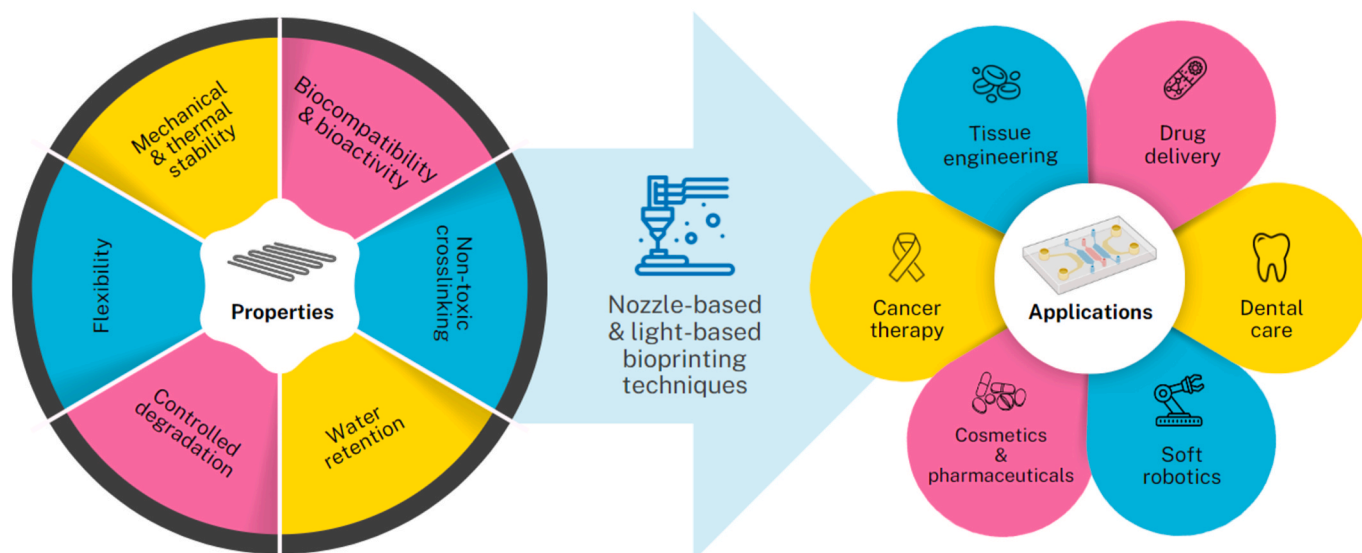
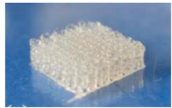
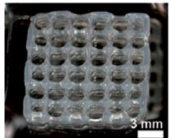
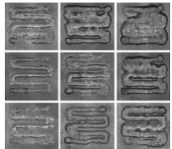
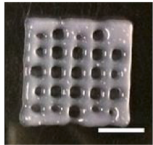
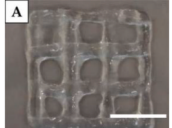
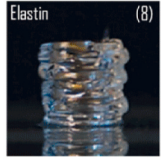


Fig. 3. Properties required by biomaterials for 3D bioprinting in biomedical applications.



**Table 1**  
Properties and printability of protein-based materials for biomedical applications.

| Material | Type                                     | Biological properties                                                                                                                       | Physical properties                                                                                                                                                                                         | Printability                                                                                | Visual examples                                                                       | Ref.          |
|----------|------------------------------------------|---------------------------------------------------------------------------------------------------------------------------------------------|-------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|---------------------------------------------------------------------------------------------|---------------------------------------------------------------------------------------|---------------|
| Collagen | Structural/fibrous protein               | <ul style="list-style-type: none"> <li>- Supports cell functions</li> <li>- High cell adhesion</li> </ul>                                   | <ul style="list-style-type: none"> <li>- Poor mechanical stability</li> <li>- Slow gelation</li> <li>- Not shear thinning</li> <li>- Thermal and enzymatic crosslinking</li> </ul>                          | Printable using extrusion, inkjet, and laser-assisted bioprinting techniques                |    | [18–21]       |
| Gelatin  | Protein                                  | <ul style="list-style-type: none"> <li>- Good biocompatibility</li> <li>- Medium cell adhesion</li> <li>- Good biodegradability</li> </ul>  | <ul style="list-style-type: none"> <li>- Thermal, chemical, and enzymatic crosslinking</li> <li>- Temperature responsive</li> <li>- Shear thinning</li> <li>- Poor rheological properties</li> </ul>        | Printable using nozzle-based and light-assisted techniques                                  |    | [6,20,23,24]  |
| Silk     | Fibrous protein                          | <ul style="list-style-type: none"> <li>- Low cell adhesion</li> <li>- High enzymatic degradation rate</li> </ul>                            | <ul style="list-style-type: none"> <li>- Shear thinning (spider silk)</li> <li>- Self-crosslinking</li> <li>- Good structural integrity</li> </ul>                                                          | Printable using nozzle-based and light-assisted techniques                                  |    | [18,20,23,26] |
| Fibrin   | Fibrous protein                          | <ul style="list-style-type: none"> <li>- High biocompatibility</li> <li>- High cell adhesion</li> <li>- High biodegradability</li> </ul>    | <ul style="list-style-type: none"> <li>- Thermal, chemical, and enzymatic crosslinking</li> <li>- Elastic and viscous</li> <li>- Not shear thinning</li> <li>- Has tunable mechanical properties</li> </ul> | Printable mainly using nozzle-based techniques                                              |    | [20,28]       |
| dECM     | Mixture of proteins and other components | <ul style="list-style-type: none"> <li>- Excellent biocompatibility</li> <li>- Source of bioactive molecules</li> </ul>                     | <ul style="list-style-type: none"> <li>- Water soluble</li> <li>- Weak mechanical properties</li> <li>- Chemical and physical crosslinking</li> </ul>                                                       | Printable using nozzle-based and light-assisted techniques                                  |   | [6,30,31]     |
| Matrigel | Protein-based biomaterial                | <ul style="list-style-type: none"> <li>- High cell adhesion</li> <li>- mimics human extracellular matrix</li> </ul>                         | <ul style="list-style-type: none"> <li>- Shear thinning</li> <li>- Thermal crosslinking</li> <li>- poor mechanical strength</li> </ul>                                                                      | Mainly printable using nozzle-based techniques. Can also be used in laser-assisted methods. |  | [7,20,33]     |
| Keratin  | Fibrous protein                          | <ul style="list-style-type: none"> <li>- High biocompatibility</li> <li>- Biodegradable</li> </ul>                                          | <ul style="list-style-type: none"> <li>- Weak mechanical properties</li> </ul>                                                                                                                              | Mainly printed using light-base techniques. Can also be printed using nozzle-based methods  | –                                                                                     | [23,34,35]    |
| Elastin  | Fibrous protein                          | <ul style="list-style-type: none"> <li>- Essential component of the ECM</li> <li>- Biodegradable</li> <li>- High cell attachment</li> </ul> | <ul style="list-style-type: none"> <li>- High elasticity, stretchability, and flexibility</li> </ul>                                                                                                        | Mainly printable using nozzle-based techniques.                                             |  | [18]          |

Other fibrous proteins, including silk, fibrin, and elastin, are also used in bioinks. Silk offers good mechanical stability but has poor rheology, enzymatic degradation, and potential crosslinking toxicity [49–51]. Innovations like light-assisted bioprinting [52], hybrid bioinks [53,54], and computational modeling [51] address these issues, though challenges remain. Fibrin is elastic and biocompatible but has viscosity challenges and high degradation rates [6,7,19]. Solutions include blending with glycerol [27], gelatin [55,56], collagen [56], hyaluronic acid [27,56], alginate [57], and chitosan [57]. Elastin promotes flexibility and cell adhesion [23,58], but its natural extraction is difficult, leading to the use of synthetic alternatives [59] or blends with materials like resilin [60] and gelatin [61]. Keratin supports cell growth but requires modifications such as water solubility, photocrosslinking [23,62], and nanoparticle coatings [63].

Other protein complexes like dECM and Matrigel are also utilized. dECM, obtained after decellularization, is tissue-specific and highly biocompatible but faces challenges in reproducibility, decellularization, and weak mechanical stability [6,19,31,64]. Matrigel, extracted from mice tumors, mimics human ECM and has gelling properties but struggles with reproducibility, low mechanical stability, and temperature sensitivity [7].

#### 2.1.2. Polysaccharide-based materials

Polysaccharides offer superior mechanical stability compared to protein-based materials, making them highly attractive for use as bioink base materials. These materials are abundant and derived from various natural sources, such as cellulose, pectin, and starches from plants, and alginate, carrageenan, and chitosan from marine sources as shown in

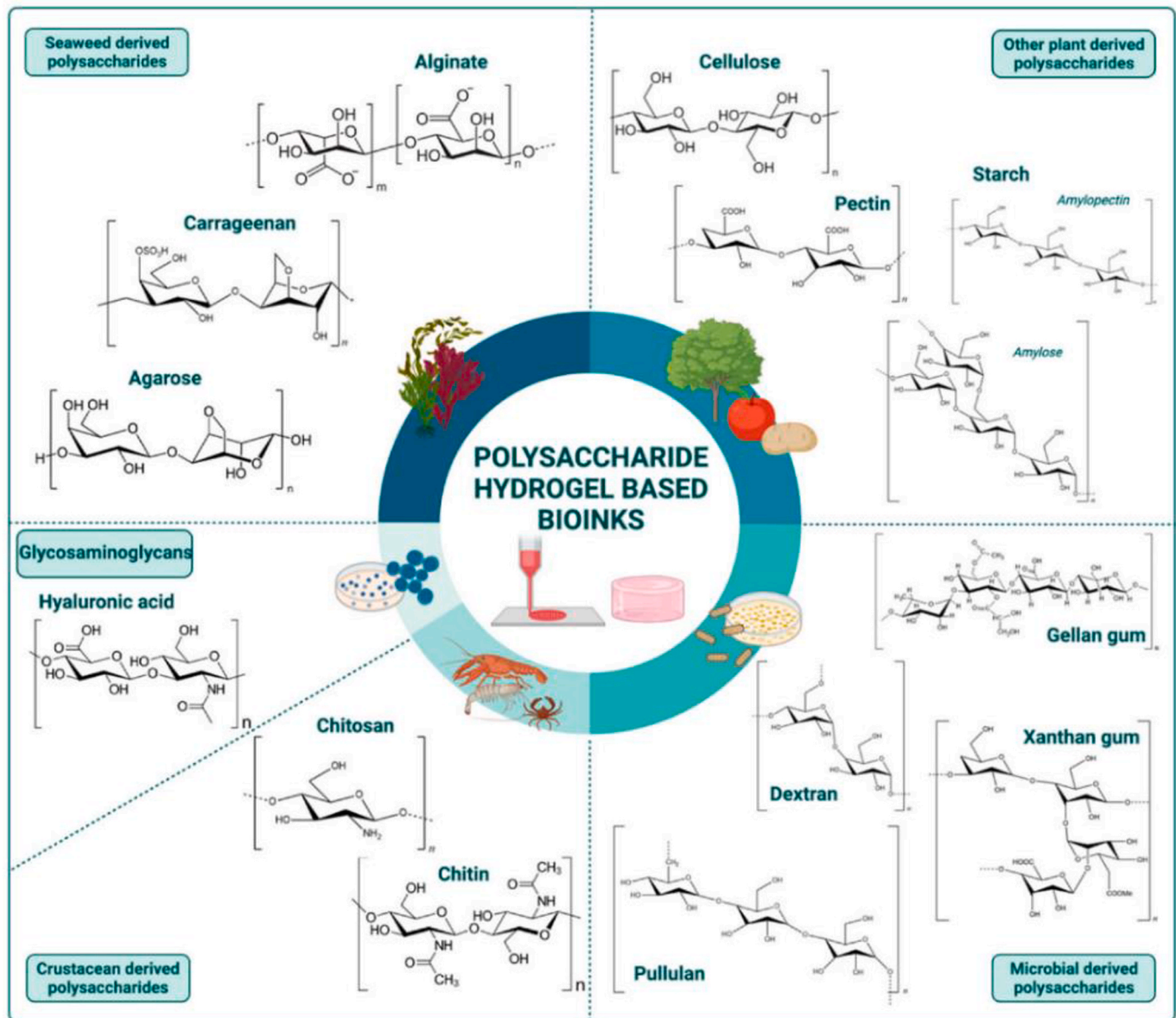


Fig. 4. Chemical structures of polysaccharides from various natural sources [3].

Fig. 4. Their properties are influenced by factors such as chemical structure, hydrogel concentration, and gelation mechanisms. Additionally, polysaccharides do not carry the risk of transmissible diseases that some protein-based materials do [65]. However, one challenge is their relatively lower cell adhesion, which affects their biocompatibility compared to protein-based hydrogels.

A commonly used polysaccharide in biomedical applications is hyaluronic acid (HA), which is known for its high biocompatibility and favorable rheological properties. However, HA's mechanical stability is often insufficient, which requires modifications such as methacrylate or cysteine to improve its gelation rates and mechanical properties [6,19,66]. Another polysaccharide, dextran, is used in biomedical fields due to its biocompatibility and biodegradability. However, its limited cell adhesion reduces its effectiveness in tissue engineering applications [67–70].

Among the polysaccharides sourced from algae, alginate and carrageenan are the most widely used in biomedical applications. Alginate's favorable biological properties support cell encapsulation, but it faces issues such as low-resolution printing and brittleness due to its low viscosity in its native state [5,71]. These challenges can be overcome by

creating hybrid hydrogels [72–74], or modifying the alginate structure [75,76]. Carrageenan, while also beneficial for its bioactive properties, shares similar limitations in terms of brittleness, but can be enhanced by incorporating nanostructures or using alternating-layer bioprinting techniques [3,77,78]. Fucoidan, another polysaccharide from algae, has bioactive properties but limited experimental evidence for its printability [4].

Chitosan, distinct due to its cationic nature, is often used in combination with anionic polysaccharides to improve bioink properties. Its biocompatibility, biodegradability, and easy gelation properties make it suitable for various bioprinting techniques, including nozzle-based and light-assisted methods [79–82]. Agarose, while mechanically robust and stable, has poor cell adhesion and limited degradation [5,83–85]. It is commonly used as a sacrificial support material for weak biomedical scaffolds [86,87].

Cellulose, another versatile polysaccharide, is commonly used in modified forms such as methylcellulose and carboxymethylcellulose. These derivatives offer favorable viscosity, thixotropy, and shear thinning behavior, making them useful in bioink formulations. Additionally, cellulose-based bioinks are biodegradable, but challenges such as

insolubility and low shape fidelity at low concentrations remain [88–91]. Other polysaccharides, such as starch and pectin, are derived from food sources. While starch can form biodegradable gels, it requires heating and cooling, which limits its applicability in cell encapsulation [3]. Pectin, on the other hand, is often used as an additive to improve the biological properties of bioinks without compromising their mechanical integrity [92]. Lastly, natural gums, including guar gum and xanthan gum, are used to enhance the rheological properties of bioinks [3]. Table 2 summarizes the properties and printability of polysaccharide-based bioinks for biomedical applications.

### 2.1.3. Other materials

In addition to proteins and polysaccharides, there are a few other types of materials that are naturally sourced and used in biomedical applications mainly due to their strong mechanical properties. PLA (Polylactic acid) and PHAs (Polyhydroxyalkanoates) are two biodegradable nontoxic polyesters that have excellent printability and mechanical/thermal stability. They are used to make high shape fidelity biomedical scaffolds for tissue regeneration [154]. Although PLA is widely used in conventional 3D printing, its crystallinity and low cell adhesion limits its use in biomedical applications. To enhance the biological behavior of the PLA-based bioinks, studies are investigating the use of PLA nanofibers [155], PLA hybrid bioinks [155,156], and PLA nanocomposites [157]. Similarly, PHAs exhibit better biological properties when blended with other materials than when used independently [158]. PHAs also have tunable biodegradability that can vary from weeks to months, making them suitable for biomedical scaffold fabrication. Additionally, they can be used to develop smart materials that sense external stimuli and respond as programmed [158]. Interestingly, diatom-sourced biosilica, which is a ceramic material, or a mineral is also used for the fabrication of bioinks for biomedical applications. In terms of mechanical properties, biosilica is crystalline, highly stable, and resistance to moisture and chemicals, however it lags in biological properties. Therefore, it is used as fillers and nanocomposites in combination with other materials to leverage its benefits without compromising the overall bioink bioactivity [159–161]. Table 3 summarizes the properties and printability of PLA, PHAs, and biosilica.

## 2.2. Biomaterials for water treatment applications

Besides biotechnological applications that were discussed in the previous section, 3D printing using natural materials is becoming more popular in other fields such as the water treatment industry due to the demand for intricate structures with high surface area, controlled porosity, and advanced properties. Materials such as cellulose, PLA, biosilica, alginate, and chitosan are used to fabricate various designs including membranes, meshes, filters, among other structures that are used for oil/water separation, metal and microplastic removal, and water desalination.

Cellulose and its derivatives are commonly used to make functionalized filter papers, membranes, and fibers that are used for water treatment. 3D printing is gradually replacing the conventional methods of preparing cellulose-based separation membranes such as the solution casting method and replacing them with more advanced structures such as foams, aerogels, and thin meshes. Fig. 5 illustrates the process of preparing a cellulose-based ink and 3D printing it into filtration meshes. The results by Koh et al. [176] show that the microporous hydrophilic thin filters are able to separate oil and water with an efficiency of ~95 %, with the best results when the pores are smaller than 280  $\mu\text{m}$ . Additionally, they were able to achieve an efficiency of 99 % with a more complex structure, emphasizing the importance of 3D printing in enhancing the efficiency of oil/water separation. Similarly, Li et al. [177] achieved an oil/water separation efficiency of up to 99 % by using a 3D printed anti-fouling membrane made of cellulose acetate, polyvinyl alcohol, and silica nanoparticles. Interestingly, besides hydrophilic membranes, hydrophobic membranes are also used to treat water. For

instance, Ning et al. [178] used a membrane made of cellulose and silica to achieve an oil/water separation efficiency of 99.31 % after cycles and Xu et al. [179] 3D printed a cellulose-based hydrophobic foam that has the ability to remove microplastics. In another study [178], a cellulose-based aerogel was developed in way that it can switch between hydrophobicity for oil/water separation (efficiency: ~99.5 %) and hydrophilicity for water desalination using evaporation (efficiency: ~89.7 %). 3D printed cellulose-based membranes are also used for metal ions, dye, and CO<sub>2</sub> treatment [180]. However, further investigations and evaluations should be done to prove the efficiency of these cellulose-based materials under different scenarios and on larger scales.

Additionally, 3D printing has the potential to replace mesh coating in the water treatment industry. Fig. 6 shows the difference in uniformity and homogeneity between 3D printed meshes and coated meshes, which enhances the performance and durability of the 3D printed counterparts. Due to its stability and other favorable properties, PLA is commonly used to 3D print matrices and scaffolds that can be further surface-modified [181,182] and reinforced [183] to achieve a final composite that has good adsorption and water treatment properties.

Moreover, chitosan- and alginate-based materials have the potential to adsorb heavy metals such as copper and lead. In one study [185] a chitosan-based filter was 3D printed and used to adsorb lead, while in another study [186] chitosan was grafted on a scaffold made of a different material to enhance its removal of dye from textile wastewater. Also, chitosan catalysts incorporating metal oxides and other nanoparticles are reported for use in wastewater treatment [187]. However, the involvement of 3D printing is yet new to this field and expected to advance this field rapidly, especially on the nanoscale. Similarly, 3D printed alginate-based materials are also used to adsorb and remove water pollutants such as uranium [188], copper [189], zinc [189], Methylene Blue [190], and others [189,191].

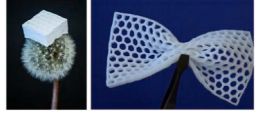
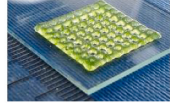
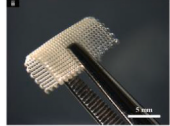
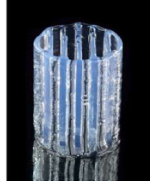
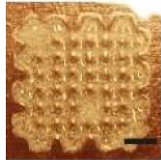
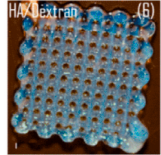
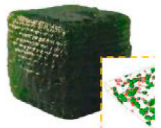
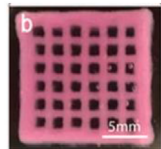
Natural hydrogels such as alginate, chitosan, and silica gel are also used to host and immobilize living cells such as algae that have the ability to treat water by recovering nutrients [192], removing heavy metals [182,188,193], detecting pollutants [194], and monitoring toxicity [195,196]. In this regard, 3D printing can either be used to print a hydrogel that is already loaded with cells or can be used to seed the cells into the hydrogel after printing the scaffold [4].

## 2.3. Biomaterials for food and agriculture applications

Aligning with Goal 2 of the United Nation's Sustainable Development Goals "End hunger, achieve food security and improved nutrition and promote sustainable agriculture", the food industry is increasingly focusing on exploring and implementing innovative ways to ensure sustainable food supply that meets the growing demand of the nations and the changing challenges of the new generations. Among these challenges in the food industry are the spread of highly selective diets, the increasing demand for functional foods, and the limited availability of certain food types. To address these issues, the incorporation of 3D printing has shown the potential to provide customizable cost-effective solutions using extrusion, inkjet, binder jetting, and light-assisted 3D printing [197]. In terms of materials, studies are continuously developing new 3D printing formulations made of edible inks including natural biomaterials such as alginate [198–200], carrageenan [201–204], chitosan [205], fish gelatin [206,207], pectin [208], starches [209], natural gums [153,210,211], milk protein [212], and lipids [213]. These formulations are used for many purposes such as high-nutrition functional foods, personalized therapeutic foods, and vegan alternative foods.

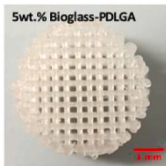
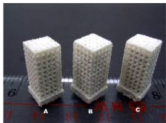
One of the main drivers for food 3D printing using edible biomaterials is the need for precisely prepared foods for specific diets and lifestyles. Recently, there have been many trials to 3D print vegan alternatives with similar sensory attributes and nutritional value to food types of non-vegan origin. Fig. 7 shows examples of seafood-like prototypes made of sunflower oil, carnauba wax, soy protein isolate and

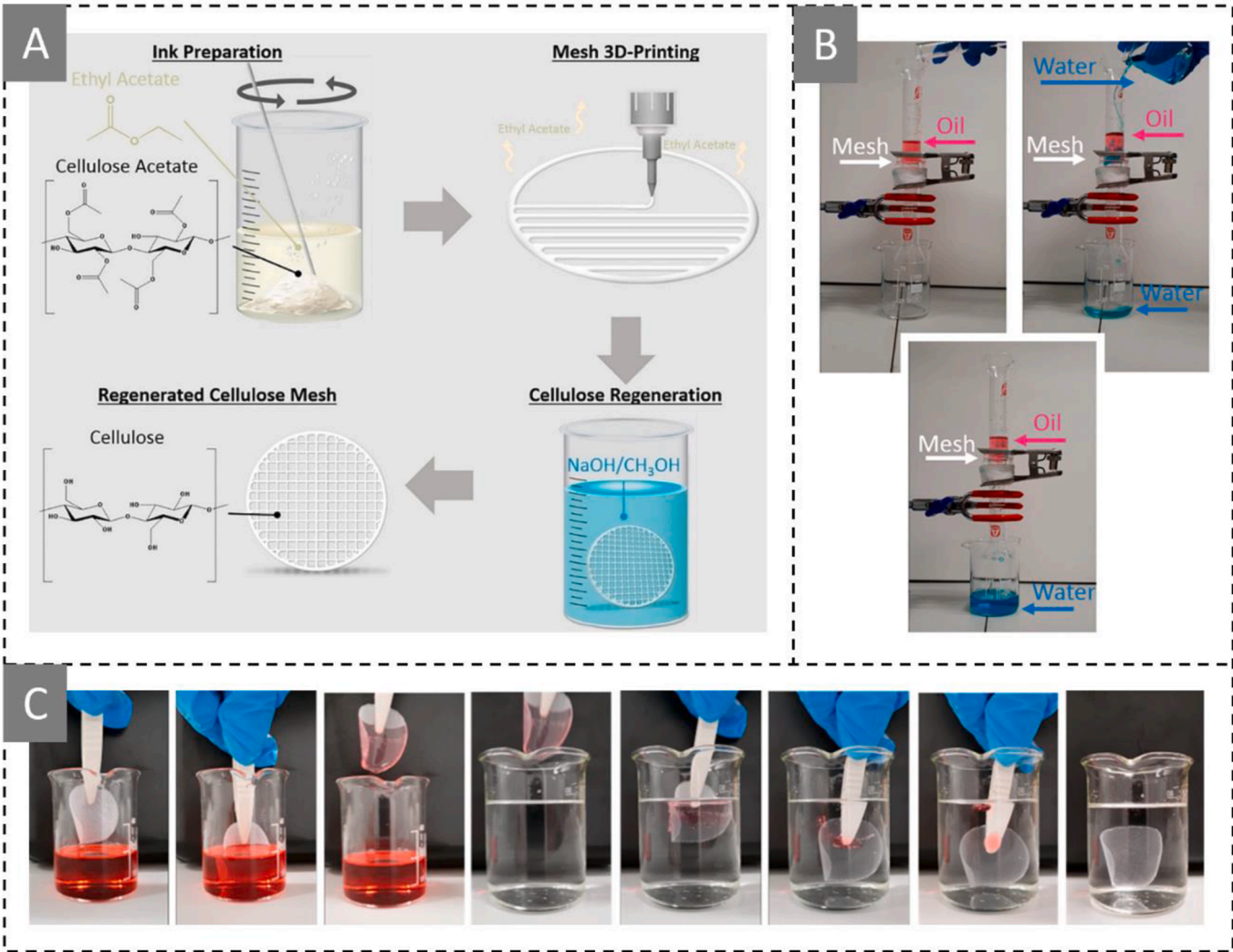
**Table 2**  
Properties and printability of polysaccharide-based materials for biomedical applications.

| Material        | Biological properties                                                                                                                                                     | Physical properties                                                                                                                                                                                  | Printability                                                                                                                          | Visual examples                                                                                | Refs.               |
|-----------------|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------|------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|---------------------------------------------------------------------------------------------------------------------------------------|------------------------------------------------------------------------------------------------|---------------------|
| Cellulose       | Tunable biodegradability                                                                                                                                                  | Properties vary depending on composition, can be: Crystalline, thixotropic, shear thinning, thermally stable.                                                                                        | Printable using extrusion [93–95], inkjet [96], and laser-assisted [97,98] techniques.                                                | <br>[99]    | [100,101]           |
| Alginate        | <ul style="list-style-type: none"> <li>- Biocompatible</li> <li>- Low cell adhesion</li> <li>- Nontoxic</li> <li>- biodegradable</li> </ul>                               | <ul style="list-style-type: none"> <li>- Chemical and ionic crosslinking</li> <li>- Shear thinning</li> </ul>                                                                                        | Printable using extrusion [102–104] and laser assisted bioprinting [105, 106]                                                         | <br>[107]   | [20]                |
| Chitosan        | <ul style="list-style-type: none"> <li>- Biocompatible</li> <li>- Biodegradable</li> <li>- Bioactive properties</li> <li>- Low cell adhesion</li> </ul>                   | <ul style="list-style-type: none"> <li>- Ionic crosslinking</li> <li>- Good mechanical stability</li> </ul>                                                                                          | Printable using various printing techniques (e.g., extrusion [108–111], inkjet [112], binder jetting [113], laser-assisted [114–117]) | <br>[118]   | [20, 119–122]       |
| Agarose         | <ul style="list-style-type: none"> <li>- Limited biocompatibility</li> <li>- Uncontrolled biodegradation</li> <li>- Low cell adhesion</li> <li>- pH-responsive</li> </ul> | <ul style="list-style-type: none"> <li>- Shear thinning</li> <li>- Thermal crosslinking</li> <li>- Thermoreversible hydrogel</li> </ul>                                                              | Printable using extrusion techniques [123–125]                                                                                        | <br>[126]   | [20,84,85, 127,128] |
| Carrageenan     | <ul style="list-style-type: none"> <li>- Biocompatible</li> <li>- High cell viability</li> <li>- Tunable biodegradability</li> </ul>                                      | <ul style="list-style-type: none"> <li>- High viscosity</li> <li>- High shape fidelity</li> <li>- Shear thinning</li> <li>- Thermal and ionic crosslinking</li> </ul>                                | Printable using nozzle-based [129, 130] and laser assisted [1] techniques                                                             | <br>[131]  | [20, 132–135]       |
| Hyaluronic acid | <ul style="list-style-type: none"> <li>- Essential in connective, epithelial, and neural tissues</li> <li>- Tunable properties</li> </ul>                                 | <ul style="list-style-type: none"> <li>- Poor mechanical properties</li> <li>- Rapid degradation</li> <li>- Physical and chemical crosslinking</li> <li>- Viscoelasticity is pH sensitive</li> </ul> | Printable using nozzle-based techniques [136]. Can be modified to suit laser-assisted methods [137].                                  | <br>[138] | [6,19]              |
| Dextran         | <ul style="list-style-type: none"> <li>- Biocompatible</li> <li>- Biodegradable</li> </ul>                                                                                | <ul style="list-style-type: none"> <li>- Should be modified to form a hydrogel</li> </ul>                                                                                                            | Printable using extrusion techniques [139]                                                                                            | <br>[140] | [3,141]             |
| Fucoidan        | <ul style="list-style-type: none"> <li>- Biocompatible</li> <li>- Bioactive properties</li> </ul>                                                                         | <ul style="list-style-type: none"> <li>- Shear thinning</li> <li>- Viscoelastic</li> </ul>                                                                                                           | Limited literature available                                                                                                          | –                                                                                              | [142–145]           |
| Pectin          | <ul style="list-style-type: none"> <li>- Contains bioactive molecules</li> </ul>                                                                                          | <ul style="list-style-type: none"> <li>- Varying degrees of gelling</li> <li>- Low mechanical strength</li> <li>- Rapid degradation</li> </ul>                                                       | Printable mainly through extrusion methods [146]                                                                                      | <br>[147] | [146,148]           |
| Starches        | <ul style="list-style-type: none"> <li>- Biodegradable</li> <li>- Nontoxic</li> <li>- Hydrophilic</li> </ul>                                                              | <ul style="list-style-type: none"> <li>- Low mechanical stability</li> <li>- Temperature sensitivity</li> <li>- Humidity sensitivity</li> <li>- viscoelasticity</li> </ul>                           | Printable mainly through extrusion printing [149]                                                                                     | <br>[150] | [149]               |
| Natural gums    | <ul style="list-style-type: none"> <li>- Biodegradable</li> <li>- Biocompatible</li> </ul>                                                                                | Water soluble, salt tolerant, gelling and thickening (i.e., guar gum, locust bean gum, xanthan gum)                                                                                                  | Useful as viscosity modifiers in nozzle-based printing                                                                                | –                                                                                              | [151–153]           |



**Table 3**  
Properties and printability of biosilica, PLA, and PHAs for biomedical applications.

| Material  | Type                        | Biological properties                                                                                                     | Physical properties                                                                                                                                                                   | Printability                                                                   | Visual examples                                                                              | Refs.     |
|-----------|-----------------------------|---------------------------------------------------------------------------------------------------------------------------|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|--------------------------------------------------------------------------------|----------------------------------------------------------------------------------------------|-----------|
| Biosilica | Ceramic material or mineral | <ul style="list-style-type: none"><li>- Commonly used as fillers and nanocomposites due to limited bioactivity</li></ul>  | <ul style="list-style-type: none"><li>- Crystalline</li><li>- High thermal stability</li><li>- Excellent mechanical strength</li><li>- Resistance to moisture and chemicals</li></ul> | Printable using various methods depending on the base components of the bioink | <br>[162] | [163]     |
| PLA       | Linear aliphatic polyester  | <ul style="list-style-type: none"><li>- Varying biodegradability</li><li>- Nontoxic</li><li>- Low cell affinity</li></ul> | <ul style="list-style-type: none"><li>- High rigidity</li><li>- Shape fidelity</li><li>- Crystalline and brittle</li></ul>                                                            | Widely used in 3D printing                                                     | <br>[164] | [165]     |
| PHAs      | Polyester                   | <ul style="list-style-type: none"><li>- Biodegradable</li><li>- Biocompatible</li></ul>                                   | <ul style="list-style-type: none"><li>- Semi-crystalline</li><li>- Tensile strength between 9 and MPa [166]</li></ul>                                                                 | Printable using extrusion [167–169] and laser-assisted [170,171] techniques    | <br>[172] | [173–175] |



**Fig. 5.** (A) Fabrication of a cellulose mesh via 3D printing for water treatment, and (B) the setup of the oil/water separation process, (C) self-cleaning ability of the mesh [176].

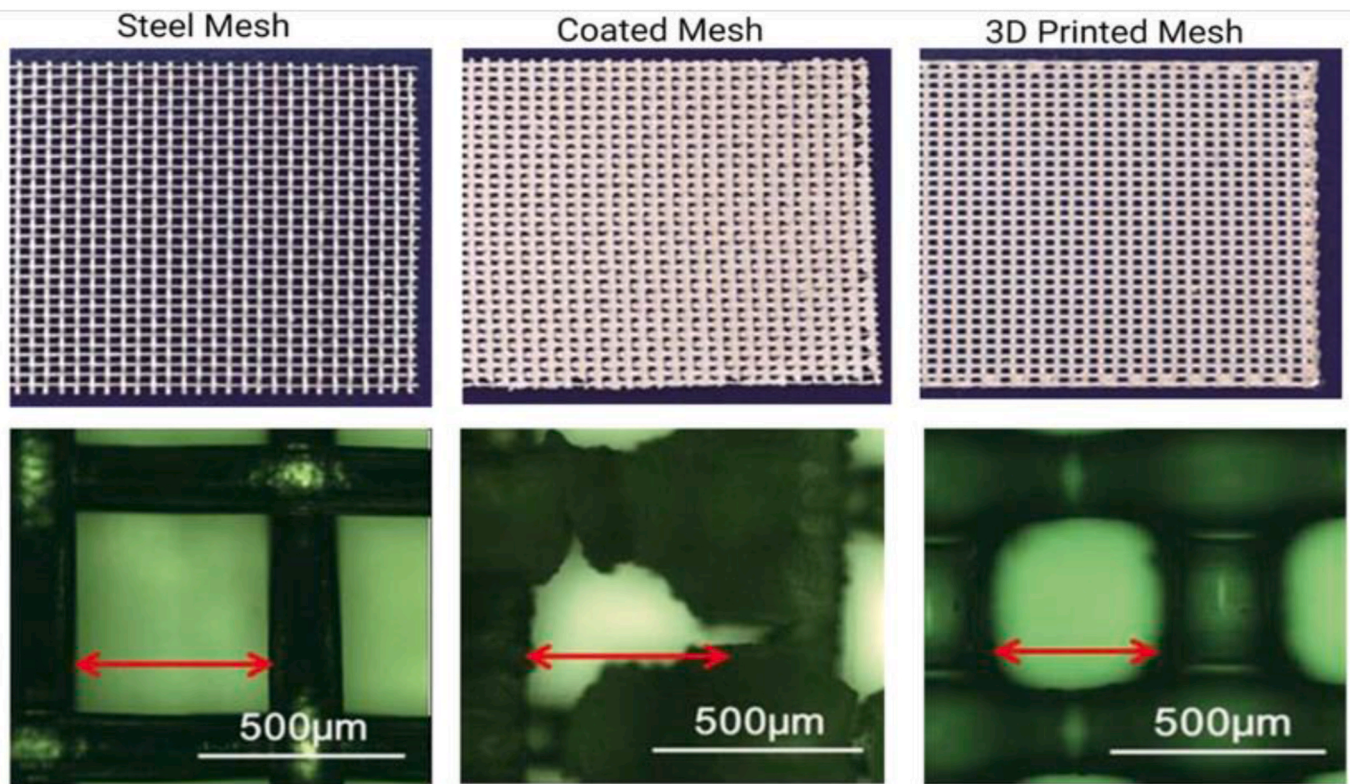


Fig. 6. Difference in uniformity and homogeneity between 3D printed meshes and coated meshes [184].

$\kappa$ -carrageenan [214]. Although the results from this study are promising and highlight the ability of plant-based lipids in improving the mechanical properties of the final print, the overall sensory properties are yet to be optimized to achieve customer acceptance.

Besides the vegan industry, people with difficulty swallowing (dysphagia) and the elderly need special food that is soft but does not compromise the necessary nutritional value. One of the main components used in the dysphagia diet is xanthan gum [215,216] and other natural gums [217]. However, although gums are well printable, they are not enough in terms of nutrition and textural properties for a complete diet. Therefore, studies are investigating the incorporation of various food groups in the preparation of edible 3D printing inks without compromising its printability. These include konjac [215,218], mung bean protein [217], rose powder [217], orange concentrate [153], whey protein isolate [216], gelatin [216], and cellulose nanocrystals [216]. Interestingly, adding cellulose nanocrystals and storing the ink at 4 °C raised its quality from below the standards of the dysphagia diet to a level 5 class [216]. However, further improving the sensory aspects of gum-based inks is required to increase the food's desirability and avoid malnutrition in patients. Additionally, other types of diets require materials with different properties. For example, 3D printing food in space saves costs, space, time and allows room for more variety in food as the inks can be stored in powder form and printed on demand. However, the inks should have a long shelf life to ensure durability in space missions [219,220].

To further enhance the quality of 3D printed food, living cells of various plants can be cultured inside the hydrogel. For example, Vancauwenberghe et al. [147] encapsulated lettuce cells in a 3D printed pectin hydrogel and observed successful cell proliferation that changed the color of the hydrogel from white to green as shown in Fig. 8 (A). Similarly, Park et al. [221] cultured carrot tissues in an alginate-based hydrogel in a process shown in Fig. 8 (B). Although both studies achieved successful cell growth, 3D printing edible plant cultures still suffers from low mechanical stability upon increase in cell density and low

cell adhesion. Further investigations using materials that are compatible with plant cells are needed to advance this field.

In addition to plant cell culturing, natural hydrogels can also be used as a water and nutrient supply in the field of agriculture due to their high water retention and ability to release particles controllably [222]. For example, Dissanayake et al. [223] used 3D printing to prepare an agar/urea hydrogel that provides a slow release of nitrogen as a potential natural fertilizer. 3D printing is also used in other applications in the food and agricultural industries, such as monitoring food safety, food quality, and agricultural environments [224], and ongoing efforts are working on using biomaterials to develop the food and agricultural sectors sustainably.

#### 2.4. Biomaterials for energy applications

As the energy demand is increasing on one side, and the call for sustainable sourcing, manufacturing, and consumption are becoming more urgent on the other side, innovative solutions are necessary to address the challenges of the energy industry. 3D printing using natural materials has emerged as a promising solution in this regard due to its ability to utilize a wide range of materials to fabricate highly detailed structures on micro- and nanoscales for energy devices. These advanced structures possessing hierarchical porosity, high surface area, and controlled microstructures have shown enhancements in the electrochemical and mechanical properties of 3D printed energy devices as opposed to conventionally fabricated devices [225]. Additionally, Fig. 9 shows an example of how 3D printing shortens the manufacturing process by eliminating a few steps. Moreover, the use of naturally sourced materials such as cellulose, PLA, alginate, biochar, biosilica, and carrageenan can be used in the fabrication of energy devices to reduce their environmental footprint and enhance their performance sustainably, however their use in 3D printed energy devices is still in its infancy.

In energy generation and conversion applications, natural materials can be used as 3D printing inks in fuel cells, solar cells, water splitting





Fig. 7. 3D printed vegan seafood-like carrageenan-based prototypes [214].

systems, and other electric generators [225,226]. This is due to their naturally intricate and porous microstructures that provide increased active sites and therefore enhance their energy conversion ability. For example, in the case of conventionally fabricated fuel cells, the range of peak power density is typically 240–415 mW/cm<sup>2</sup> while the highest value achieved by 3D printing fabrication is 727 mW/cm<sup>2</sup> [225,227]. Similarly, in the case of solar cells, the typical range of power conversion efficiency is 12.2 %–18.6 % while the highest achieved by 3D printing fabrication is 26.47 % [225,228]. Although these values prove the potential of 3D printing in improving the efficiency of energy generation devices, they do not specifically represent the results of naturally sourced materials. In other studies, PLA-based inks were used in combination with graphene, nickel, and copper, and other active materials to fabricate electrodes through fused deposition modelling (FDM) [226]. On the other hand, cellulose derivatives, lignin, and alginate were used as base materials to fabricate electrodes through direct ink writing (DIW) [226]. Although these materials can reduce the overall toxicity of the fabricated devices, increase their degradability, and minimize the need for synthetic surface modifications, their use in energy generation devices is yet to be developed and optimized to achieve industry-level standards.

In a similar manner, natural materials including PLA, cellulose, alginate, gelatin, and starch are used in energy storage applications such

as batteries and supercapacitors [226]. Using conventional battery fabrication methods, even biochar, biosilica, carrageenan, and agar are used to make various battery components like electrodes, electrolytes, binders, and separators [229], however, further efforts are required to test and optimize their performance under 3D printing conditions. Fig. 10 summarizes the main properties and biomaterials suitable for use in 3D printing for energy applications. The increasing demand for wearable electronics is driving the energy industry to explore novel materials that have good electrochemical and mechanical properties while providing flexibility and biocompatibility. 3D printing using natural materials not only serves the environmental needs, but also allows the implementation of innovative solutions rapidly, on micro-scales, and with precise accuracy. What remains is applying the knowledge of electrochemical and morphological properties of natural materials and tuning them to leverage their advantages and serve the requirements of energy generation and storage devices.

## 2.5. Biomaterials for bioplastics and food packaging applications

In addition to biomedical, water, food, and energy applications, the utilization of biodegradable natural materials in the packaging and textile industries is becoming increasingly necessary to mitigate the pollution caused by plastics and other materials derived from fossil fuel.

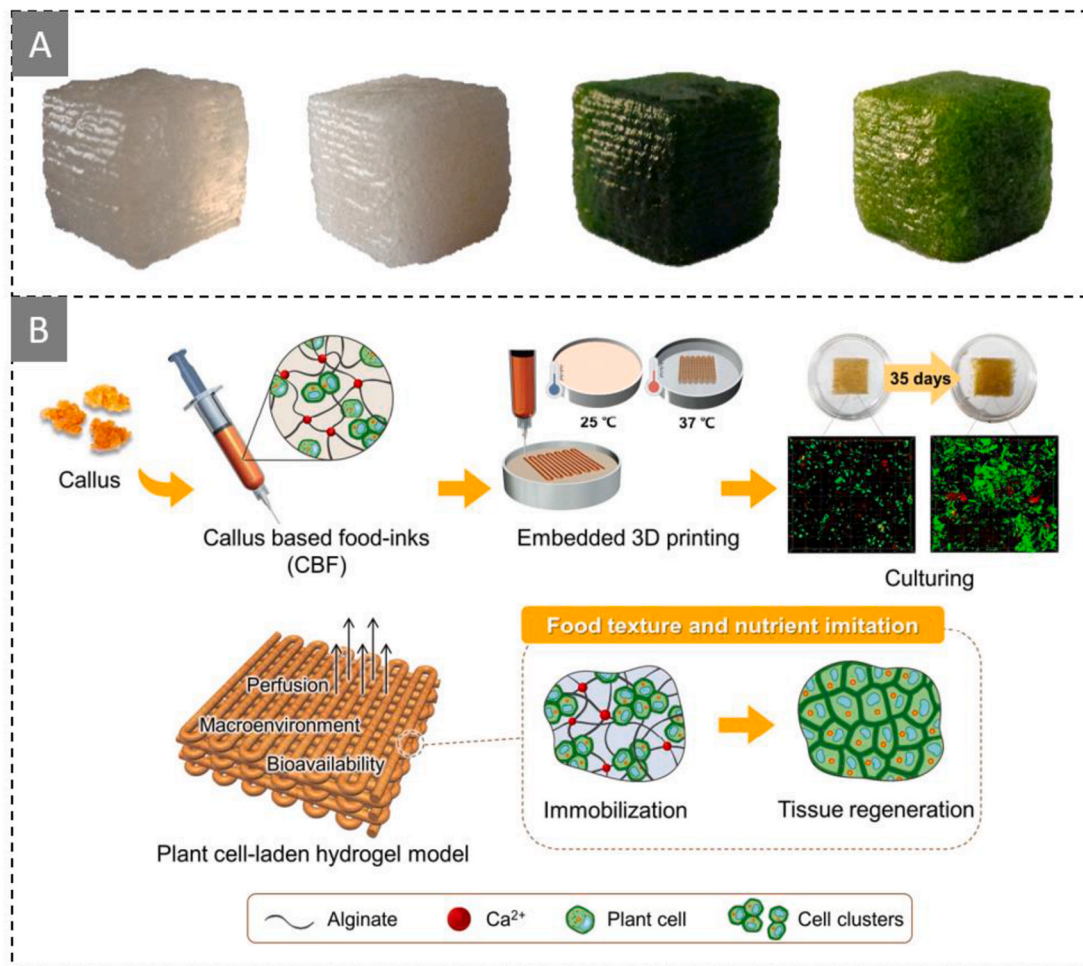


Fig. 8. (A) the growth of lettuce cells in pectin [147]. (B) The process of 3D printing carrot tissues in alginate [221].

However, strength, flexibility, and other mechanical properties are of utmost importance in these industries, and proposed biomaterials should be well-developed to meet the requirements of daily consumer use and the standards set by the plastics, textile, and other related industries. It is worth noting here that although biodegradability is a common feature in naturally occurring materials, modifying and treating these materials to achieve industrial standards can negatively impact their biodegradability.

Studies are investigating the potential of naturally sourced PLA, PHAs, cellulose [230,231], lignocellulose [232,233], starch [230,231], chitosan [234], proteins [235–237], and other biomaterials to develop novel packaging films and foils, edible packaging films, coatings, straws, and even plant pots [235]. PLA is one of the commonly used 3D printing materials due to its favorable mechanical properties and environmental friendliness [238]. Similarly, PHAs are compatible with many nozzle-based and light-based 3D printing techniques [239]. They are desired for their nontoxicity and multifunctionality, however, more efforts are still needed to achieve the processing methods and materials that are compatible with the different PHA types and printing techniques while keeping the cost at the feasible level [239,240].

Food packaging is one of the most important applications in this field, in which various 3D printing techniques can be employed, as shown in Fig. 11. Interestingly, as the technology advances, food packaging is moving towards active and smart types of packaging, which require the use of biomaterials that have certain bioactive properties. In active packaging, biomaterials with anti-microbial and antioxidant properties such as chitosan, soy protein, and gelatin are used to extend the shelf life of the packed food and prevent its spoilage [241]. On the

other hand, smart packaging utilizes the sensitivity of certain materials to humidity, pH, and other parameters to sense and indicate any changes occurring in the packed food. While 3D printing serves the purpose of active packaging by allowing rapid prototyping of customized products, 4D printing is employed in the case of smart packaging to manufacture products that deform over time when exposed to certain stimuli. Reviews of the detailed mechanisms and materials used in 3D/4D printing of active and smart food packaging are available elsewhere in literature [241–243]. Additionally, edible biomaterials such as starch, natural gums, and fruit and vegetable extracts and residues are used to fabricate edible films for food packaging [244]. 3D printing also helps in incorporating nanoparticles in the packaging films, however although these particles are attracting research interest, they still face potential toxicity hazards [244].

### 3. New venues to improve biomaterial printability using artificial intelligence

As researchers strive to enhance the functionality and adaptability of printable natural materials to meet the set requirements for various industries, obstacles related to material compatibility, printability, and stability are frequently encountered. The sensitivity of biomaterial properties towards numerous parameters and conditions requires creative solutions to overcome the huge number of possible combinations and formulations that would otherwise need to be tested experimentally, wasting resources, efforts, and time. Herein, using artificial intelligence comes to serve this purpose.

The integration of AI in bioprinting is a transformative approach that



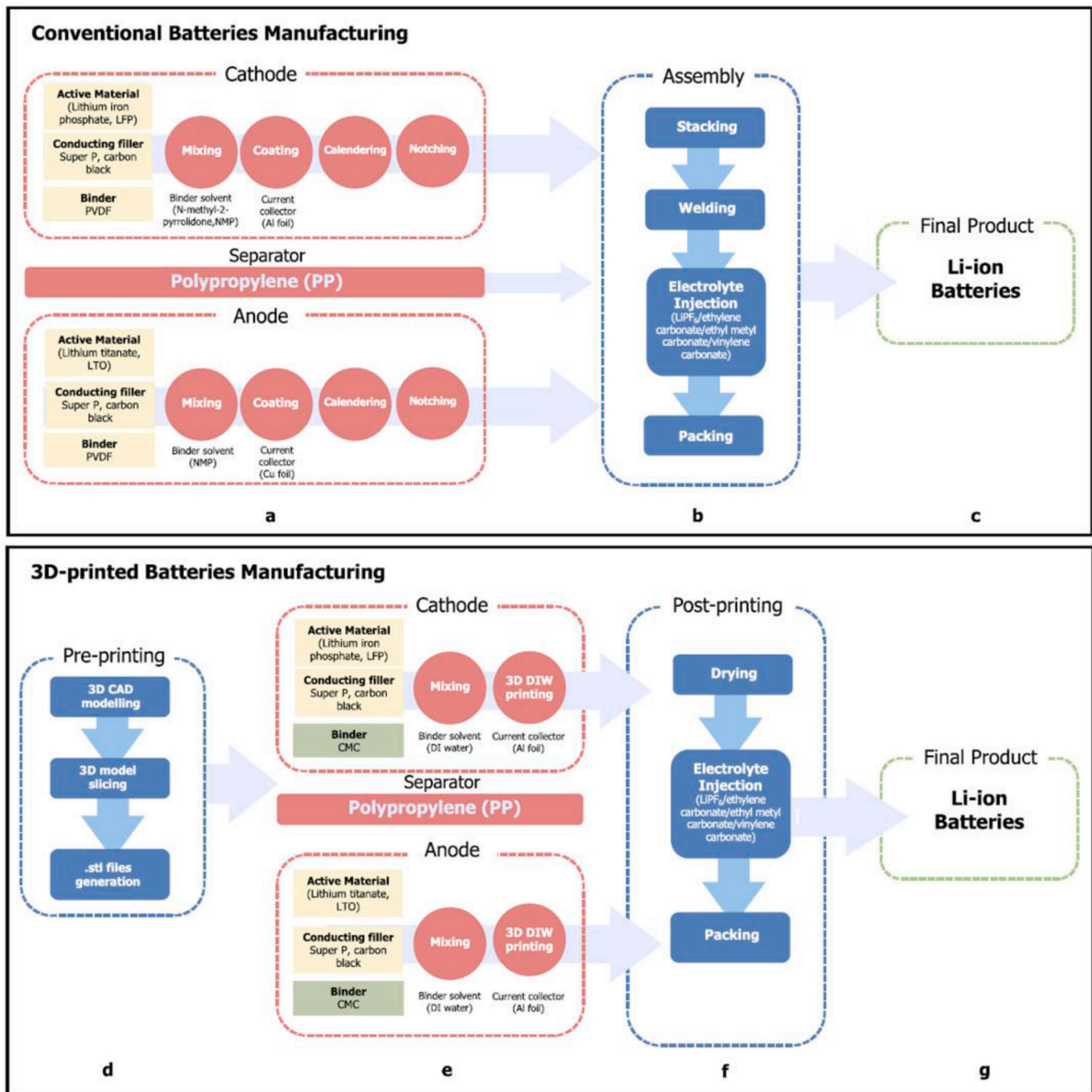


Fig. 9. A comparison between conventional battery manufacturing process and 3D printing process (the case of Li-ion battery) [226].

accelerates developments in the field and opens new opportunities for innovative solutions. Generally, AI can be employed in all stages of 3D printing, including preprinting (image processing, computer-aided design, material selection, printing parameters), through printing (printing method selection, control print defects, identify print failures), and post printing (quality inspection and real-time monitoring) [10]. In another study [11], the involvement of AI in 3D printing is better categorized into 3 levels: (1) open loop AI printing, (2) closed loop AI printing, and (3) predictive AI printing. In open loop printing, AI is used offline for purposes such as scanning nonplanar surfaces before printing or designing and shaping programming structures for 4D printing. In closed loop printing, AI is used online (during printing) for monitoring and real time adaptation based on any defects or moving parts. Lastly,

predictive AI printing is used to predict deformations before they occur and adjust on time. This sophisticated level of involvement is expected to be further developed in the future for applications such as AI-driven surgical 3D printing. Although the aforementioned classifications are valid, this review focuses solely on the aspects related to enhancing biomaterial printability, which are (1) material property prediction, (2) smart material selection, and (3) printing parameter optimization, as shown in Fig. 12. It is worth noting that although most of these methods are currently in the open-loop stage of involvement, they are developing rapidly and have the potential to be integrated in closed loop AI printing. Additionally, the discussions presented in the following subsections are mainly based on information and studies conducted using natural materials, although a few studies using unnaturally sourced materials

# Properties and biomaterials used in 3D printing for energy applications

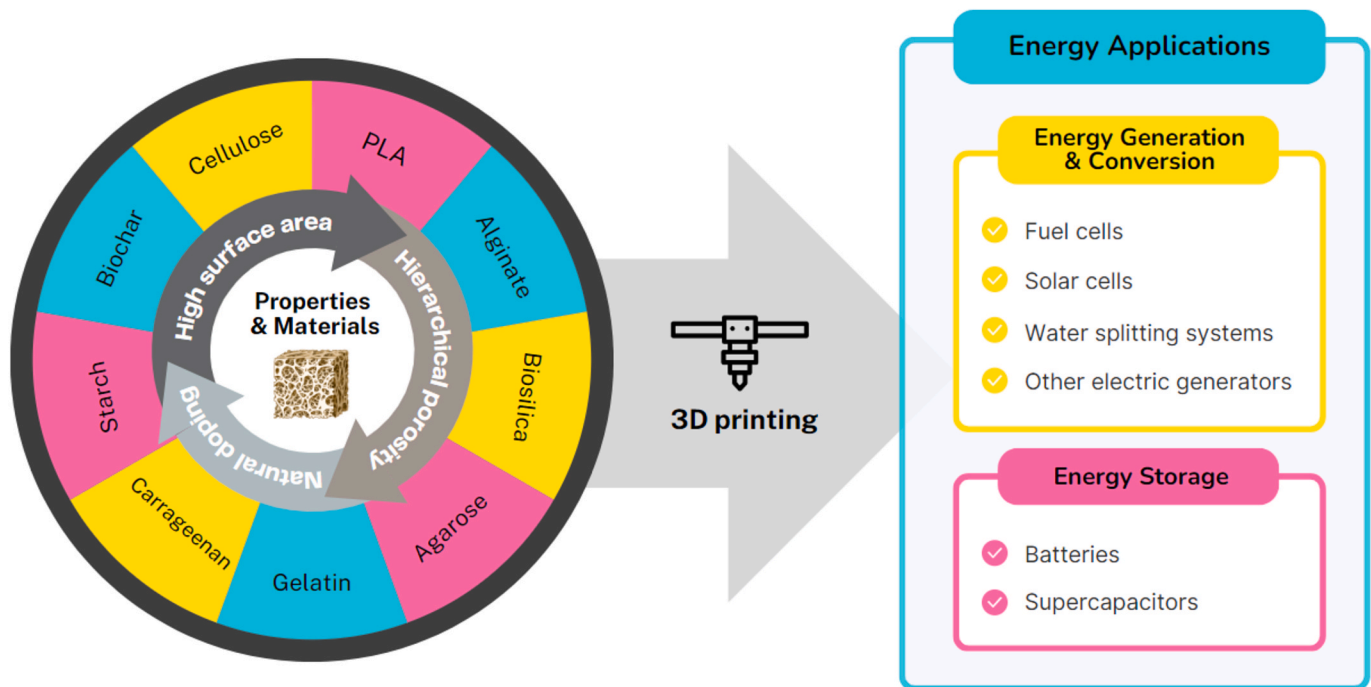


Fig. 10. Properties and biomaterials used in 3D printing for energy applications.

are referred to when deemed necessary and suitable to inspire similar trends to be performed for natural materials.

To apply artificial intelligence for enhancing biomaterial printability, it is essential to understand the various types of machine learning algorithms available and how they can be applied to different challenges in bioprinting. Each type of machine learning—whether supervised, unsupervised, semi-supervised, or reinforcement learning—offers distinct advantages depending on the specific problem at hand, and can be used for different applications. Understanding the characteristics and appropriate applications of these algorithms will enable researchers to select the most effective approaches for improving printability and overcoming material compatibility issues. Table 4 provides a comparative overview of these machine learning types and summarizes their key features.

Among the diverse machine learning models, some are more commonly used in bioprinting, such as decision trees, random forests, artificial neural networks (ANNs), and convolutional neural networks (CNNs). Each of the mentioned models have their distinct advantages, limitations, and applications as discussed below.

Linear and regression-based models are basic tools that can be used in bioprinting for parameter analysis and prediction. Multiple Linear Regression (MLR) is a straightforward model that can predict outcomes such as cell viability or tissue properties based on printing parameters [251]. Its relatively direct interpretability makes it ideal for baseline predictions and identifying key variables, although it assumes linear relationships, which can limit its applicability in more complex scenarios. LASSO (Least Absolute Shrinkage and Selection Operator) Regression takes a step further by focusing on feature selection to pinpoint critical parameters influencing outcomes, simplifying models for easier interpretation [252]. However, its reliance on linearity may cause it to miss potential interactions between variables. Bayesian Optimization (BO) stands out in design optimization by significantly

reducing the number of required experiments [253]. Using limited data, BO identifies optimal process parameters and offers efficient prediction capabilities, making it a powerful tool for streamlining experimental workflows.

Tree-based models are widely utilized in different fields for their ability to handle both linear and non-linear relationships effectively. Decision trees (DT) are particularly valued for their interpretability, as they can classify tissue types or predict properties based on printing parameters. They are especially useful in preliminary analyses and feature selection due to their simplicity and visual appeal [254]. For more complex tasks, Random Forests (RF) provide a robust alternative. By aggregating multiple decision trees, RF can manage intricate relationships and produce advanced predictions while reducing the risk of overfitting. Additionally, RF offers valuable insights into variable importance, enhancing its utility in bioprinting applications.

Additionally, neural networks have the ability to adapt to complex patterns and datasets. Artificial Neural Networks (ANNs) excel in generating new designs or predicting outcomes with remarkable accuracy. Despite their effectiveness, ANNs operate as black-box models and demand extensive training data, which can be a limitation. Moreover, Convolutional Neural Networks (CNNs) are specifically tailored for image analysis, making them indispensable for real-time feedback and quality control in bioprinting. By analyzing structure images, CNNs provide detailed insights into printed outputs, although their computational intensity requires significant resources [255].

Moreover, generative models offer innovative solutions for design and parameter generation. Generative Adversarial Networks (GANs) are renowned for their ability to create synthetic designs by capturing detailed relationships within large datasets. This capability allows for the exploration of novel parameters and structures, expanding the boundaries of bioprinting innovation. Conditional GANs (CGANs) build on this foundation by introducing condition-specific data generation,

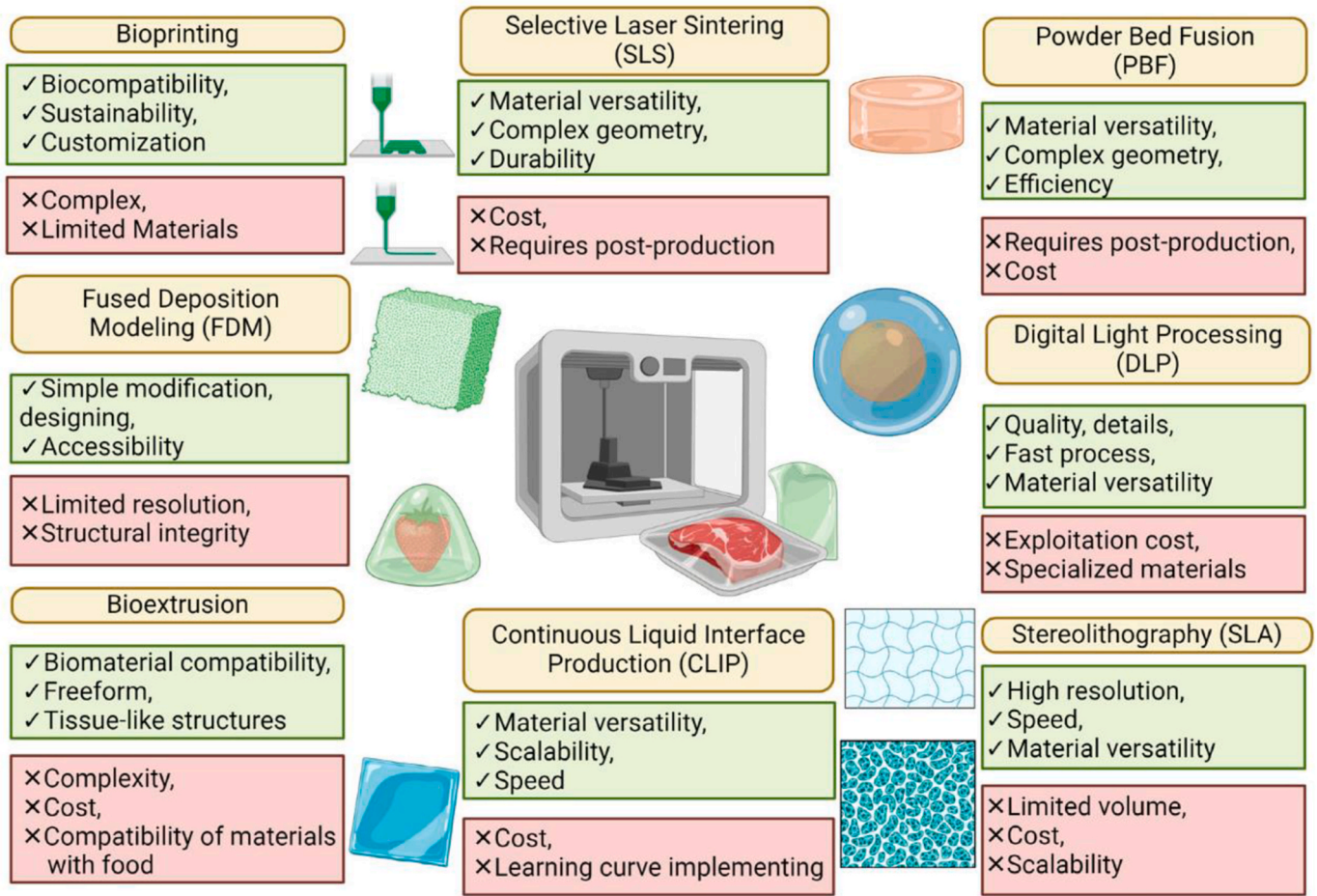


Fig. 11. Comparison of 3D printing techniques for food packaging applications [241].

## AI for Printable Material Development

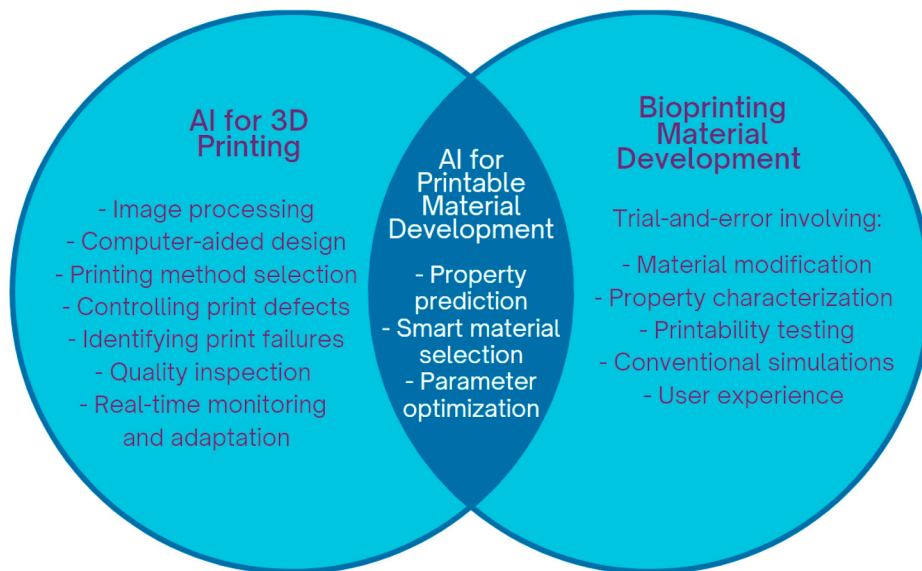


Fig. 12. AI for printable material development.

enabling tailored designs and fostering condition-driven advancements in bioprinting applications. These models find their applications in biomimetic design [256].

Furthermore, reinforcement learning models are uniquely suited for real-time optimization in dynamic bioprinting processes. For example, Q-Learning can be used to optimize printing parameters by making



**Table 4**  
Comparative overview of machine learning types.

| Type                     | Description                                                                                                                                                                                                             | Key Features                                                                                                                                                                                              | Examples of Algorithms/Models                                                                                                                                                                                        | Refs.          |
|--------------------------|-------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|----------------|
| Supervised Learning      | <ul style="list-style-type: none"> <li>- Learns a function that maps input to output based on labeled input-output pairs.</li> <li>- Uses labeled data to infer a function.</li> </ul>                                  | <ul style="list-style-type: none"> <li>- Requires labeled data.</li> <li>- Goal-oriented approach.</li> <li>- Error is calculated based on the difference between predicted and actual output.</li> </ul> | <ul style="list-style-type: none"> <li>- Linear and logistic regression</li> <li>- Naïve Bayes classification</li> <li>- Support Vector Machines (SVM)</li> <li>- Decision Trees</li> <li>- Random Forest</li> </ul> | [245, 246]     |
| Unsupervised Learning    | <ul style="list-style-type: none"> <li>- Analyzes unlabeled data without human supervision.</li> <li>- The algorithm identifies patterns or structures within the data by itself.</li> </ul>                            | <ul style="list-style-type: none"> <li>- Data-driven approach.</li> <li>- No labeled data required.</li> <li>- Focus on extracting features, trends, and groupings.</li> </ul>                            | <ul style="list-style-type: none"> <li>- Clustering</li> <li>- Dimensionality reduction</li> <li>- Anomaly detection</li> <li>- Feature learning</li> </ul>                                                          | [246, 247]     |
| Semi-Supervised Learning | <ul style="list-style-type: none"> <li>- A hybrid approach combining labeled and unlabeled data.</li> <li>- Uses a small amount of labeled data and a large amount of unlabeled data for better prediction.</li> </ul>  | <ul style="list-style-type: none"> <li>- Combines both labeled and unlabeled data.</li> <li>- Effective when labeled data is scarce.</li> </ul>                                                           | <ul style="list-style-type: none"> <li>- Semi-supervised clustering.</li> <li>- Semi-supervised classification.</li> </ul>                                                                                           | [246, 248,249] |
| Reinforcement Learning   | <ul style="list-style-type: none"> <li>- Learns by interacting with an environment and receiving feedback in the form of rewards or penalties.</li> <li>- Focuses on optimizing actions to maximize rewards.</li> </ul> | <ul style="list-style-type: none"> <li>- Environment-driven approach, based on reward/punishment.</li> <li>- Optimizes decisions over time.</li> </ul>                                                    | <ul style="list-style-type: none"> <li>- Q-Learning</li> <li>- Deep Q-Network (DQN)</li> <li>- Proximal Policy Optimization (PPO)</li> </ul>                                                                         | [246, 250]     |

adaptive state-action decisions, ensuring process adjustments are made efficiently during operations [257]. Deep Q-Networks (DQN) take this optimization a step further by focusing on spatial aspects, such as enhancing printer head movements. Through their ability to learn complex relationships and apply them in real-time, DQNs contribute to achieving precise and efficient printing processes.

The following subsections discuss the recent developments in using machine learning to enhance material properties for bioprinting applications.

### 3.1. Material property prediction

Predicting the properties of printable biomaterials serves as a basic building block in developing advanced 3D printing inks for specialized applications. Based on these predictions, the printing techniques, conditions, and parameters can be adjusted. Biomaterial-based ink formulations can also be further optimized according to the suggested

predictions without undergoing a long series of experimental trial-and-error or even lengthy simulations. For example, using the linear regression (LR) model to predict and rank various geometries of soft/brittle materials based on their toughness and strength was proven around 250 times faster than achieving the same results using finite element model (FEM) simulations [258]. Although this high speed does not predict detailed values of strength and toughness, it successfully predicts the rankings of 1 million geometries based on limited labeled data in less than a second. On a large scale, this approach is useful in filtering a huge number of possible combinations and shortlisting the best ones for further exploration and optimization. Interestingly, this study proves that using more complex machine learning methods such as convolutional neural networks (CNN) does not necessarily yield better ranking predictions despite their higher accuracy.

On a more specific level, machine learning methods can predict the mechanical properties of various biomaterials, as shown in Table 5. These properties include tensile strength, yield strength, stiffness,

**Table 5**  
Properties predicted by AI methods to improve the printability of various biomaterials.

| Property                            | AI methods                                        | 3D printing material                                                     | Prediction accuracy & Best methods                                                                                                                            | Refs.          |
|-------------------------------------|---------------------------------------------------|--------------------------------------------------------------------------|---------------------------------------------------------------------------------------------------------------------------------------------------------------|----------------|
| Shape fidelity                      | Inductive logic programming – ILP                 | Atelocollagen (AC)                                                       | Higher elastic modulus gives better shape fidelity while extrusion is best achieved below the critical yield stress.                                          | [251]          |
| Extrudability                       | (relative least general generalization Algorithm) |                                                                          |                                                                                                                                                               |                |
| Extrusion pressure                  | Regression and classification methods             | Alginate and gelatin                                                     | RF prediction error: 34.4%–44.4 %<br>LR prediction error: 13.2%–73.4 %                                                                                        | [260]          |
| Printing temperature                | PLSR and PCR<br>Regression analyses               | Surimi paste<br>968 drug formulations were studied                       | PLS has the best prediction accuracy<br>ANN: best results                                                                                                     | [261]<br>[262] |
| Microneedle features                | Classification methods and DL                     | Biodegradable PLA                                                        | ConvNeXt_Base DL model has highest accuracy (0.96) and precision (0.96)                                                                                       | [263]          |
| Inkjet droplet velocity             | RFs, LASSO, XGB, SVR                              | Sodium alginate hydrogel                                                 | XGB: best prediction performance                                                                                                                              | [254]          |
| Inkjet droplet volume               | DT, RF, DL                                        | 210 biomaterial formulations                                             | RF: best accuracy (88.1 %) and precision (90.6 %)                                                                                                             | [264]          |
| Printability or printability window | RF, MLP, SVM<br>SVM, RF, ANN, KNN, LR             | 687 drug formulations were studied<br>968 drug formulations were studied | DL: best recall (87.3 %)<br>Printability prediction accuracy: 97.22 %<br>Print quality prediction accuracy: 97.14 %<br>Printability prediction accuracy: 93 % | [265]<br>[262] |
| Tensile strength                    | OLS LR, RF, LGBM, XGB, ANN                        | PLA                                                                      | XGB: best tensile strength prediction accuracy                                                                                                                | [259]          |
| Stiffness, strength, and toughness  | Image-driven CNN                                  | PDMS                                                                     | Process-structure-property studied                                                                                                                            | [266]          |
| Plastic viscosity                   | LR, SVR, RF, XGB, MLP                             | Concrete                                                                 | XGB: best plastic viscosity and yield stress prediction accuracy                                                                                              | [267]          |
| Yield stress                        |                                                   |                                                                          |                                                                                                                                                               |                |
| Dissolution profile                 | MLR, DT, SVM                                      | PCL with various amounts of ciprofloxacin and PEG                        | DT: best accuracy                                                                                                                                             | [268]          |

toughness, plastic viscosity, and shape fidelity. Ege et al. [259] employed five machine learning models to analyze the impact of 3D printing parameters (i.e., nozzle temperature, printing speed, and layer height) on the tensile strength of PLA, and predict the tensile strength values. To achieve this, datasets containing 68 combinations of process parameters and tensile strengths were used from literature: 80 % for training and 20 % for testing. Their results show that in comparison to the ordinary least squares linear regression (OLS LR), random forest (RF), light gradient boost (LGBM), and artificial neural network (ANN) models, the extreme gradient boost (XGBoost or XGB) model shows the best accuracy in predicting the tensile strength values as illustrated in Fig. 13. Additionally, the study concludes that high infill density and low printing speed result in better prediction accuracy. This method can further be developed to understand the importance of other 3D printing parameters such as nozzle pressure and strut size. Further research should also investigate the methods' accuracy using other biomaterials.

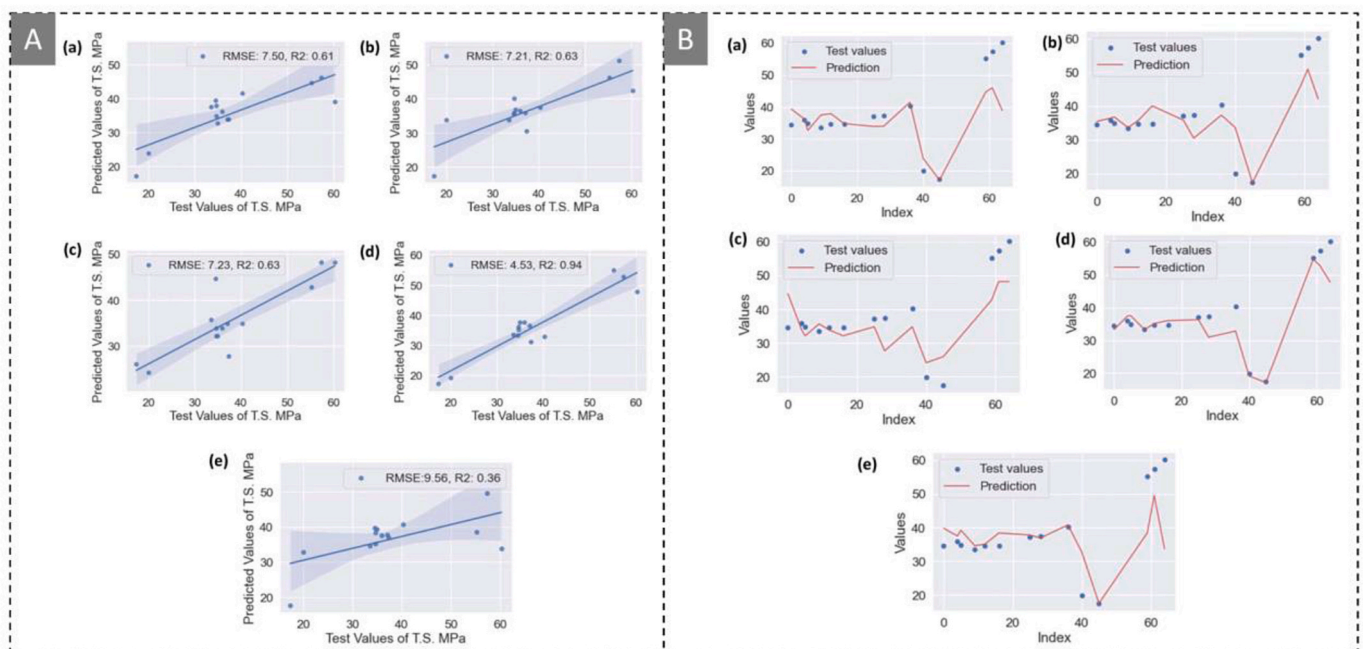
Similarly, plastic viscosity and yield stress were best predicted by the XGB model over linear regression (LR), support vector regression (SVR), random forest (RF), and multilayer perceptron (MLP) models [267]. Based on the plastic viscosity and yield stress, the printability window was predicted, and close results were obtained experimentally, as shown in Fig. 14. Although these results were obtained for concrete rather than natural biomaterials, the same approach of varying experimental parameters and obtaining rheological data to train the predictive models can be applied to other types of natural materials. In another study [264], the printability windows of various biomaterial formulations were predicted using decision tree (DT), random forest (RF), and deep learning (DL) models, as shown in Fig. 15. The models were trained using the printability data and formulation compositions of 210 formulations which consist of 16 biomaterials and that were 3D printed to test their printability (Fig. 15 A). Among the three models, RF shows the best prediction accuracy (88.1 %) and precision (90.6 %). RF excels in overall performance due to its ensemble approach, which combines multiple DT trained on different samples of the data. This diversity among trees reduces overfitting and enhances the model's generalization ability. On the other hand, DL has the highest recall (87.3 %) and finest map granularity as noticed in Fig. 15 B. These printability window prediction methods are of high importance when dealing with large

numbers of bioink formulations, such as in the pharmaceutical industry. Studies have collected 900+ drug formulations to train machine learning models to predict the ink printability using filament hot melt extrusion [262] and inkjet [265] printing. Furthermore, the drug release times and dissolution profiles can also be predicted using machine learning algorithms [262,268].

The relationship between printability and mechanical properties is further analyzed by Lee et al. [251]. They used an inductive logic programming method to analyze the importance of various properties and found that higher elastic modulus gives better shape fidelity while extrusion is best achieved below the critical yield stress. This guideline was used to develop mathematical prediction models and optimize atelocollagen (AC) bioink formulations. Although the developed models are material dependent, a similar approach can be used in future research to predict the relationship between the properties and printability of other types of bioinks.

Furthermore, machine learning can also be used to predict the 3D printing process parameters such as extrusion pressure [260,261], temperature [262], inkjet droplet velocity and volume [254], and even microneedle features [263] for biomaterials such as alginate, gelatin, surimi paste, PLA, and various drug formulations as shown in Table 5. Prediction of printing parameters is of extreme importance as it leads to parameter optimization which is necessary since the parameters determine the print quality and they are material dependent. Parameter optimization is discussed in section 3.3.

The mechanical properties of biomaterial-based inks can also be predicted through its microstructure using image-driven conventional neural networks (CNN) [266]. Fig. 16 shows the experimental and predicted values of stiffness, strength, and toughness of a 3D printed soft hyperelastic polydimethylsiloxane-based (PDMS) hybrid composite using a CNN prediction algorithm that was trained on images of microstructures from the printed specimens. This method needs more effort to be developed because it has the potential to minimize trial-and-error to a great extent as it depends on the visual microstructures rather than the printability data like other methods.



**Fig. 13.** A: Experimental values of tensile strength vs. predicted values by (a) OLS LR (b) RF (c) LGBM (d) XGB (e) ANN. B: Comparison between experimental and predicted values [259].

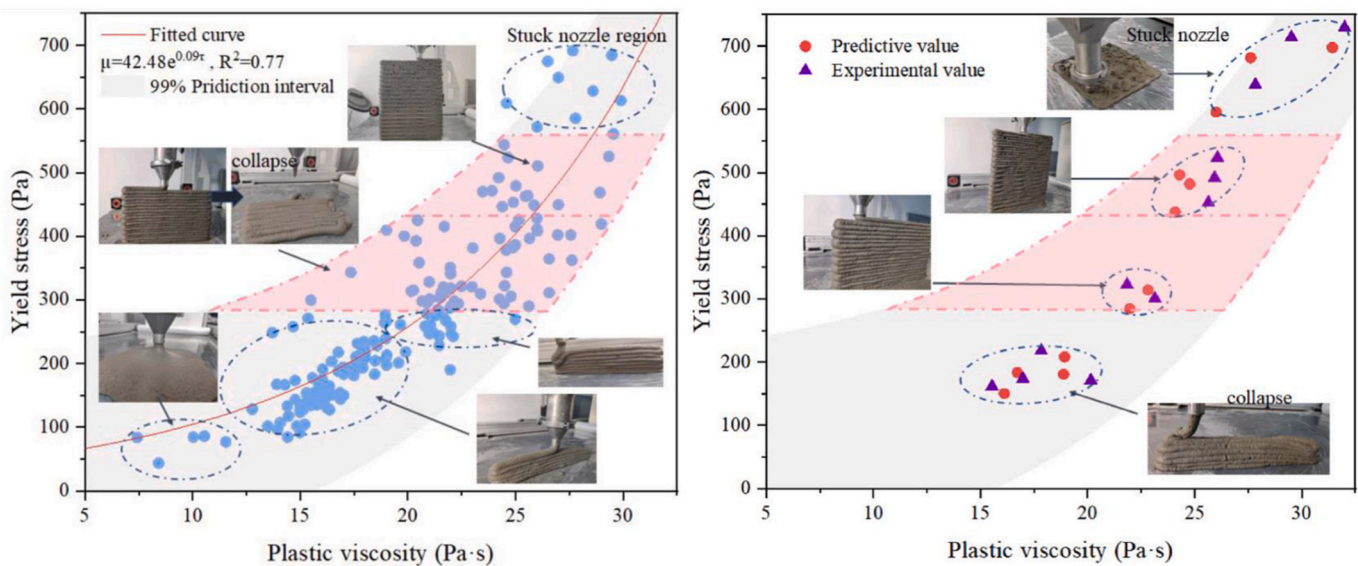


Fig. 14. Predicted values (left) and alignment between experimental and predicted values by XGB model (right) [267].

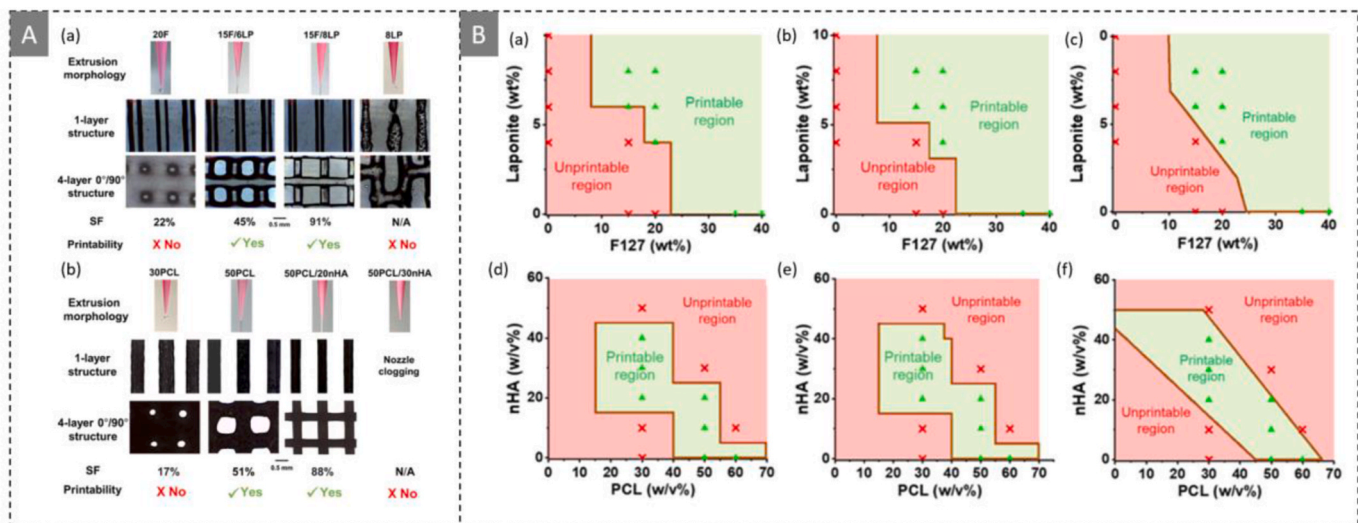


Fig. 15. A: examples of the printability assessment performed for all 210 formulations. B: printability windows for various combinations of biomaterials as predicted by DT (a, d), RF (b, e), and DL (c, f) [264].

### 3.2. Smart biomaterial selection

Upon predicting the properties of various material combinations, it becomes possible for the AI algorithms to select the material that has the most suitable properties for the desired application, whether it has previously been used or not. For example, in an attempt to optimize the printing process of a silicon elastomer using hierarchical machine learning, the results suggested a unique silicone ink formulation that was not used before [269]. On the other hand, Rojek et al. used ANN [270] and CNN [271] to select the best out of two known materials which are PLA and PLA+. Their results indicate that PLA+ is the most suitable for printing the targeted hand exoskeleton parts and optimized printing parameters are also obtained. The selection of the material was based on optimum tensile strength, however, further explorations following similar methods should take into account various parameters that determine the suitability of the material for the targeted 3D printing application.

Additionally, the best combination of two materials out of a large number of combinations can also be selected by machine learning (ML)

models. In a study by Gu et al. [258], the mechanical properties of 1 million combinations of soft and brittle materials are predicted by machine learning models and then the best 25 composites are shortlisted based on initial data that only indicates whether the combination is “good” or “bad”. The models use classification probabilities and rankings to quickly identify the best designs, saving significant time and computational effort compared to FEM simulations. Although this method gives results that are comparable to FEM simulations (Fig. 17) in a much shorter time, its performance is yet to be investigated on a larger scale and with more advanced structures and geometries.

Similarly, Yu et al. [272] employed deep learning to find the best design of a soft/brittle hydrogel that has the highest fracture toughness. They propose a hierarchical design method based on reinforcement learning, in which it starts with low resolution images and once the best design is obtained the resolution is increased leading to larger sized systems, as illustrated in Fig. 18. This method selects the optimized material in less time and cost compared to non-hierarchical design methods. Moreover, in terms of material structure, artificial intelligence plays an important role in optimizing designs to achieve the required



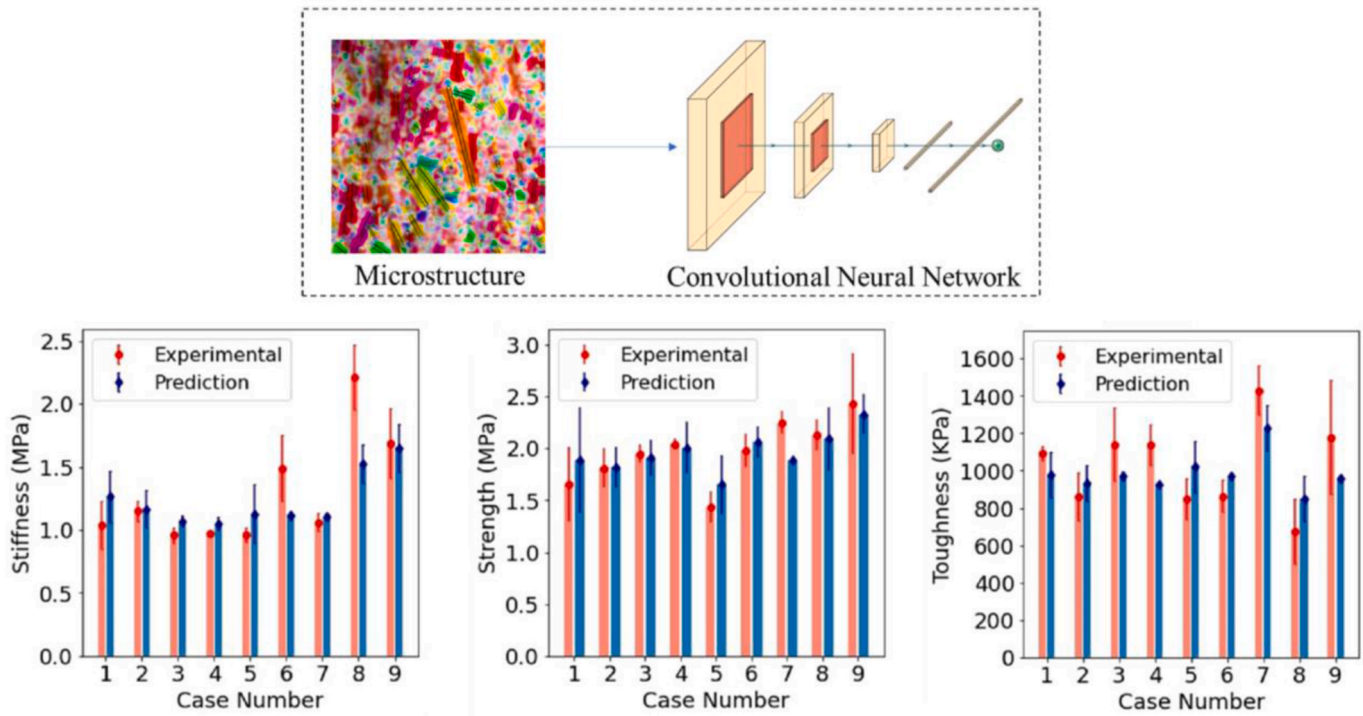


Fig. 16. Values of stiffness, strength, and toughness of PDMS-based specimens obtained experimentally and via image-driven CNN predictions [266].

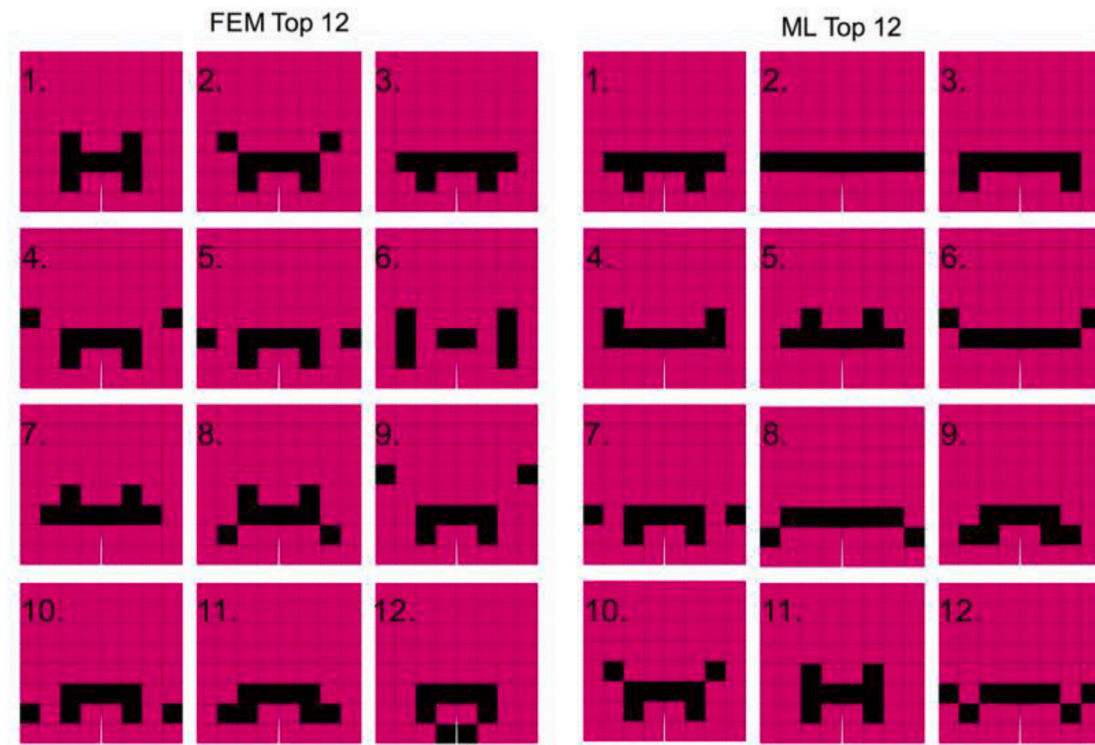


Fig. 17. Comparison between FEM and ML generated high-toughness geometries made of stiff (pink) and brittle (black) materials [258].

properties. Comprehensive reviews on the use of machine learning and artificial intelligence in the field of mechanical design are available in the literature [273].

### 3.3. Printing parameter optimization

The quality of 3D printed structures not only depends on the material properties, but also on the 3D printing technique and process parameters. Optimizing printing parameters for biomaterials is challenging due to many reasons. One of the main reasons is that the compositions of

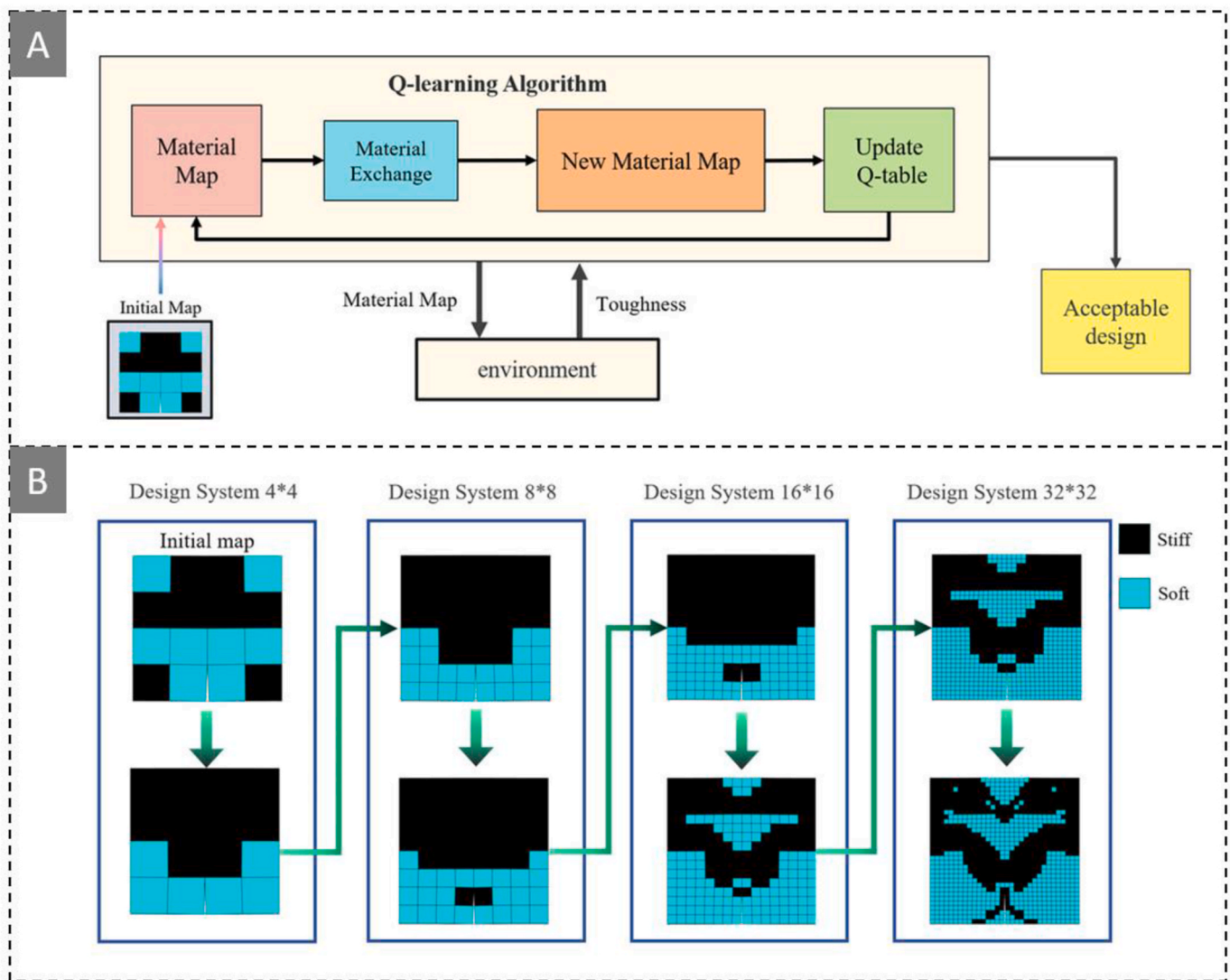


Fig. 18. A: Hierarchical reinforcement learning algorithm for best soft/stiff composite. B: Gradual increase in resolution throughout the design process [272].

biomaterial formulations in research are continuously changing and many times they are unique to the case they are used in. The composition and properties can also vary from batch to batch since the materials are naturally sourced. This means each material has limited documentation on its properties and behavior upon 3D printing. Consequently, every time a new material is to be explored, lengthy processes of parameter optimization should be undertaken. This slows down the development of novel materials and formulations that have the potential to solve certain problems but are limited by their compatibility with the available 3D printing processes and the user's experience regarding parameter optimization. Additionally, many 3D printing techniques are highly customizable and allow the adjustment of a wide array of printing parameters, and most of the parameters are dependent on the type of material used. This results in a huge number of possible combinations that should be tested experimentally to select the best setup, meaning excess resources, time, and money should be spent.

Alternatively, conventional simulation methods can be used, but in the case of novel biomaterials their accuracy is limited. For example, cases that involve spreading, dynamic viscosity, interlayer interactions, and multiple nozzle printing might not be accurately simulated [274]. To increase the accuracy of the optimization process and decrease its reliance on the knowledge of the user, conventional simulation methods can be coupled with machine learning approaches to achieve a process that learns from a given number of experiments and suggests optimized

parameters. This method does not only speed up the optimization process, but also uses less resources and requires a reduced number of experimental trials. Finite element modeling (FEM) is one of the most used numerical methods in combination with machine learning approaches [275]. FEM and machine learning methods are complementary because FEM requires detailed knowledge of the material's properties and behavior upon printing which is not always available in the case of newly developed biomaterials, while on the other hand machine learning techniques can predict these properties, though requiring a large amount of data which can be generated using a combination of FEM and experimental data. This concept is explained in more detail by Baturynska et al. [275] in their conceptual framework study developed for a powder bed fusion printing system, and is expected to further be investigated for other systems. Another optimization algorithm that can be coupled with machine learning is the whale optimization algorithm (WOA) [276]. In a study by Kumar et al. [276] a hybrid WOA-ANN method was used to optimize the surface roughness, production time, and volume percentage error of an FDM printing process using PLA. Using this approach, a large dataset of different combinations is first generated, then the relations between inputs and outputs were determined by the coupled optimization and machine learning approach, revealing that the layer thickness and nozzle temperature have the most significant impact on the surface roughness of the prints [276].

Machine learning is also used independently with experimental trials

of various 3D printing techniques to optimize printing process parameters that are unique to each technique, despite the type of ink. For example, CNN was used for TPL (Two-Photon Lithography) to optimize the light dosage, ANN for WAAM (wire-arc additive manufacturing) to optimize the printing toolpath, and GPR (Gaussian Process Regression) for PBF to optimize its process window [277]. However, when introducing biomaterials that have sensitive properties and require strictly optimized conditions, optimization becomes challenging and more complicated, as the number of involved parameters increases. In spite of its complexity, it is at this point where machine learning is actually needed because otherwise either developing such types of novel 3D printing biomaterials will be hindered, or exhaustive experimental trials and lengthy conventional simulation runs should be performed. Table 6 shows the parameters that were optimized by various studies using machine learning algorithms for 3D printing using various biomaterials and printing techniques. Ideally, using machine learning algorithms, it should be possible for the 3D printer user to set the desired output properties, and the system suggests the best input parameters based on its optimized values. This scenario is possible, but research is currently working on the training phase of the machine learning algorithms to reduce the time and energy they consume while maintaining high accuracies and similarities to the experimental results.

Moreover, integrating a graphical user interface (GUI) to facilitate communication between the user and the system allows material scientists and engineers that are not well experienced in machine learning and/or additive manufacturing to utilize the benefits of the complex optimization systems conveniently. A user-friendly interface was developed by Rahmani et al. [281] in which it takes in the CAD design that is desired to be printed and printing parameters (i.e., pressure and temperature) as an input and outputs an index representing the similarity between the design and the predicted printing quality, as shown in Fig. 19. The system was trained on various printing parameters and designs having a variety of complexity indices obtained from the number of intersections in each design. This approach is beneficial when a few trials using a certain material are available and the material's behavior when printing different structures is to be investigated. However, only simple structures were tested using this method and limited parameters were adjustable. For the development of biomaterial inks, more parameters should be involved, and more complex structures should be explored.

Upon integrating more parameters, the machine learning optimization system becomes more advanced and various training approaches can be taken. Among these approaches, Ruberu et al. [253] employed a

system based on Bayesian optimization (BO) to reduce the number of experiments needed to optimize the GelMA/HAMA extrusion process from 6000+ experiments down to a range of 4–47. To achieve this, filament morphology and layer stacking are first assessed and scored, as shown in Fig. 20 (A). Then, the data is fed into the BO system, it suggests new improved data, the new data is tested experimentally, the experimental data is fed into the optimizer, and the cycle goes on until the optimal print is achieved, as illustrated in Fig. 20 (B). Although this approach reduces the number of experimental trials needed considerably, it still requires human interference in each cycle. This interference can perhaps be replaced in the future by automatic robots that perform the required experimental tests to achieve a fully automated optimization process. The advantages of this system include significantly reducing the number of required experiments, enabling step-by-step refinement for improved accuracy, and creating opportunities for full automation through robotic integration in the future.

Another method to train machine learning tools to achieve printing parameter optimization is by using image analysis and classification as a first step. In extrusion 3D printing, combinations of various printing parameters can be used to generate prints which are then photographed and fed into the AI-based optimizer to obtain the best combination. In a study by Chen et al. [279], a pneumatic extruder is programmed to use multiple combinations of printing pressures and speeds, an inclined printing surface is used to alter the nozzle distance as shown in Fig. 21 (A), and the printed trials are imaged. The combination of the programmable extruder and the AI-assisted image analysis algorithm constructs a High-Throughput Screening system (AI-HTPCSS) that is able to produce phase diagrams of the printable window and optimize the 3 given parameters. Results show that the proposed system can optimize the printing conditions of the alginate/gelatin ink and achieve satisfying mechanical and biological properties to accelerate the process of wound healing. This method can further be tested with other biomaterials as it shows good potential for extrusion printing optimization. As this system becomes more mature, additional parameters such as temperature can be incorporated, as many biomaterials are generally temperature sensitive.

Similarly, image analysis in combination with machine learning can be used to optimize printing parameters of aerosol jet printing (AJP) and minimize the effects of high overspray, limited line thickness, and high edge roughness [280]. Liu et al. [280] developed an optimization system based on machine learning algorithms shown in Tables 6 and in which line features and morphology are extracted from images of experimental trials (Fig. 21 B), and then the correlation between the input and output

**Table 6**  
Printing parameters optimized, and the respective ML methods, biomaterials, 3D printing techniques used.

| Optimized parameters                                                      | Input parameters                                                                       | AI methods                                       | 3D printing material                                                            | Printing method                     | Refs. |
|---------------------------------------------------------------------------|----------------------------------------------------------------------------------------|--------------------------------------------------|---------------------------------------------------------------------------------|-------------------------------------|-------|
| Ink composition, temperature, pressure, speed                             | Filament formation, layer stacking, printability score                                 | Bayesian optimization (BO)                       | GelMA & HAMA                                                                    | Extrusion printing                  | [253] |
| Layer fusion, stringiness, infill                                         | Ink material & flow speed, bath material & conc., needle diameter, retraction distance | Hierarchical machine learning (HML)              | Silicon                                                                         | Freeform reversible embedding (FRE) | [269] |
| Droplet diameter & speed                                                  | Voltage, viscosity, surface tension, nozzle diameter                                   | Fully connected neural networks (FCNNs)          | Cell laden bioinks: Dulbecco's Modified Eagle Medium (DMEM) and sodium alginate | Drop-on-demand (DOD) bioprinting    | [278] |
| Material, layer height, shell thickness, fill density, speed, temperature | Max. Tensile force & dimensions                                                        | Artificial neural networks (ANN)                 | PLA, PLA+                                                                       | Fused deposition modelling (FFD)    | [271] |
| Printing pressure, nozzle speed, and printing distance                    | Images of bioink during and after printing                                             | Deep learning (DL)                               | Alginate & gelatin                                                              | Pneumatic extrusion bioprinting     | [279] |
| Surface roughness, production time, volume percentage error               | Layer thickness, nozzle temp., speed, and raster width                                 | Artificial neural networks (ANN)                 | PLA                                                                             | FDM                                 | [276] |
| Line morphology                                                           | Line image analysis                                                                    | Support vector machine (SVM), K-means clustering | Clariant® silver nanoparticles                                                  | Aerosol jet 3D printing (AJP)       | [280] |
| Similarity index (proper printing, over-printing, or under-printing)      | Image analysis (prints with varying pressure, temperature, and complexity index)       | LR, SVR, DT, RF, GBR & XGB, BR, NNs              | Nivea Crème (proof-of-concept study)                                            | Pneumatic extrusion bioprinting     | [281] |



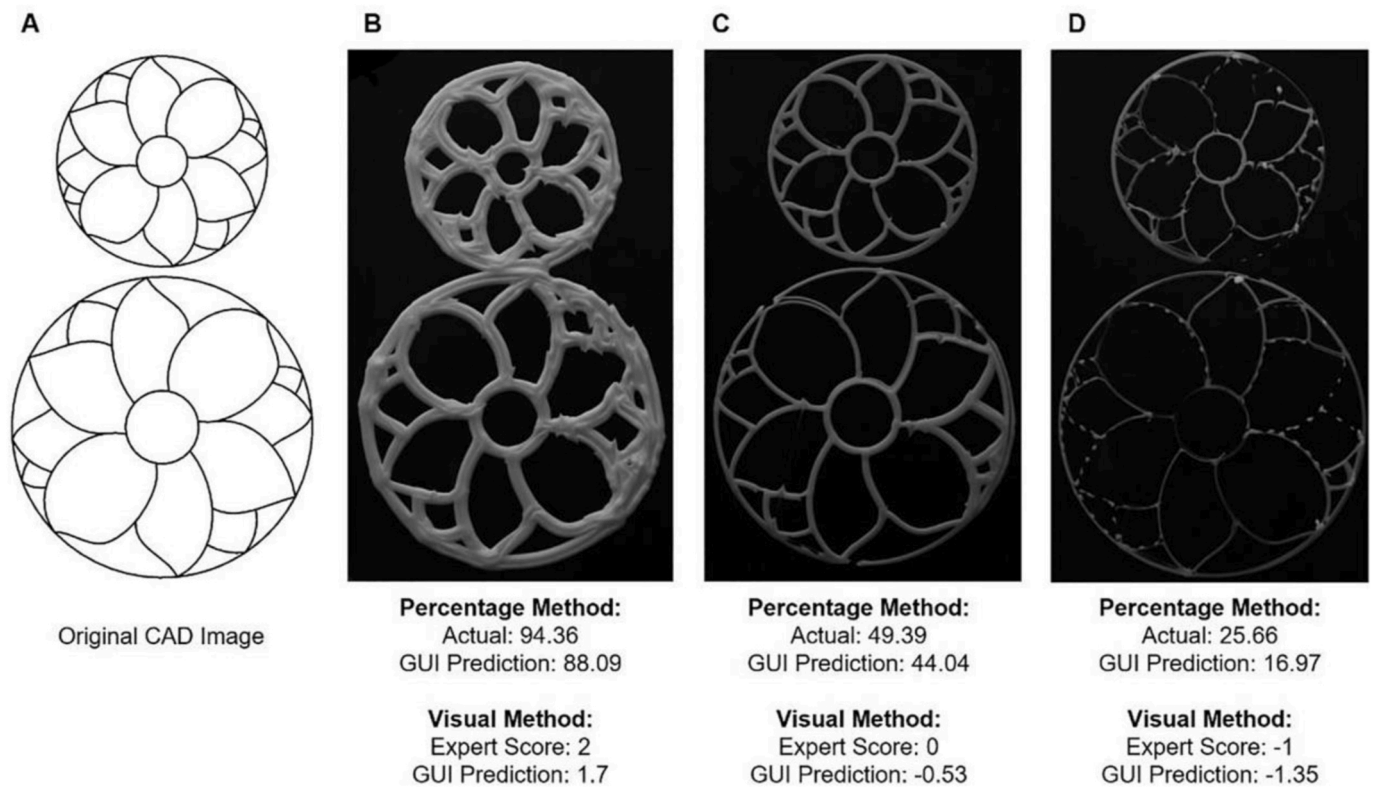


Fig. 19. Images showing the CAD designs (A) and their respective printed samples (B–D) having different print qualities and similarity indices [281].

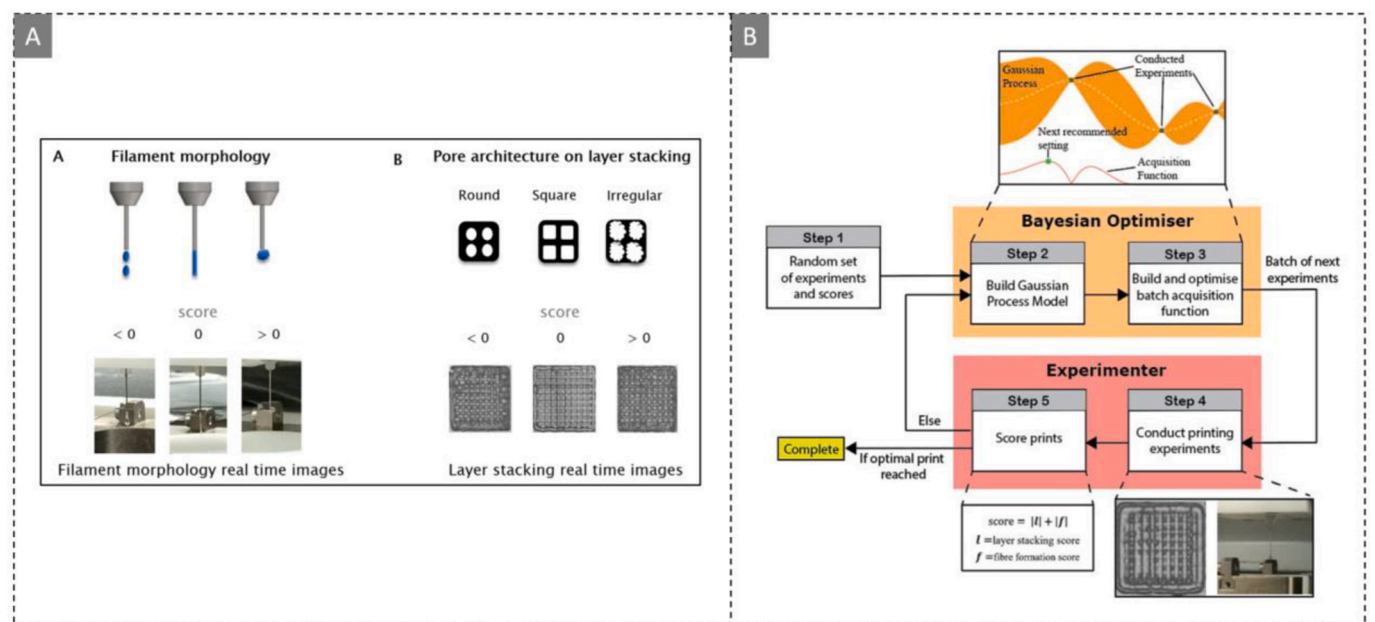
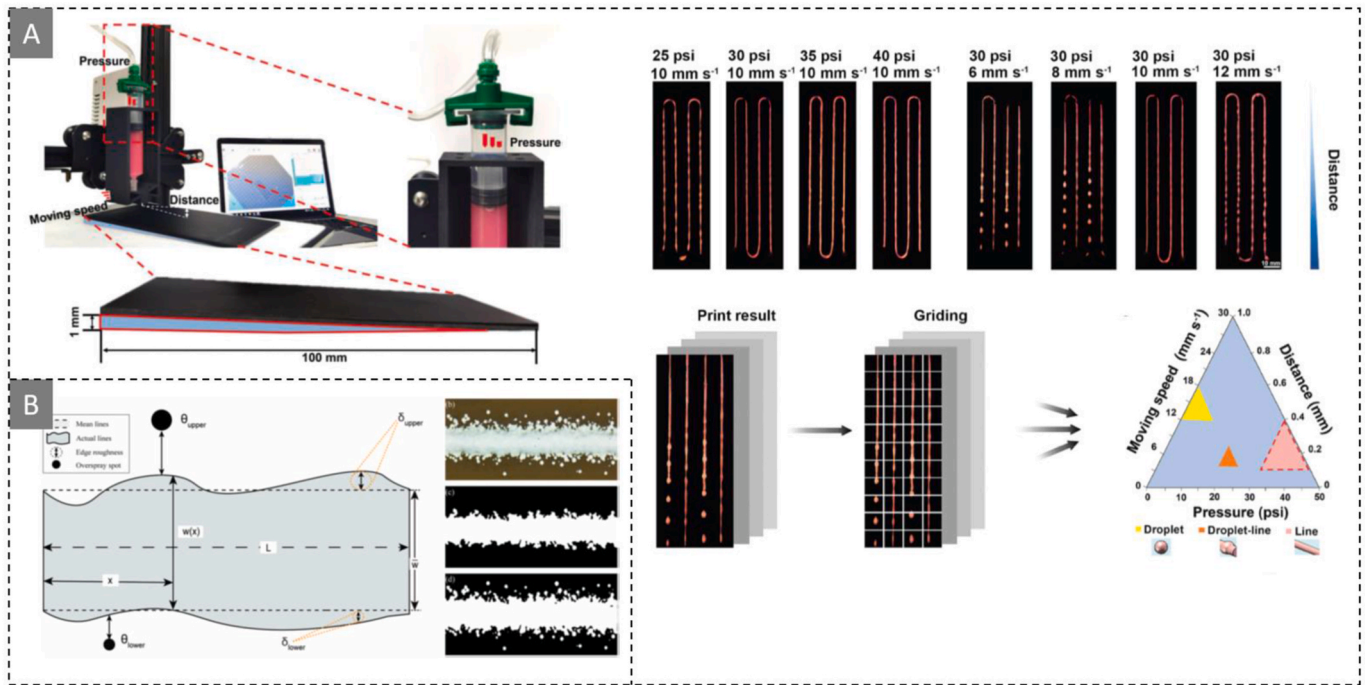


Fig. 20. A: scoring of filament morphology and pore architecture upon stacking, B: machine learning based optimization system for extrusion printing by [253].

parameters is analyzed and the line morphology is optimized. This method does not take into consideration the impact of the ink and substrate properties as it mainly focuses on the line features. Nevertheless, it shows the potential of AI-assisted image analysis in advancing the field of 3D printing parameter optimization.

In the light of the above, it becomes clear that advancing natural materials to meet the requirements of 3D printing and other industry-specific characteristics is crucial to address the growing demand for

highly intricate structures, case-sensitive properties, and rapid production rate. This fast pace of growth can only be achieved by the help of AI due to its ability to deal with large amounts of data in a few seconds. However, due to the complexity of natural materials and other reasons discussed in this review, the progress in this field is developing slowly. Building on the growth rate of AI-assisted models in other fields, the emergence of transfer learning [282], and the horizons and goals that AI-assisted techniques can achieve in the field of natural material



**Fig. 21.** A: AI-based optimization system setup for extrusion printing, examples of printed lines at different conditions, and a printability phase diagram [279] B: features of a printed line using AJP [280].

development for 3D printing, an exponential growth in this field is expected soon.

#### 4. Challenges and future prospects

As discussed, integrating AI into 3D bioprinting holds immense potential to achieve significant advancements, enabling improvements in precision, efficiency, and customization. However, despite these promising developments, several challenges persist that limit the full potential of AI in this domain. These challenges include limited data availability, material property variability, high dependency on multiple factors, etc. Addressing these obstacles requires a multidisciplinary approach that utilizes advanced AI algorithms, collaborative data sharing, and hybrid modeling techniques as is discussed below.

##### 4.1. Property prediction

One of the key challenges in using AI for property prediction in 3D printing is the limited availability of reliable and extensive data on novel biomaterials. Many materials are new and still in the research phase, meaning there is insufficient data for accurate machine learning model training. Additionally, the natural variability of biomaterials leads to inconsistent properties that complicate the prediction process. For example, while methods like linear regression are fast and useful for ranking geometries, they do not predict detailed material properties accurately, highlighting the trade-offs between speed and precision in some AI models [258]. Furthermore, prediction errors in extrusion pressure using random forest and linear regression models emphasize the challenge of achieving high accuracy across diverse biomaterials [260]. To overcome this, there is a need for collaborative data sharing between academia and industry to create large and standardized datasets for training AI models.

Available studies also reveal that some predictions for parameters like plastic viscosity and yield stress are often limited to specific non-biomaterials such as concrete, and still need to be generalized for natural biomaterials [267]. Even in cases where methods like extreme gradient boosting (XGB) excel, challenges remain in extending these

findings to other biomaterials and exploring the influence of additional parameters like nozzle pressure and strut size [259].

Furthermore, the complexity of the material's behavior upon 3D bioprinting, such as viscosity, spreading, and layer adhesion, is not always predictable with existing simulation tools. Future work should focus on integrating hybrid modeling techniques, combining simulation-based models with AI algorithms to enhance prediction accuracy. The continued exploration of AI-driven tools can also enable more dynamic and adaptive predictions, which can even account for changes in material properties during the printing process.

##### 4.2. Smart biomaterial selection

The ability of AI techniques to select the most suitable biomaterials based on desired properties is currently hindered by the lack of extensive datasets on different material combinations, especially for new and unique biomaterials. This limitation affects the scalability of AI models, as highlighted by Gu et al. [258], where the efficiency of material selection for 1 million combinations is possible but constrained by the absence of advanced structural data. While AI methods like neural networks and reinforcement learning have been used to predict suitable material combinations, the challenge of accurate material ranking persists, as many properties are multi-faceted and may conflict. For instance, ANN and CNN models demonstrated by Rojek et al. [270,271] highlighted the demand to incorporate diverse parameters beyond tensile strength to achieve more reliable material rankings for specific applications.

Another issue is the complexity of material behavior in various 3D printing processes, where the material's performance can depend heavily on the specific application and environmental conditions. Yu et al. [272] emphasized that optimizing designs using deep learning methods, while cost-efficient, is complicated by the variability in resolution and size scaling during the design process. To address these challenges, future research should aim to develop AI algorithms that can handle multi-dimensional material data and incorporate environmental and application-specific factors. The creation of open-access material databases could also help standardize material selection, allowing AI

systems to suggest the best material for novel applications with higher reliability and efficiency. However, cybersecurity threats in bioprinting may rise as a new type of challenge [283].

#### 4.3. Parameter optimization

The optimization of 3D printing parameters for biomaterials presents significant challenges due to the inherent variability in material properties and the wide range of customizable parameters across different printing techniques. The dependency of printing outcomes on both material and process parameters makes it difficult to generalize solutions. Additionally, since traditional simulation methods such as FEM struggle to accurately predict the complex behaviors associated with new biomaterials, such as spreading, dynamic viscosity, and interlayer interactions, machine learning offers a promising alternative, yet its data dependency and high computational costs can slow down the process [274]. Moreover, machine learning models used for different printing techniques, such as TPL, WAAM, and PBF, must account for the challenges posed by different parameter requirements for each biomaterial and process [277].

To improve optimization, future work should focus on combining FEM and other simulation methods with machine learning to create more accurate and data-efficient models. For example, machine learning methods, such as hybrid Whale Optimization Algorithm (WOA)-ANN, could help tackle the challenge of parameter optimization by generating large datasets and establishing the relations between inputs and outputs to reduce computational efforts, especially in the optimization of parameters like surface roughness and production time [276]. Moreover, advancing machine learning algorithms to handle large parameter spaces without excessive computation will be essential for real-time optimization.

Furthermore, the development of user-friendly optimization interfaces that require minimal expertise could allow non-specialists to take full advantage of AI-driven parameter optimization. As a further step, integrating robotic automation in experimental workflows, as seen in image-based AI systems for extrusion and aerosol jet printing, could reduce human intervention and accelerate the optimization process, as these systems could autonomously test and adjust parameters [279, 280].

#### 5. Conclusion

The purpose of this review is to provide a guide for selecting suitable biomaterial-based inks for specific applications based on the material's inherent properties, and the recent progress in AI-assisted techniques that can be utilized to enhance their printability. As previous works related to 3D printing bioinks mostly focus on various materials for biomedical applications, this paper discusses only the naturally sourced materials and gives equal importance to other fields in which natural materials can be used as 3D printing inks to achieve significant improvements. Results show that there is a huge need for rapid enhancements in the printable materials that have functional properties suited for use in the biomedical, water, food, energy, and bioplastics industry. There are 20+ natural materials each possessing a unique set of properties that make them promising for many applications, as discussed and analyzed in this study. However, further improvements should be made so that these materials find their way into industrial applications. Since the development process of biomaterial-based inks involves lengthy trial-and-error experiments that waste resources and efforts, machine learning approaches can alternatively be used to perform thousands of trials in a few seconds with minimal human intervention. Findings of this work demonstrate that multiple machine learning and deep learning techniques have the ability to advance the printability of biomaterials by the prediction of their properties, smart selection of the bioink formulations, and optimization of the printing parameters. Training of these techniques can be based on many aspects such as binary scoring of the

printability of experimental trials, image analysis, etc. Results can be in the form of predicted mechanical properties, optimized set of parameters, suggested printability windows, and many more which are discussed in the paper. Overall, in the age of big data, the integration of AI in printable biomaterial development is essential to keep up with the rapid progress of multiple industries that rely on 3D bioprinting while maintaining sustainability. However, this concept requires more efforts from scientists and researchers to enhance the accuracy of the models, minimize the experimental data required for training, and consequently establish standard procedures that can be incorporated into the industry.

#### CRediT authorship contribution statement

**Soumaya Grira:** Writing – original draft, Validation, Investigation, Formal analysis, Data curation, Conceptualization. **Mohammad Sayem Mozumder:** Writing – review & editing, Validation, Supervision, Investigation, Formal analysis, Data curation. **Abdel-Hamid I. Mourad:** Writing – review & editing, Visualization, Validation, Supervision, Project administration, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Mohamad Ramadan:** Writing – review & editing, Validation, Investigation, Formal analysis. **Hadil Abu Khalifeh:** Writing – review & editing, Formal analysis, Data curation. **Mohammad Alkhedher:** Writing – review & editing, Supervision, Formal analysis.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

#### Acknowledgements

ADU author acknowledges the financial support received from the United AI-Saqer Grants Initiative through Project Number 19300842, and UAEU author acknowledges the financial support from UAEU Strategic Research Grant (No. 12R227).

#### Data availability

Data will be made available on request.

#### References

- [1] Y. Zhang, et al., Biomaterials based on marine resources for 3D bioprinting applications, *Mar. Drugs* 17 (10) (Sep. 2019) 555, <https://doi.org/10.3390/md17100555>.
- [2] W. Aljohani, M.W. Ullah, X. Zhang, G. Yang, Bioprinting and its applications in tissue engineering and regenerative medicine, *Int. J. Biol. Macromol.* 107 (Feb. 2018) 261–275, <https://doi.org/10.1016/j.ijbiomac.2017.08.171>.
- [3] M.C. Teixeira, N.S. Lameirinhas, J.P.F. Carvalho, A.J.D. Silvestre, C. Vilela, C.S. R. Freire, A guide to polysaccharide-based hydrogel bioinks for 3D bioprinting applications, *Int. J. Mol. Sci.* 23 (12) (Jun. 2022) 6564, <https://doi.org/10.3390/ijms23126564>.
- [4] S. Grira, H.A. Khalifeh, M. Alkhedher, M. Ramadan, 3D printing algae-based materials: pathway towards 4D bioprinting, *Bioprinting* 33 (Sep. 2023) e00291, <https://doi.org/10.1016/j.bprint.2023.E00291>.
- [5] T.H. Jovic, G. Kungwengwe, A.C. Mills, I.S. Whitaker, Plant-derived biomaterials: a review of 3D bioprinting and biomedical applications, *Front. Mech. Eng.* 5 (Apr. 2019), <https://doi.org/10.3389/fmech.2019.00019>.
- [6] X.B. Chen, et al., Biomaterials/bioinks and extrusion bioprinting, *Bioact. Mater.* 28 (Oct. 2023) 511–536, <https://doi.org/10.1016/j.bioactmat.2023.06.006>.
- [7] C. Benwood, et al., Natural biomaterials and their use as bioinks for printing tissues, *Bioengineering* 8 (2) (Feb. 2021) 27, <https://doi.org/10.3390/bioengineering8020027>.
- [8] J. Qin, et al., Research and application of machine learning for additive manufacturing, *Addit. Manuf.* 52 (Apr. 2022) 102691, <https://doi.org/10.1016/j.addma.2022.102691>.
- [9] D.R. Gunasegaram, et al., Machine learning-assisted in-situ adaptive strategies for the control of defects and anomalies in metal additive manufacturing, *Addit. Manuf.* 81 (Feb. 2024) 104013, <https://doi.org/10.1016/j.addma.2024.104013>.
- [10] L. Ma, S. Yu, X. Xu, S. Moses Amadi, J. Zhang, Z. Wang, Application of artificial intelligence in 3D printing physical organ models, *Mater. Today Bio.* 23 (Dec. 2023) 100792, <https://doi.org/10.1016/j.mtbio.2023.100792>.



- [11] Z. Zhu, D.W.H. Ng, H.S. Park, M.C. McAlpine, 3D-printed multifunctional materials enabled by artificial-intelligence-assisted fabrication technologies, *Nat. Rev. Mater.* 6 (1) (Oct. 2020) 27–47, <https://doi.org/10.1038/s41578-020-00235-2>.
- [12] S. Ramesh, A. Deep, A. Tamayol, A. Kamaraj, C. Mahajan, S. Madhally, Advancing 3D bioprinting through machine learning and artificial intelligence, *Bioprinting* 38 (Apr. 2024) e00331, <https://doi.org/10.1016/j.bprint.2024.e00331>.
- [13] J. Sun, K. Yao, J. An, L. Jing, K. Huang, D. Huang, Machine learning and 3D bioprinting, *Int. J. Bioprint* 9 (4) (Aug. 2024) 717, <https://doi.org/10.18063/ijb.717>.
- [14] S.S. Babu, A.-H.I. Mourad, K.H. Harib, S. Vijayavenkataraman, Recent developments in the application of machine-learning towards accelerated predictive multiscale design and additive manufacturing, *Virtual Phys. Prototyp.* 18 (1) (Dec. 2023), <https://doi.org/10.1080/17452759.2022.2141653>.
- [15] S. Vanaei, S. Vanaei, M. Kanhouou, S. Khelladi, A. Tcharkhtchi, H.R. Vanaei, Importance of machine learning in 3D bioprinting, in: *3D Bioprinting from Lab to Industry*, Wiley, 2024, pp. 391–409, <https://doi.org/10.1002/9781119894407.ch13>.
- [16] H. Chen, B. Zhang, J. Huang, Recent advances and applications of artificial intelligence in 3D bioprinting, *Biophys. Rev.* 5 (3) (Sep. 2024), <https://doi.org/10.1063/5.0190208>.
- [17] J. Yin, M. Yan, Y. Wang, J. Fu, H. Suo, 3D bioprinting of low-concentration cell-laden gelatin methacrylate (GelMA) bioinks with a two-step cross-linking strategy, *ACS Appl. Mater. Interfaces* 10 (8) (Feb. 2018) 6849–6857, <https://doi.org/10.1021/acsami.7b16059>.
- [18] S. Yigit, N. Dinjaski, D.L. Kaplan, Fibrous proteins: at the crossroads of genetic engineering and biotechnological applications, *Biotechnol. Bioeng.* 113 (5) (May 2016) 913–929, <https://doi.org/10.1002/bit.25820>.
- [19] A. Fatimi, O.V. Okoro, D. Podstawczyk, J. Siminska-Stanny, A. Shavandi, Natural hydrogel-based bio-inks for 3D bioprinting in tissue engineering: a review, *Gels* 8 (3) (Mar. 2022) 179, <https://doi.org/10.3390/gels8030179>.
- [20] R. Pugliese, B. Beltrami, S. Regondi, C. Lunetta, Polymeric biomaterials for 3D printing in medicine: an overview, *Annals of 3D Printed Medicine* 2 (Jun. 2021) 100011, <https://doi.org/10.1016/j.jstlm.2021.100011>.
- [21] J. Stepanovska, M. Supova, K. Hanzalek, A. Broz, R. Matejka, Collagen bioinks for bioprinting: a systematic review of hydrogel properties, bioprinting parameters, protocols, and bioprinted structure characteristics, *Biomedicines* 9 (9) (Sep. 2021) 1137, <https://doi.org/10.3390/biomedicines9091137>.
- [22] G. Ying, et al., Aqueous two-phase emulsion bioink-enabled 3D bioprinting of porous hydrogels, *Adv. Mater.* 30 (50) (Dec. 2018), <https://doi.org/10.1002/adma.201805460>.
- [23] A. Veiga, I.V. Silva, M.M. Duarte, A.L. Oliveira, Current trends on protein driven bioinks for 3D printing, *Pharmaceutics* 13 (9) (Sep. 2021) 1444, <https://doi.org/10.3390/pharmaceutics13091444>.
- [24] Y.O. Waidi, I. Kariim, S. Datta, Bioprinting of gelatin-based materials for orthopedic application, *Front. Bioeng. Biotechnol.* 12 (Mar) (2024), <https://doi.org/10.3389/fbioe.2024.1357460>.
- [25] K. Schacht, T. Jüngst, M. Schweinlin, A. Ewald, J. Groll, T. Scheibel, Biofabrication of cell-loaded 3D spider silk constructs, *Angew. Chem. Int. Ed.* 54 (9) (Feb. 2015) 2816–2820, <https://doi.org/10.1002/anie.201409846>.
- [26] X. Cui, et al., 3D printing strategies for precise and functional assembly of silk-based biomaterials, *Engineering* 34 (Mar. 2024) 92–108, <https://doi.org/10.1016/j.eng.2023.09.022>.
- [27] F. Iberite, et al., 3D bioprinting of thermosensitive inks based on gelatin, hyaluronic acid, and fibrinogen: reproducibility and role of printing parameters, *Bioprinting* 39 (Jun. 2024) e00338, <https://doi.org/10.1016/j.bprint.2024.e00338>.
- [28] B.A.G. de Melo, Y.A. Jodot, E.M. Cruz, J.C. Benincasa, S.R. Shin, M. A. Porcionatto, Strategies to use fibrinogen as bioink for 3D bioprinting fibrin-based soft and hard tissues, *Acta Biomater.* 117 (Nov. 2020) 60–76, <https://doi.org/10.1016/j.actbio.2020.09.024>.
- [29] F. Pati, et al., Printing three-dimensional tissue analogues with decellularized extracellular matrix bioink, *Nat. Commun.* 5 (1) (Jun. 2014) 3935, <https://doi.org/10.1038/ncomms4935>.
- [30] S. Chae, et al., 3D cell-printing of gradient multi-tissue interfaces for rotator cuff regeneration, *Bioact. Mater.* 19 (Jan. 2023) 611–625, <https://doi.org/10.1016/j.bioactmat.2022.05.004>.
- [31] L. Elomaa, A. Almalla, E. Keshi, K.H. Hillebrandt, I.M. Sauer, M. Weinhart, Rise of tissue- and species-specific 3D bioprinting based on decellularized extracellular matrix-derived bioinks and bioresins, *Biomater. Biosyst.* 12 (Dec. 2023) 100084, <https://doi.org/10.1016/j.bbiosys.2023.100084>.
- [32] P. De Stefano, et al., Bioprinting of Matrigel scaffolds for cancer research, *Polymers* 13 (12) (Jun. 2021) 2026, <https://doi.org/10.3390/polym13122026>.
- [33] A. Sorkio, et al., Human stem cell based corneal tissue mimicking structures using laser-assisted 3D bioprinting and functional bioinks, *Biomaterials* 171 (Jul. 2018) 57–71, <https://doi.org/10.1016/j.biomaterials.2018.04.034>.
- [34] H. Kwak, S. Shin, H. Lee, J. Hyun, Formation of a keratin layer with silk fibroin-polyethylene glycol composite hydrogel fabricated by digital light processing 3D printing, *J. Ind. Eng. Chem.* 72 (Apr. 2019) 232–240, <https://doi.org/10.1016/j.jiec.2018.12.023>.
- [35] J.K. Placone, et al., Development and characterization of a 3D printed, keratin-based hydrogel, *Ann. Biomed. Eng.* 45 (1) (Jan. 2017) 237–248, <https://doi.org/10.1007/s10439-016-1621-7>.
- [36] S. Salinas-Fernández, M. Santos, M. Alonso, L. Quintanilla, J.C. Rodríguez-Cabello, Genetically engineered elastin-like recombinamers with sequence-based molecular stabilization as advanced bioinks for 3D bioprinting, *Appl. Mater. Today* 18 (Mar. 2020) 100500, <https://doi.org/10.1016/j.apmt.2019.100500>.
- [37] E.O. Osidak, V.I. Kozhukhov, M.S. Osidak, S.P. Domogatsky, Collagen as bioink for bioprinting: a comprehensive review, *Int. J. Bioprint* 6 (3) (Apr. 2020) 17–26, <https://doi.org/10.18063/ijb.v6i3.270>.
- [38] H. Yoon, J.-S. Lee, H. Yim, G. Kim, W. Chun, Development of cell-laden 3D scaffolds for efficient engineered skin substitutes by collagen gelation, *RSC Adv.* 6 (26) (2016) 21439–21447, <https://doi.org/10.1039/C5RA19532B>.
- [39] E.E. Antoine, P.P. Vlachos, M.N. Rylander, Review of collagen I hydrogels for bioengineered tissue microenvironments: characterization of mechanics, structure, and transport, *Tissue Eng., Part B* 20 (6) (Dec. 2014) 683–696, <https://doi.org/10.1089/ten.teb.2014.0086>.
- [40] Y. Koo, G. Kim, An approach for fabricating hierarchically porous cell-laden constructs utilizing a highly porous collagen-bioink, *Adv. Funct. Mater.* 34 (26) (Jun. 2024), <https://doi.org/10.1002/adfm.202316222>.
- [41] A. Zimmerling, Y. Zhou, X. Chen, Synthesis of alginate/collagen bioink for bioprinting respiratory tissue models, *J. Funct. Biomater.* 15 (4) (Apr. 2024) 90, <https://doi.org/10.3390/jfb15040090>.
- [42] X. Lan, et al., Double crosslinked hyaluronic acid and collagen as a potential bioink for cartilage tissue engineering, *Int. J. Biol. Macromol.* 273 (Jul. 2024) 132819, <https://doi.org/10.1016/j.jbiomac.2024.132819>.
- [43] W. Sun, et al., Three-dimensional bioprinting of strontium-modified controlled assembly of collagen polylactic acid composite scaffold for bone repair, *Polymers* 16 (4) (Feb. 2024) 498, <https://doi.org/10.3390/polym16040498>.
- [44] Q. Lihao, et al., 3D bioprinting of Salivianolic acid B-sodium alginate-gelatin skin scaffolds promotes diabetic wound repair via antioxidant, anti-inflammatory, and proangiogenic effects, *Biomed. Pharmacother.* 171 (Feb. 2024) 116168, <https://doi.org/10.1016/j.biopha.2024.116168>.
- [45] K. Abedi, H. Keshvari, M. Solati-Hashjin, Extrusion-based bioprinting: considerations toward gelatin-alginate bioink, *Rapid Prototyp. J.* 30 (6) (Jul. 2024) 1094–1104, <https://doi.org/10.1108/RPJ-06-2023-0207>.
- [46] J.H. Palma, M. Bertuola, É.B. Hermida, Modeling calcium diffusion and crosslinking dynamics in a thermogelling Alginate-Gelatin-Hyaluronic acid ink: 3D bioprinting applications, *Bioprinting* 38 (Apr. 2024) e00329, <https://doi.org/10.1016/j.bprint.2024.e00329>.
- [47] A. Kara Özenler, T. Distler, A.R. Akkineni, F. Tihminlioglu, M. Gelinsky, A. R. Boccacini, 3D bioprinting of mouse pre-osteoblasts and human MSCs using bioinks consisting of gelatin and decellularized bone particles, *Biofabrication* 16 (2) (Apr. 2024) 025027, <https://doi.org/10.1088/1758-5090/ad2c98>.
- [48] F. Bider, et al., 3D bioprinting of multifunctional alginate dialdehyde (ADA)-gelatin (GEL) (ADA-GEL) hydrogels incorporating ferulic acid, *Int. J. Biol. Macromol.* 257 (Feb. 2024) 128449, <https://doi.org/10.1016/j.jbiomac.2023.128449>.
- [49] Y.P. Singh, A. Bandyopadhyay, S. Dey, N. Bhardwaj, B.B. Mandal, Trends and advances in silk based 3D printing/bioprinting towards cartilage tissue engineering and regeneration, *Prog. Biomed. Eng.* 6 (2) (Apr. 2024) 022002, <https://doi.org/10.1088/2516-1091/ad2d59>.
- [50] S.H. Kim, Y.J. Lee, J.S. Lee, O. Ajiteru, O.J. Lee, C.H. Park, Three-dimensional bioprinting using silk biomaterial ink: where we are trying to move?, in: *Silk-Based Biomaterials for Tissue Engineering, Regenerative and Precision Medicine* Elsevier, 2024, pp. 477–509, <https://doi.org/10.1016/B978-0-323-96017-5.00018-2>.
- [51] C. Guo, C. Li, D.L. Kaplan, Enzymatic degradation of *Bombyx mori* silk materials: a review, *Biomacromolecules* 21 (5) (May 2020) 1678–1686, <https://doi.org/10.1021/acs.biomac.0c00090>.
- [52] S.H. Kim, et al., Precisely printable and biocompatible silk fibroin bioink for digital light processing 3D printing, *Nat. Commun.* 9 (1) (Apr. 2018) 1620, <https://doi.org/10.1038/s41467-018-03759-y>.
- [53] J. Lee, et al., Development of silk fibroin-based non-crosslinking thermosensitive bioinks for 3D bioprinting, *Polymers* 15 (17) (Aug. 2023) 3567, <https://doi.org/10.3390/polym15173567>.
- [54] K.H. Schneider, et al., Silk fibroin, gelatin, and human placenta extracellular matrix-based composite hydrogels for 3D bioprinting and soft tissue engineering, *Biomater. Res.* 27 (1) (Feb. 2023), <https://doi.org/10.1186/s40824-023-00431-5>.
- [55] M.S. Hakam, R. Imani, N. Abolfathi, H. Fakhrazadeh, A.M. Sharifi, Evaluation of fibrin-gelatin hydrogel as biopaper for application in skin bioprinting: an in-vitro study, *Bio Med. Mater. Eng.* 27 (6) (Feb. 2017) 669–682, <https://doi.org/10.3233/BME-161617>.
- [56] E. Mazari-Arrighi, et al., Self-organization of long-lasting human endothelial capillary-like networks guided by DLP bioprinting, *Adv. Healthcare Mater.* 13 (14) (Jun. 2024), <https://doi.org/10.1002/adhm.202302830>.
- [57] C. Lee, E. Abelseh, L. de la Vega, S.M. Willerth, Bioprinting a novel glioblastoma tumor model using a fibrin-based bioink for drug screening, *Mater. Today Chem.* 12 (Jun. 2019) 78–84, <https://doi.org/10.1016/j.mtchem.2018.12.005>.
- [58] S. Posniak, J.H.Y. Chung, X. Liu, P. Mukherjee, G.G. Wallace, The importance of elastin and its role in auricular cartilage tissue engineering, *Bioprinting* 32 (Jul. 2023) e00276, <https://doi.org/10.1016/j.bprint.2023.e00276>.
- [59] S. Lee, E.S. Sani, A.R. Spencer, Y. Guan, A.S. Weiss, N. Annabi, Human-recombinant-elastin-based bioinks for 3D bioprinting of vascularized soft tissues, *Adv. Mater.* 32 (45) (Nov. 2020), <https://doi.org/10.1002/adma.202003915>.
- [60] V. Cecuda-Adamczewska, et al., Characterization of a chimeric resilin-elastin structural protein dedicated to 3D bioprinting as a bioink component, *Nanomaterials* 14 (9) (Apr. 2024) 749, <https://doi.org/10.3390/nano14090749>.

- [61] D. Wang, et al., Extrusion bioprinting of elastin-containing bioactive double-network tough hydrogels for complex elastic tissue regeneration, *Aggregate* 5 (3) (Jun. 2024), <https://doi.org/10.1002/agt2.477>.
- [62] K.-F. Yu, et al., Design and synthesis of stem cell-laden keratin/glycol chitosan methacrylate bioinks for 3D bioprinting, *Biomacromolecules* 23 (7) (Jul. 2022) 2814–2826, <https://doi.org/10.1021/acs.biomac.2c00191>.
- [63] M. Chirivi, et al., Biomimetic keratin-coated gold nanoparticles for photo-thermal therapy in a 3D bioprinted glioblastoma tumor model, *Int. J. Mol. Sci.* 23 (17) (Aug. 2022) 9528, <https://doi.org/10.3390/ijms23179528>.
- [64] A. Abaci, M. Guvendiren, Designing decellularized extracellular matrix-based bioinks for 3D bioprinting, *Adv. Healthcare Mater.* 9 (24) (Dec. 2020), <https://doi.org/10.1002/adhm.202000734>.
- [65] M.K. Włodarczyk-Biegun, A. del Campo, 3D bioprinting of structural proteins, *Biomaterials* 134 (Jul. 2017) 180–201, <https://doi.org/10.1016/j.biomaterials.2017.04.019>.
- [66] S. Tavakoli, N. Krishnan, H. Mokhtari, O.P. Oommen, O.P. Varghese, Fine-tuning dynamic cross-linking for enhanced 3D bioprinting of hyaluronic acid hydrogels, *Adv. Funct. Mater.* 34 (4) (Jan. 2024), <https://doi.org/10.1002/adfm.202307040>.
- [67] S. Banerjee, et al., Dextran-based scaffolds for in-situ hydrogelation: use for next generation of bioartificial cardiac tissues, *Carbohydr. Polym.* 262 (Jun. 2021) 117924, <https://doi.org/10.1016/j.carbpol.2021.117924>.
- [68] N. Riahi, B. Liberelle, O. Henry, G. De Crescenzo, Impact of RGD amount in dextran-based hydrogels for cell delivery, *Carbohydr. Polym.* 161 (Apr. 2017) 219–227, <https://doi.org/10.1016/j.carbpol.2017.01.002>.
- [69] Y. Liu, M.B. Chan-Park, Hydrogel based on interpenetrating polymer networks of dextran and gelatin for vascular tissue engineering, *Biomaterials* 30 (2) (Jan. 2009) 196–207, <https://doi.org/10.1016/j.biomaterials.2008.09.041>.
- [70] P. Nikpour, et al., Dextran hydrogels incorporated with bioactive glass-ceramic: nanocomposite scaffolds for bone tissue engineering, *Carbohydr. Polym.* 190 (Jun. 2018) 281–294, <https://doi.org/10.1016/j.carbpol.2018.02.083>.
- [71] K.Y. Lee, D.J. Mooney, Alginate: properties and biomedical applications, *Prog. Polym. Sci.* 37 (1) (Jan. 2012) 106–126, <https://doi.org/10.1016/j.procpolymsci.2011.06.003>.
- [72] E. Axpe, M. Oyen, Applications of alginate-based bioinks in 3D bioprinting, *Int. J. Mol. Sci.* 17 (12) (Nov. 2016) 1976, <https://doi.org/10.3390/ijms17121976>.
- [73] I.P.S. Fernando, W. Lee, E.J. Han, G. Ahn, Alginate-based nanomaterials: fabrication techniques, properties, and applications, *Chem. Eng. J.* 391 (Jul. 2020) 123823, <https://doi.org/10.1016/j.cej.2019.123823>.
- [74] R. Ahmad Raus, W.M.F. Wan Nawawi, R.R. Nasaruddin, Alginate and alginate composites for biomedical applications, *Asian J. Pharm. Sci.* 16 (3) (May 2021) 280–306, <https://doi.org/10.1016/j.ajps.2020.10.001>.
- [75] S.N. Pawar, Chemical modification of alginate, in: *Seaweed Polysaccharides*, Elsevier, 2017, pp. 111–155, <https://doi.org/10.1016/B978-0-12-809816-5.00008-6>.
- [76] Y. Deng, A. Shavandi, O.V. Okoro, L. Nie, Alginate modification via click chemistry for biomedical applications, *Carbohydr. Polym.* 270 (Oct. 2021) 118360, <https://doi.org/10.1016/j.carbpol.2021.118360>.
- [77] S.A. Wilson, L.M. Cross, C.W. Peak, A.K. Gaharwar, Shear-thinning and thermo-reversible nanoengineered inks for 3D bioprinting, *ACS Appl. Mater. Interfaces* 9 (50) (Dec. 2017) 43449–43458, <https://doi.org/10.1021/acsami.7b13602>.
- [78] H. Li, Y.J. Tan, L. Li, A strategy for strong interface bonding by 3D bioprinting of oppositely charged  $\kappa$ -carrageenan and gelatin hydrogels, *Carbohydr. Polym.* 198 (Oct. 2018) 261–269, <https://doi.org/10.1016/j.carbpol.2018.06.081>.
- [79] K. Pal, et al., Selected applications of chitosan composites, *Int. J. Mol. Sci.* 22 (20) (Oct. 2021) 10968, <https://doi.org/10.3390/ijms222010968>.
- [80] J. Venkatesan, S.-K. Kim, Chitosan composites for bone tissue engineering—an overview, *Mar. Drugs* 8 (8) (Aug. 2010) 2252–2266, <https://doi.org/10.3390/md8082252>.
- [81] M.A. Elgadir, Md S. Uddin, S. Ferdosh, A. Adam, A.J.K. Chowdhury, Md Z. I. Sarker, Impact of chitosan composites and chitosan nanoparticle composites on various drug delivery systems: a review, *J. Food Drug Anal.* 23 (4) (Dec. 2015) 619–629, <https://doi.org/10.1016/j.jfda.2014.10.008>.
- [82] W.S. Wan Ngah, L.C. Teong, M.A.K.M. Hanafiah, Adsorption of dyes and heavy metal ions by chitosan composites: a review, *Carbohydr. Polym.* 83 (4) (Feb. 2011) 1446–1456, <https://doi.org/10.1016/j.carbpol.2010.11.004>.
- [83] J.J. Roberts, P.J. Martens, Engineering biosynthetic cell encapsulation systems, in: *Biosynthetic Polymers for Medical Applications*, Elsevier, 2016, pp. 205–239, <https://doi.org/10.1016/B978-1-78242-105-4.00009-2>.
- [84] C. Jiang, Z. Liu, D. Cheng, X. Mao, Agarose degradation for utilization: enzymes, pathways, metabolic engineering methods and products, *Biotechnol. Adv.* 45 (Dec. 2020) 107641, <https://doi.org/10.1016/j.biotechadv.2020.107641>.
- [85] X.T. Fu, S.M. Kim, Agarase: review of major sources, categories, purification method, enzyme characteristics and applications, *Mar. Drugs* 8 (1) (Jan. 2010) 200–218, <https://doi.org/10.3390/md8010200>.
- [86] J.J. Senior, M.E. Cooke, L.M. Grover, A.M. Smith, Fabrication of complex hydrogel structures using suspended layer additive manufacturing (SLAM), *Adv. Funct. Mater.* 29 (49) (Dec. 2019), <https://doi.org/10.1002/adfm.201904845>.
- [87] G. Cidonio, M. Cooke, M. Glinka, J.I. Dawson, L. Grover, R.O.C. Oreffo, Printing bone in a gel: using nanocomposite bioink to print functionalised bone scaffolds, *Mater. Today Bio.* 4 (Sep. 2019) 100028, <https://doi.org/10.1016/j.mtbio.2019.100028>.
- [88] J. Jose, et al., Recent advances in the design and development of bioink formulations for various biomedical applications, *Results Eng.* 22 (Jun. 2024) 102060, <https://doi.org/10.1016/j.rineng.2024.102060>.
- [89] X. Wang, Q. Wang, C. Xu, Nanocellulose-based inks for 3D bioprinting: key aspects in research development and challenging perspectives in applications—a mini review, *Bioengineering* 7 (2) (Apr. 2020) 40, <https://doi.org/10.3390/bioengineering7020040>.
- [90] X. Li, G. Jiang, G. Wang, J. Zhou, Y. Zhang, D. Zhao, Promising cellulose-based functional gels for advanced biomedical applications: a review, *Int. J. Biol. Macromol.* 260 (Mar. 2024) 129600, <https://doi.org/10.1016/j.ijbiomac.2024.129600>.
- [91] B.S. Tabatabaei Hosseini, K. Meadows, V. Gabriel, J. Hu, K. Kim, Biofabrication of cellulose-based hydrogels for advanced wound healing: a special emphasis on 3D bioprinting, *Macromol. Biosci.* 24 (5) (May 2024), <https://doi.org/10.1002/mabi.202300376>.
- [92] S. Hu, et al., An immune regulatory 3D-printed alginate-pectin construct for immunoisolation of insulin producing  $\beta$ -cells, *Mater. Sci. Eng. C* 123 (Apr. 2021) 112009, <https://doi.org/10.1016/j.msec.2021.112009>.
- [93] Q. Wang, et al., Kinetic thermal behavior of nanocellulose filled polylactic acid filament for fused filament fabrication 3D printing, *J. Appl. Polym. Sci.* 137 (7) (Feb. 2020) 48374, <https://doi.org/10.1002/app.48374>.
- [94] T. Ambone, A. Torris, K. Shanmuganathan, Enhancing the mechanical properties of  $\langle \text{scp} \rangle$  3D  $\langle \text{scp} \rangle$  printed polylactic acid using nanocellulose, *Polym. Eng. Sci.* 60 (8) (Aug. 2020) 1842–1855, <https://doi.org/10.1002/pem.25421>.
- [95] L.A.E. Müller, T. Zimmermann, G. Nyström, I. Burgert, G. Siqueira, Mechanical properties tailoring of 3D printed photoreactive nanocellulose composites, *Adv. Funct. Mater.* 30 (35) (Aug. 2020) 2002914, <https://doi.org/10.1002/adfm.202002914>.
- [96] Y. Teramoto, Material development using the inherent features of nano-cellulose and nano-chitin: necessity of simple processes and cross-disciplinary collaboration, *Adv. Powder Technol.* 31 (2) (Feb. 2020) 528–532, <https://doi.org/10.1016/j.apt.2019.11.009>.
- [97] X. Sun, P. Tyagi, S. Agate, M.G. McCord, L.A. Lucia, L. Pal, Highly tunable bioadhesion and optics of 3D printable PNIPAm/cellulose nanofibrils hydrogels, *Carbohydr. Polym.* 234 (Apr. 2020) 115898, <https://doi.org/10.1016/j.carbpol.2020.115898>.
- [98] L.Y. Ee, S.F. Yau Li, Recent advances in 3D printing of nanocellulose: structure, preparation, and application prospects, *Nanoscale Adv.* 3 (5) (2021) 1167–1208, <https://doi.org/10.1039/D0NA00408A>.
- [99] J. Jiang, H. Oguzlu, F. Jiang, 3D printing of lightweight, super-strong yet flexible all-cellulose structure, *Chem. Eng. J.* 405 (Feb. 2021) 126668, <https://doi.org/10.1016/j.cej.2020.126668>.
- [100] J. Leppiniemi, et al., 3D-Printable bioactivated nanocellulose–alginate hydrogels, *ACS Appl. Mater. Interfaces* 9 (26) (Jul. 2017) 21959–21970, <https://doi.org/10.1021/acsami.7b02756>.
- [101] P.G. Gan, S.T. Sam, M.F. bin Abdullah, M.F. Omar, Thermal properties of nanocellulose-reinforced composites: a review, *J. Appl. Polym. Sci.* 137 (11) (Mar. 2020) 48544, <https://doi.org/10.1002/app.48544>.
- [102] S.Y. Sonaye, E.G. Ertugral, C.R. Kothapalli, P. Sikder, Extrusion 3D (Bio)Printing of alginate-gelatin-based composite scaffolds for skeletal muscle tissue engineering, *Materials* 15 (22) (Nov. 2022) 7945, <https://doi.org/10.3390/ma15227945>.
- [103] P.P. Shah, H.B. Shah, K.K. Maniar, T. Özel, Extrusion-based 3D bioprinting of alginate-based tissue constructs, *Procedia CIRP* 95 (2020) 143–148, <https://doi.org/10.1016/j.procir.2020.06.007>.
- [104] K. Christensen, C. Xu, W. Chai, Z. Zhang, J. Fu, Y. Huang, Freeform inkjet printing of cellular structures with bifurcations, *Biotechnol. Bioeng.* 112 (5) (May 2015) 1047–1055, <https://doi.org/10.1002/bit.25501>.
- [105] R. Xiong, Z. Zhang, Y. Huang, Identification of optimal printing conditions for laser printing of alginate tubular constructs, *J. Manuf. Process.* 20 (Oct. 2015) 450–455, <https://doi.org/10.1016/j.jmapro.2015.06.023>.
- [106] J. Yan, Y. Huang, D.B. Chrisey, Laser-assisted printing of alginate long tubes and annular constructs, *Biofabrication* 5 (1) (Nov. 2012) 015002, <https://doi.org/10.1088/1758-5082/5/1/015002>.
- [107] A. Lode, et al., Green bioprinting: fabrication of photosynthetic algae-laden hydrogel scaffolds for biotechnological and medical applications, *Eng. Life Sci.* 15 (2) (Mar. 2015) 177–183, <https://doi.org/10.1002/elsc.201400205>.
- [108] Shengjie Li, Zhuo Xiong, Xiaohong Wang, Yongnian Yan, Haixia Liu, Renji Zhang, Direct fabrication of a hybrid cell/hydrogel construct by a double-nozzle assembling technology, *J. Bioact. Compat. Polym.* 24 (3) (May 2009) 249–265, <https://doi.org/10.1177/0883911509104094>.
- [109] Y. Sun, et al., 3D printing collagen/chitosan scaffold ameliorated axon regeneration and neurological recovery after spinal cord injury, *J. Biomed. Mater. Res.* 107 (9) (Sep. 2019) 1898–1908, <https://doi.org/10.1002/jbm.a.36675>.
- [110] Q. Wu, M. Maire, S. Lerouge, D. Theriault, M.-C. Heuzey, 3D printing of microstructured and stretchable chitosan hydrogel for guided cell growth, *Adv. Biosyst.* 1 (6) (Jun. 2017) 1700058, <https://doi.org/10.1002/adbi.201700058>.
- [111] W.L. Ng, W.Y. Yeong, M.W. Naing, Polyelectrolyte gelatin-chitosan hydrogel optimized for 3D bioprinting in skin tissue engineering, *Int. J. Bioprint* 2 (0) (Jan. 2016), <https://doi.org/10.18063/IJB.2016.01.009>.
- [112] S. Nkanga, et al., Inkjet printing of Chitlac-nanosilver—a method to create functional coatings for non-metallic bone implants, *Biofabrication* 6 (4) (Oct. 2014) 041001, <https://doi.org/10.1088/1758-5082/6/4/041001>.
- [113] F. Fu, et al., Magnetic resonance imaging-three-dimensional printing technology fabricates customized scaffolds for brain tissue engineering, *Neural Regen. Res.* 12 (4) (2017) 614, <https://doi.org/10.4103/1673-5374.205101>.
- [114] V.B. Morris, S. Nimbalkar, M. Younesi, P. McClellan, O. Akkus, Mechanical properties, cytocompatibility and manufacturability of chitosan:PEGDA hybrid-

- gel scaffolds by stereolithography, *Ann. Biomed. Eng.* 45 (1) (Jan. 2017) 286–296, <https://doi.org/10.1007/s10439-016-1643-1>.
- [115] Y.-L. Cheng, F. Chen, Preparation and characterization of photocured poly (ε-caprolactone) diacrylate/poly (ethylene glycol) diacrylate/chitosan for photopolymerization-type 3D printing tissue engineering scaffold application, *Mater. Sci. Eng. C* 81 (Dec. 2017) 66–73, <https://doi.org/10.1016/j.msec.2017.07.025>.
- [116] K.N. Bardakova, et al., 3D printing biodegradable scaffolds with chitosan materials for tissue engineering, *IOP Conf. Ser. Mater. Sci. Eng.* 347 (Apr. 2018) 012009, <https://doi.org/10.1088/1757-899X/347/1/012009>.
- [117] D. Fan, U. Stauffer, A. Accardo, Engineered 3D polymer and hydrogel microenvironments for cell culture applications, *Bioengineering* 6 (4) (Dec. 2019) 113, <https://doi.org/10.3390/bioengineering6040113>.
- [118] W. Qinghua, M. Marion, L. Sophie, T. Daniel, H. Marie-Claude, Solvent-cast 3D printing of Chitosan hydrogel scaffolds for guided cell growth, *Front. Bioeng. Biotechnol.* 4 (2016), <https://doi.org/10.3389/conf.FBIOE.2016.01.00336>.
- [119] S. Rodrigues, M. Dionísio, C.R. López, A. Grenha, Biocompatibility of chitosan carriers with application in drug delivery, *J. Funct. Biomater.* 3 (3) (Sep. 2012) 615–641, <https://doi.org/10.3390/jfb3030615>.
- [120] M. Zubair, M. Arshad, R.A. Pradhan, A. Ullah, Chitosan/chitin-based composites for food packaging applications, in: *Handbook of Chitin and Chitosan*, Elsevier, 2020, pp. 641–670, <https://doi.org/10.1016/B978-0-12-817966-6.00020-0>.
- [121] R. Priyadarshi, J.-W. Rhim, Chitosan-based biodegradable functional films for food packaging applications, *Innovat. Food Sci. Emerg. Technol.* 62 (Jun. 2020) 102346, <https://doi.org/10.1016/j.ifset.2020.102346>.
- [122] M. Rajabi, M. McConnell, J. Cabral, M.A. Ali, Chitosan hydrogels in 3D printing for biomedical applications, *Carbohydr. Polym.* 260 (May 2021) 117768, <https://doi.org/10.1016/j.carbpol.2021.117768>.
- [123] A. Forget, et al., Mechanically tunable bioink for 3D bioprinting of human cells, *Adv. Healthcare Mater.* 6 (20) (Oct. 2017) 1700255, <https://doi.org/10.1002/adhm.201700255>.
- [124] N. Arya, A. Forget, M. Sarem, V.P. Shastri, RGDSP functionalized carboxylated agarose as extrudable carriers for chondrocyte delivery, *Mater. Sci. Eng. C* 99 (Jun. 2019) 103–111, <https://doi.org/10.1016/j.msec.2019.01.080>.
- [125] C. Gong, Z. Kong, X. Wang, The effect of agarose on 3D bioprinting, *Polymers* 13 (22) (Nov. 2021) 4028, <https://doi.org/10.3390/polym13224028>.
- [126] L. Wenger, et al., Systematic evaluation of agarose- and agar-based bioinks for extrusion-based bioprinting of enzymatically active hydrogels, *Front. Bioeng. Biotechnol.* 10 (Nov) (2022), <https://doi.org/10.3389/fbioe.2022.928878>.
- [127] Y. Hu, Y. Kim, I. Hong, M. Kim, S. Jung, Fabrication of flexible pH-responsive agarose/succinoglycan hydrogels for controlled drug release, *Polymers* 13 (13) (Jun. 2021) 2049, <https://doi.org/10.3390/polym13132049>.
- [128] B. Mahendiran, et al., Recent trends in natural polysaccharide based bioinks for multiscale 3D printing in tissue regeneration: a review, *Int. J. Biol. Macromol.* 183 (Jul. 2021) 564–588, <https://doi.org/10.1016/j.ijbiomac.2021.04.179>.
- [129] M.-A. Kamlow, F. Spyropoulos, T. Mills, 3D printing of kappa-carrageenan emulsion gels, *Food Hydrocoll. Health* 1 (2021) 100044, <https://doi.org/10.1016/j.fhfh.2021.100044>.
- [130] C. Kelder, A. Bakker, J. Klein-Nulend, D. Wismeijer, The 3D printing of calcium phosphate with K-carrageenan under conditions permitting the incorporation of biological components—a method, *J. Funct. Biomater.* 9 (4) (Oct. 2018) 57, <https://doi.org/10.3390/jfb9040057>.
- [131] V.A. Zakharova, P.M. Gotovtsev, P.A. Poletaeva, A.G. Rogov, N.R. Kildeeva, 3D printing using iota-carrageenan-based hydrogel with immobilized chlorella vulgaris microalgae, *Nanobiotechnol. Rep.* 18 (1) (Feb. 2023) 91–97, <https://doi.org/10.1134/S2635167623010202>.
- [132] E.-M. Pacheco-Quito, R. Ruiz-Caro, M.-D. Veiga, Carrageenan: drug delivery systems and other biomedical applications, *Mar. Drugs* 18 (11) (Nov. 2020) 583, <https://doi.org/10.3390/md18110583>.
- [133] H.N.P. Vo, C. Chaiwong, L. Zheng, T.M.H. Nguyen, T. Koottatep, T.T. Nguyen, Algae-based biomaterials in 3D printing for applications in medical, environmental remediation, and commercial products, in: *Algae-Based Biomaterials for Sustainable Development*, Elsevier, 2022, pp. 185–202, <https://doi.org/10.1016/B978-0-323-96142-4.00011-7>.
- [134] S.M. Mihaila, A.K. Gaharwar, R.L. Reis, A.P. Marques, M.E. Gomes, A. Khademhosseini, Photocrosslinkable kappa-carrageenan hydrogels for tissue engineering applications, *Adv. Healthcare Mater.* 2 (6) (Jun. 2013) 895–907, <https://doi.org/10.1002/adhm.201200317>.
- [135] H. Mokhtari, et al., Recent advances in chemically-modified and hybrid carrageenan-based platforms for drug delivery, wound healing, and tissue engineering, *Polymers* 13 (11) (May 2021) 1744, <https://doi.org/10.3390/polym13111744>.
- [136] D. Petta, U. D'Amora, L. Ambrosio, D.W. Grijpma, D. Eglin, M. D'Este, Hyaluronic acid as a bioink for extrusion-based 3D printing, *Biofabrication* 12 (3) (May 2020) 032001, <https://doi.org/10.1088/1758-5090/ab8752>.
- [137] T. Lam, et al., Photopolymerizable gelatin and hyaluronic acid for stereolithographic 3D bioprinting of tissue-engineered cartilage, *J. Biomed. Mater. Res. B Appl. Biomater.* 107 (8) (Nov. 2019) 2649–2657, <https://doi.org/10.1002/jbm.b.34354>.
- [138] M.T. Poldervaart, et al., 3D bioprinting of methacrylated hyaluronic acid (MeHA) hydrogel with intrinsic osteogenicity, *PLoS One* 12 (6) (Jun. 2017) e0177628, <https://doi.org/10.1371/journal.pone.0177628>.
- [139] P.R. Turner, E. Murray, C.J. McAdam, M.A. McConnell, J.D. Cabral, Peptide chitosan/dextran core/shell vascularized 3D constructs for wound healing, *ACS Appl. Mater. Interfaces* 12 (29) (Jul. 2020) 32328–32339, <https://doi.org/10.1021/acsami.0c07212>.
- [140] L. Pescosolido, et al., Hyaluronic acid and dextran-based semi-IPN hydrogels as biomaterials for bioprinting, *Biomacromolecules* 12 (5) (May 2011) 1831–1838, <https://doi.org/10.1021/bm200178w>.
- [141] P. Alves, A.F. Simão, M.F.P. Graça, M.J. Mariz, I.J. Correia, P. Ferreira, Dextran-based injectable hydrogel composites for bone regeneration, *Polymers* 15 (23) (Nov. 2023) 4501, <https://doi.org/10.3390/polym15234501>.
- [142] J. Venkatesan, I. Bhatnagar, S.-K. Kim, Chitosan-alginate biocomposite containing fucoidan for bone tissue engineering, *Mar. Drugs* 12 (1) (Jan. 2014) 300–316, <https://doi.org/10.3390/md12010300>.
- [143] D.-H. Ngo, S.-K. Kim, Sulfated polysaccharides as bioactive agents from marine algae, *Int. J. Biol. Macromol.* 62 (Nov. 2013) 70–75, <https://doi.org/10.1016/j.ijbiomac.2013.08.036>.
- [144] T.-S. Vo, S.-K. Kim, Fucoidans as a natural bioactive ingredient for functional foods, *J. Funct. Foods* 5 (1) (Jan. 2013) 16–27, <https://doi.org/10.1016/j.jff.2012.08.007>.
- [145] M. Tako, Rheological characteristics of fucoidan isolated from commercially cultured cladosiphon okamuranus, *Bot. Mar.* 46 (5) (Jan. 2003), <https://doi.org/10.1515/BOT.2003.047>.
- [146] T. Agarwal, M. Costantini, T.K. Maiti, Extrusion 3D printing with Pectin-based ink formulations: recent trends in tissue engineering and food manufacturing, *Biomed. Eng. Adv.* 2 (Dec. 2021) 100018, <https://doi.org/10.1016/j.bea.2021.100018>.
- [147] V. Vancanwenberghe, V. Baiye Mfortaw Mbong, E. Vanstreels, P. Verboven, J. Lammertyn, B. Nicolai, 3D printing of plant tissue for innovative food manufacturing: encapsulation of alive plant cells into pectin-based bio-ink, *J. Food Eng.* 263 (Dec. 2019) 454–464, <https://doi.org/10.1016/j.jfoodeng.2017.12.003>.
- [148] N.S. Bostanci, S. Büyüksungur, N. Nasirci, A. Tezcaner, Potential of pectin for biomedical applications: a comprehensive review, *J. Biomater. Sci. Polym. Ed.* 33 (14) (Sep. 2022) 1866–1900, <https://doi.org/10.1080/09205063.2022.2088525>.
- [149] L. Rong, et al., The application of 3D printing technology on starch-based product: a review, *Trends Food Sci. Technol.* 134 (Apr. 2023) 149–161, <https://doi.org/10.1016/j.tifs.2023.02.015>.
- [150] P. Zhuang, Z. Greenberg, M. He, Biologically enhanced starch bio-ink for promoting 3D cell growth, *Adv. Mater. Technol.* 6 (12) (Dec. 2021), <https://doi.org/10.1002/admt.202100551>.
- [151] H. Yu, S. Chi, D. Li, L. Wang, Y. Wang, Effect of gums on the multi-scale characteristics and 3D printing performance of potato starch gel, *Innovat. Food Sci. Emerg. Technol.* 80 (Aug. 2022) 103102, <https://doi.org/10.1016/j.ifset.2022.103102>.
- [152] H. Strother, R. Moss, M.B. McSweeney, Comparison of <sc>3D</sc> printed and molded carrots produced with gelatin, guar gum and xanthan gum, *J. Texture Stud.* 51 (6) (Dec. 2020) 852–860, <https://doi.org/10.1111/jtxs.12545>.
- [153] R.S.M. Azam, M. Zhang, B. Bhandari, C. Yang, Effect of different gums on features of 3D printed object based on vitamin-D enriched orange concentrate, *Food Biophys.* 13 (3) (Sep. 2018) 250–262, <https://doi.org/10.1007/s11483-018-9531-x>.
- [154] R. Khoeini, et al., Natural and synthetic bioinks for 3D bioprinting, *Adv. Nanobiomed. Res.* 1 (8) (Aug. 2021), <https://doi.org/10.1002/anbr.202000097>.
- [155] L.K. Narayanan, P. Huebner, M.B. Fisher, J.T. Spang, B. Starly, R.A. Shirwaiker, 3D-Bioprinting of polylactic acid (PLA) nanofiber-alginate hydrogel bioink containing human adipose-derived stem cells, *ACS Biomater. Sci. Eng.* 2 (10) (Oct. 2016) 1732–1742, <https://doi.org/10.1021/acsbiomaterials.6b00196>.
- [156] K.C.R. Kolan, J.A. Semon, A.T. Bindbeutel, D.E. Day, M.C. Leu, Bioprinting with bioactive glass loaded polylactic acid composite and human adipose stem cells, *Bioprinting* 18 (Jun. 2020) e00075, <https://doi.org/10.1016/j.bprint.2020.e00075>.
- [157] Y. Wang, et al., 3D printing biocompatible l-Arg/GNPs/PLA nanocomposites with enhanced mechanical property and thermal stability, *J. Mater. Sci.* 55 (12) (Apr. 2020) 5064–5078, <https://doi.org/10.1007/s10853-020-04353-8>.
- [158] J. Żur-Pińska, M.Z. Gładysz, D. Ubels, J. Siebring, M.K. Włodarczyk-Biegun, Smart and sustainable: exploring the future of PHAs biopolymers for 3D printing in tissue engineering, *Sustain. Mater. Technol.* 38 (Dec. 2023) e00750, <https://doi.org/10.1016/j.susmat.2023.e00750>.
- [159] A. Rajabi-Abhari, et al., Diatom bio-silica and cellulose nanofibril for bio-triboelectric nanogenerators and self-powered breath monitoring masks, *ACS Appl. Mater. Interfaces* 13 (1) (Jan. 2021) 219–232, <https://doi.org/10.1021/acsami.0c18227>.
- [160] M. Dobrosielska, et al., Biogenic composite filaments based on polylactide and diatomaceous earth for 3D printing, *Materials* 13 (20) (Oct. 2020) 4632, <https://doi.org/10.3390/ma13204632>.
- [161] R. Han, F. Buchanan, L. Ford, M. Julius, P.J. Walsh, A comparison of the degradation behaviour of 3D printed PDLGA scaffolds incorporating bioglass or biosilica, *Mater. Sci. Eng. C* 120 (Jan. 2021) 111755, <https://doi.org/10.1016/j.msec.2020.111755>.
- [162] R. Han, F. Buchanan, M. Julius, P.J. Walsh, Filament extrusion of bioresorbable PDLGA for additive manufacturing utilising diatom biosilica to inhibit process-induced thermal degradation, *J. Mech. Behav. Biomed. Mater.* 116 (Apr. 2021) 104265, <https://doi.org/10.1016/j.jmbbm.2020.104265>.
- [163] C. Qin, C. Wu, Inorganic biomaterials-based bioinks for three-dimensional bioprinting of regenerative scaffolds, *View* 3 (4) (Jul. 2022), <https://doi.org/10.1002/viw.20210018>.
- [164] D. Qin, L. Sang, Z. Zhang, S. Lai, Y. Zhao, Compression performance and deformation behavior of 3D-printed PLA-based lattice structures, *Polymers* 14 (5) (Mar. 2022) 1062, <https://doi.org/10.3390/polym14051062>.



- [165] E.H. Tümer, H.Y. Erbil, Extrusion-based 3D printing applications of PLA composites: a review, *Coatings* 11 (4) (Mar. 2021) 390, <https://doi.org/10.3390/coatings11040390>.
- [166] M. Mehrpouya, H. Vahabi, M. Barletta, P. Laheurte, V. Langlois, Additive manufacturing of polyhydroxyalkanoates (PHAs) biopolymers: materials, printing techniques, and applications, *Mater. Sci. Eng. C* 127 (Aug. 2021) 112216, <https://doi.org/10.1016/j.msec.2021.112216>.
- [167] J. Tian, R. Zhang, Y. Wu, P. Xue, Additive manufacturing of wood flour/polyhydroxyalkanoates (PHA) fully bio-based composites based on micro-screw extrusion system, *Mater. Des.* 199 (Feb. 2021) 109418, <https://doi.org/10.1016/j.matdes.2020.109418>.
- [168] F. Valentini, A. Dorigato, D. Rigotti, A. Pegoretti, Polyhydroxyalkanoates/fibrillated nanocellulose composites for additive manufacturing, *J. Polym. Environ.* 27 (6) (Jun. 2019) 1333–1341, <https://doi.org/10.1007/s10924-019-01429-8>.
- [169] A.N. Frone, et al., Morpho-structural, thermal and mechanical properties of PLA/PHB/cellulose biodegradable nanocomposites obtained by compression molding, extrusion, and 3D printing, *Nanomaterials* 10 (1) (Dec. 2019) 51, <https://doi.org/10.3390/nano10010051>.
- [170] T.F. Pereira, M.F. Oliveira, I.A. Maia, J.V.L. Silva, M.F. Costa, R.M.S.M. Thiré, 3D printing of poly(3-hydroxybutyrate) porous structures using selective laser sintering, *Macromol. Symp.* 319 (1) (Sep. 2012) 64–73, <https://doi.org/10.1002/masy.201100237>.
- [171] T.F. Pereira, et al., Effect of process parameters on the properties of selective laser sintered Poly(3-hydroxybutyrate) scaffolds for bone tissue engineering, *Virtual Phys. Prototyp.* 7 (4) (Dec. 2012) 275–285, <https://doi.org/10.1080/17452759.2012.738551>.
- [172] B. Duan, M. Wang, Encapsulation and release of biomolecules from Ca-P/PHBV nanocomposite microspheres and three-dimensional scaffolds fabricated by selective laser sintering, *Polym. Degrad. Stabil.* 95 (9) (Sep. 2010) 1655–1664, <https://doi.org/10.1016/j.polydegradstab.2010.05.022>.
- [173] K.W. Meereboer, M. Misra, A.K. Mohanty, Review of recent advances in the biodegradability of polyhydroxyalkanoate (PHA) bioplastics and their composites, *Green Chem.* 22 (17) (2020) 5519–5558, <https://doi.org/10.1039/D0GC01647K>.
- [174] H.-M. Müller, D. Seebach, Poly(hydroxyalkanoates): a fifth class of physiologically important organic biopolymers? *Angew. Chem. Int. Ed. Engl.* 32 (4) (Apr. 1993) 477–502, <https://doi.org/10.1002/ange.199304771>.
- [175] S. Gogolewski, M. Jovanovic, S.M. Perren, J.G. Dillon, M.K. Hughes, Tissue response and in vivo degradation of selected polyhydroxyacids: polylactides (PLA), poly(3-hydroxybutyrate) (PHB), and poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHB/VA), *J. Biomed. Mater. Res.* 27 (9) (Sep. 1993) 1135–1148, <https://doi.org/10.1002/jbm.820270904>.
- [176] J.J. Koh, G.J.H. Lim, X. Zhou, X. Zhang, J. Ding, C. He, 3D-Printed anti-fouling cellulose mesh for highly efficient oil/water separation applications, *ACS Appl. Mater. Interfaces* 11 (14) (Apr. 2019) 13787–13795, <https://doi.org/10.1021/acsami.9b01753>.
- [177] X. Li, H. Shan, W. Zhang, B. Li, 3D printed robust superhydrophilic and underwater superoleophobic composite membrane for high efficient oil/water separation, *Sep. Purif. Technol.* 237 (Apr. 2020) 116324, <https://doi.org/10.1016/j.seppur.2019.116324>.
- [178] S. Ning, G. Tian, F. Yang, Bionic silanized cellulose superhydrophobic paper for efficient oil-water separation, *Sep. Purif. Technol.* 353 (Jan. 2025) 128523, <https://doi.org/10.1016/j.seppur.2024.128523>.
- [179] B. Xu, et al., 3D printing of cellulose nanocrystal-based Pickering foams for removing microplastics, *Sep. Purif. Technol.* 339 (Jul. 2024) 126642, <https://doi.org/10.1016/j.seppur.2024.126642>.
- [180] H. Nasser Abdelhamid, S. Sultan, A.P. Mathew, Binder-free Three-dimensional (3D) printing of Cellulose-ZIF8 (CelloZIF-8) for water treatment and carbon dioxide (CO<sub>2</sub>) adsorption, *Chem. Eng. J.* 468 (Jul. 2023) 143567, <https://doi.org/10.1016/j.cej.2023.143567>.
- [181] S.-S. Park, et al., Facile surface treatment of 3D-printed PLA filter for enhanced graphene oxide doping and effective removal of cationic dyes, *Polymers* 15 (2) (Jan. 2023) 269, <https://doi.org/10.3390/polym15020269>.
- [182] Y. Wang, et al., 3D-Printed filters for efficient heavy metal removal from water using PLA@CS/HAP composites, *Polymers* 15 (20) (Oct. 2023) 4144, <https://doi.org/10.3390/polym15204144>.
- [183] N. Fijol, A. Aguilar-Sánchez, M.-X. Ruiz-Caldas, J. Redlinger-Pohn, A. Mautner, A. P. Mathew, 3D printed polylactic acid (PLA) filters reinforced with polysaccharide nanofibers for metal ions capture and microplastics separation from water, *Chem. Eng. J.* 457 (Feb. 2023) 141153, <https://doi.org/10.1016/j.cej.2022.141153>.
- [184] Y. Majnooni, S.O. Abioye, K. Fayazbakhsh, N. Yousefi, Nano-enabled 3D-printed structures for water treatment, *ACS ES&T Water* 4 (5) (May 2024) 1952–1965, <https://doi.org/10.1021/acsestwater.3c00770>.
- [185] T. Wu, et al., 3D printed porous chitosan/metal-organic framework composites as effective adsorbents to remove heavy metals from wastewater, *Chem. Eng. J.* 493 (Aug. 2024) 152780, <https://doi.org/10.1016/j.cej.2024.152780>.
- [186] M.Y.N. Husna, C.H. Chong, V.-L. Wong, K.H. Cheah, Y.K. Wan, 3D-printed PEGDA monolith with robust silane-grafted chitosan for enhanced textile wastewater treatment, *J. Environ. Chem. Eng.* 10 (6) (Dec. 2022) 108581, <https://doi.org/10.1016/j.jece.2022.108581>.
- [187] R. Jiang, et al., Sustainable chitosan-based materials as heterogeneous catalyst for application in wastewater treatment and water purification: an up-to-date review, *Int. J. Biol. Macromol.* 273 (Jul. 2024) 133043, <https://doi.org/10.1016/j.jbiomac.2024.133043>.
- [188] S. Fuxiang, W. Na, Z. Qiangqiang, W. Jie, L. Bin, 3D printing calcium alginate adsorbents for highly efficient recovery of U(VI) in acidic conditions, *J. Hazard Mater.* 440 (Oct. 2022) 129774, <https://doi.org/10.1016/j.jhazmat.2022.129774>.
- [189] Y. Wu, C. Tang, P. Si, D. Zhang, 3D coaxial printing of porous construct stacked with hollow filaments for heavy metal removal, *Sci. Total Environ.* 887 (Aug. 2023) 164120, <https://doi.org/10.1016/j.scitotenv.2023.164120>.
- [190] J. Luo, A. Ji, G. Xia, L. Liu, J. Yan, Construction of 3D-printed sodium alginate/chitosan/halloysite nanotube composites as adsorbents of Methylene Blue, *Molecules* 29 (7) (Apr. 2024) 1609, <https://doi.org/10.3390/molecules29071609>.
- [191] M. Shahbazi, H. Jäger, S.J. Ahmadi, M. Lacroix, Electron beam crosslinking of alginate/nanoclay ink to improve functional properties of 3D printed hydrogel for removing heavy metal ions, *Carbohydr. Polym.* 240 (Jul. 2020) 116211, <https://doi.org/10.1016/j.carbpol.2020.116211>.
- [192] S.-W. Yoon, et al., 3D-printed Chlorella vulgaris biocarriers: a novel approach to wastewater treatment, *J. Water Proc. Eng.* 57 (Jan. 2024) 104711, <https://doi.org/10.1016/j.jwpe.2023.104711>.
- [193] K. Thakare, L. Jerseth, Z. Pei, B. Tomlin, H. Qin, Three-dimensional printing of hydrogel filters containing algae cells for copper removal from contaminated water, *J. Manuf. Sci. Eng.* 143 (10) (Oct. 2021), <https://doi.org/10.1115/1.4050761>.
- [194] J. Singh, S.K. Mittal, Chlorella sp. based biosensor for selective determination of mercury in presence of silver ions, *Sensor. Actuator. B Chem.* 165 (1) (Apr. 2012) 48–52, <https://doi.org/10.1016/j.snb.2012.02.009>.
- [195] Y. Ferro, M. Perullini, M. Jobbagy, S. Bilmes, C. Durrieu, Development of a biosensor for environmental monitoring based on microalgae immobilized in silica hydrogels, *Sensors* 12 (12) (Dec. 2012) 16879–16891, <https://doi.org/10.3390/s121216879>.
- [196] C. Durrieu, Y. Ferro, M. Perullini, A. Gosset, M. Jobbagy, S.A. Bilmes, Feasibility of using a transcluid inorganic hydrogel to build a biosensor using immobilized algal cells, *Environ. Sci. Pollut. Control Ser.* 23 (1) (Jan. 2016) 9–13, <https://doi.org/10.1007/s11356-015-5023-4>.
- [197] C. He, M. Zhang, Z. Fang, 3D printing of food: pretreatment and post-treatment of materials, *Crit. Rev. Food Sci. Nutr.* 60 (14) (Aug. 2020) 2379–2392, <https://doi.org/10.1080/10408398.2019.1641065>.
- [198] J. Chen, et al., Application of soy protein isolate and hydrocolloids based mixtures as promising food material in 3D food printing, *J. Food Eng.* 261 (Nov. 2019) 76–86, <https://doi.org/10.1016/j.jfoodeng.2019.03.016>.
- [199] V.B.J. Rysenaer, S. Ahmadzadeh, F. Van Bockstaele, A. Ubeyitogullari, An extrusion-based 3D food printing approach for generating alginate-pectin particles, *Curr. Res. Food Sci.* 6 (2023) 100404, <https://doi.org/10.1016/j.cris.2022.11.023>.
- [200] Y. Liu, et al., Effects of sodium alginate and rice variety on the physicochemical characteristics and 3D printing feasibility of rice paste, *Lebensm. Wiss. Technol.* 127 (Jun. 2020) 109360, <https://doi.org/10.1016/j.lwt.2020.109360>.
- [201] L. Qiu, M. Zhang, A.F. Ghazal, Z. Chu, Z. Luo, Development of 3D printed k-carrageenan-based gummy candies modified by fenugreek gum: correlating 3D printing performance with sol-gel transition, *Int. J. Biol. Macromol.* 265 (Apr. 2024) 130865, <https://doi.org/10.1016/j.jbiomac.2024.130865>.
- [202] Y. Wang, et al., Carrageenan addition improves the melting temperature and 3D printing performance of the gelatin system, *Food Hydrocolloids* 144 (Nov. 2023) 108918, <https://doi.org/10.1016/j.foodhyd.2023.108918>.
- [203] S.M. Kim, Y. Wen, H.W. Kim, H.J. Park, Textural and sensory qualities of low-calorie surimi with carrageenan inserted as a protein substitute using coaxial extrusion 3D food printing, *J. Food Eng.* 333 (Nov. 2022) 111141, <https://doi.org/10.1016/j.jfoodeng.2022.111141>.
- [204] H. Tian, K. Wang, H. Lan, Y. Wang, Z. Hu, L. Zhao, Effect of hybrid gelator systems of beeswax-carrageenan-xanthan on rheological properties and printability of litchi inks for 3D food printing, *Food Hydrocolloids* 113 (Apr. 2021) 106482, <https://doi.org/10.1016/j.foodhyd.2020.106482>.
- [205] Y. Li, J. Wang, R. Ying, M. Huang, K. Hayat, Protein-stabilized Pickering emulsion interacting with inulin, xanthan gum and chitosan: rheological behavior and 3D printing, *Carbohydr. Polym.* 326 (Feb. 2024) 121658, <https://doi.org/10.1016/j.carbpol.2023.121658>.
- [206] M. Bian, et al., Fish gelatin and gellan gum mixture as edible ink for 3D printing, *J. Food Eng.* 362 (Feb. 2024) 111762, <https://doi.org/10.1016/j.jfoodeng.2023.111762>.
- [207] N. Carvajal-Mena, G. Tabilo-Munizaga, M. Pérez-Won, R. Lemus-Mondaca, Valorization of salmon industry by-products: evaluation of salmon skin gelatin as a biomaterial suitable for 3D food printing, *Lebensm. Wiss. Technol.* 155 (Feb. 2022) 112931, <https://doi.org/10.1016/j.lwt.2021.112931>.
- [208] J. In, H. Jeong, S. Song, S.C. Min, Determination of material requirements for 3D gel food printing using a fused deposition modeling 3D printer, *Foods* 10 (10) (Sep. 2021) 2272, <https://doi.org/10.3390/foods10102272>.
- [209] J. Zhang, et al., Hot extrusion 3D printing technologies based on starchy food: a review, *Carbohydr. Polym.* 294 (Oct. 2022) 119763, <https://doi.org/10.1016/j.carbpol.2022.119763>.
- [210] B. Yan, et al., 3D food printing curing technology based on gellan gum, *J. Food Eng.* 327 (Aug. 2022) 111036, <https://doi.org/10.1016/j.jfoodeng.2022.111036>.
- [211] Y. Zhong, S. Zeng, Y. Lv, W. Lv, H. Xiao, S. Sheng, Effect of guar gum on the rheological properties, microstructure and 3D printing performance of egg yolk powder-potato starch composite gel, *Food Hydrocolloids* 153 (Aug. 2024) 110018, <https://doi.org/10.1016/j.foodhyd.2024.110018>.
- [212] Y. Liu, D. Liu, G. Wei, Y. Ma, B. Bhandari, P. Zhou, 3D printed milk protein food simulant: improving the printing performance of milk protein concentration by

- incorporating whey protein isolate, *Innovat. Food Sci. Emerg. Technol.* 49 (Oct. 2018) 116–126, <https://doi.org/10.1016/j.ifset.2018.07.018>.
- [213] Y. Zhong, B. Wang, W. Lv, Y. Wu, Y. Lv, S. Sheng, Recent research and applications in lipid-based food and lipid-incorporated bioink for 3D printing, *Food Chem.* 458 (Nov. 2024) 140294, <https://doi.org/10.1016/j.foodchem.2024.140294>.
- [214] K. Schüller, et al., 3D printing of plant-based fat inks towards manufacturing complex cellular agriculture products with fatty structures, *Food Hydrocolloids* 157 (Dec. 2024) 110369, <https://doi.org/10.1016/j.foodhyd.2024.110369>.
- [215] X. Yu, et al., Hydrophilic colloids (Konjac gum/Xanthan gum) in 3D printing of transitional food from fish paste, *Food Hydrocolloids* 137 (Apr. 2023) 108333, <https://doi.org/10.1016/j.foodhyd.2022.108333>.
- [216] C. Zhang, C.-S. Wang, M. Girard, D. Theriault, M.-C. Heuzey, 3D printed protein/polysaccharide food simulant for dysphagia diet: impact of cellulose nanocrystals, *Food Hydrocolloids* 148 (Mar. 2024) 109455, <https://doi.org/10.1016/j.foodhyd.2023.109455>.
- [217] L. Qiu, M. Zhang, B. Adhikari, J. Lin, Z. Luo, Preparation and characterization of 3D printed texture-modified food for the elderly using mung bean protein, rose powder, and flaxseed gum, *J. Food Eng.* 361 (Jan. 2024) 111750, <https://doi.org/10.1016/j.jfoodeng.2023.111750>.
- [218] P. García-Segovia, V. García-Alcaraz, S. Balasch-Parisi, J. Martínez-Monzó, 3D printing of gels based on xanthan/konjac gums, *Innovat. Food Sci. Emerg. Technol.* 64 (Aug. 2020) 102343, <https://doi.org/10.1016/j.ifset.2020.102343>.
- [219] P. Thorakkattu, et al., 3D printing: trends and approaches toward achieving long-term sustainability in the food industry, *Crit. Rev. Biotechnol.* (May 2024) 1–21, <https://doi.org/10.1080/07388551.2024.2344577>.
- [220] Z. Liu, M. Zhang, B. Bhandari, Y. Wang, 3D printing: printing precision and application in food sector, *Trends Food Sci. Technol.* 69 (Nov. 2017) 83–94, <https://doi.org/10.1016/j.tifs.2017.08.018>.
- [221] S.M. Park, H.W. Kim, H.J. Park, Callus-based 3D printing for food exemplified with carrot tissues and its potential for innovative food production, *J. Food Eng.* 271 (Apr. 2020) 109781, <https://doi.org/10.1016/j.jfoodeng.2019.109781>.
- [222] P. Kaur, R. Agrawal, F.M. Pfeiffer, R. Williams, H.B. Bohidar, Hydrogels in agriculture: prospects and challenges, *J. Polym. Environ.* 31 (9) (Sep. 2023) 3701–3718, <https://doi.org/10.1007/s10924-023-02859-1>.
- [223] W. Dissanayake, H. Najaf Zadeh, A. Nazmi, C. Stevens, T. Huber, P. Abhayawardhana, Exploring the potential of 3D-printable agar–urea hydrogels as an efficient method of delivering nitrogen in agricultural applications, *Polysaccharides* 5 (1) (Mar. 2024) 49–66, <https://doi.org/10.3390/polysaccharides5010004>.
- [224] D.J.S. Agron, W.S. Kim, 3D printing technology: role in safeguarding food security, *Anal. Chem.* 96 (11) (Mar. 2024) 4333–4342, <https://doi.org/10.1021/acs.analchem.3c05190>.
- [225] J. Son, H. Kim, Y. Choi, H. Lee, 3D printed energy devices: generation, conversion, and storage, *Microsyst. Nanoeng.* 10 (1) (Jul. 2024) 93, <https://doi.org/10.1038/s41378-024-00708-2>.
- [226] K.N.A. Putri, V. Intasanta, V.P. Hoven, Current significance and future perspective of 3D-printed bio-based polymers for applications in energy conversion and storage system, *Heliyon* 10 (4) (Feb. 2024) e25873, <https://doi.org/10.1016/j.heliyon.2024.e25873>.
- [227] M. Cannio, et al., Smart catalyst deposition by 3D printing for polymer electrolyte membrane fuel cell manufacturing, *Renew. Energy* 163 (Jan. 2021) 414–422, <https://doi.org/10.1016/j.renene.2020.08.064>.
- [228] B. Tyagi, et al., High-performance, large-area semitransparent and tandem perovskite solar cells featuring highly scalable a-ITO/Ag mesh 3D top electrodes, *Nano Energy* 95 (May 2022) 106978, <https://doi.org/10.1016/j.nanoen.2022.106978>.
- [229] S. Grira, M. Alkhedher, H. Abu Khalifeh, M. Ramadan, M. Ghazal, Using algae in Li-ion batteries: a sustainable pathway toward greener energy storage, *Bioresour. Technol.* 394 (Feb. 2024) 130225, <https://doi.org/10.1016/j.biortech.2023.130225>.
- [230] Š. Dermol, B. Borin, D. Gregor-Sveteč, L. Slemenik Perše, G. Lavrič, The development of a bacterial nanocellulose/cationic starch hydrogel for the production of sustainable 3D-printed packaging foils, *Polymers* 16 (11) (May 2024) 1527, <https://doi.org/10.3390/polym16111527>.
- [231] Md T. Hossain, et al., Cellulose and starch-based bioplastics: a review of advances and challenges for sustainability, *Polym. Plast. Technol. Mater.* 63 (10) (Jul. 2024) 1329–1349, <https://doi.org/10.1080/25740881.2024.2329980>.
- [232] C. Zarna, M.T. Opedal, A.T. Echtermeyer, G. Chinga-Carrasco, Reinforcement ability of lignocellulosic components in biocomposites and their 3D printed applications – a review, *Composites Part C: Open Access* 6 (Oct. 2021) 100171, <https://doi.org/10.1016/j.jcomc.2021.100171>.
- [233] S.W.A. Shah, et al., Lignin-based additive materials: a review of current status, challenges, and future perspectives, *Addit. Manuf.* 74 (Jul. 2023) 103711, <https://doi.org/10.1016/j.addma.2023.103711>.
- [234] P.K. Dutta, S. Yadav, G.K. Mehrotra, Modified Chitosan Films/Coatings for Active Food Packaging, 2021, pp. 203–232, <https://doi.org/10.1007/12.2021.84>.
- [235] A. Dey, M.M. Rahman, A. Gupta, N. Yodo, C.W. Lee, A performance study on 3D-printed bioplastic pots from soybean by-products, *Sustainability* 15 (13) (Jul. 2023) 10535, <https://doi.org/10.3390/su151310535>.
- [236] X. Tian, K. Zhao, A. Teng, Y. Li, W. Wang, A rethinking of collagen as tough biomaterials in meat packaging: assembly from native to synthetic, *Crit. Rev. Food Sci. Nutr.* 64 (4) (Feb. 2024) 957–977, <https://doi.org/10.1080/10408398.2022.2111401>.
- [237] J.R.M.H. Alshehhi, et al., 3D-Printable sustainable bioplastics from gluten and keratin, *Gels* 10 (2) (Feb. 2024) 136, <https://doi.org/10.3390/gels10020136>.
- [238] T.M. Joseph, et al., 3D printing of polylactic acid: recent advances and opportunities, *Int. J. Adv. Des. Manuf. Technol.* 125 (3–4) (Mar. 2023) 1015–1035, <https://doi.org/10.1007/s00170-022-10795-y>.
- [239] A. Kovalcik, Recent advances in 3D printing of polyhydroxyalkanoates: a review, *Eurobiotech. J.* 5 (1) (Jan. 2021) 48–55, <https://doi.org/10.2478/ebjt-2021-0008>.
- [240] H. Park, H. He, X. Yan, X. Liu, N.S. Scrutton, G.-Q. Chen, PHA is not just a bioplastic, *Biotechnol. Adv.* (Jan. 2024) 108320, <https://doi.org/10.1016/j.biotechadv.2024.108320>.
- [241] A. Wittek-Krowiak, D. Szopa, B. Anwajler, Advanced packaging techniques—a mini-review of 3D printing potential, *Materials* 17 (12) (Jun. 2024) 2997, <https://doi.org/10.3390/ma17122997>.
- [242] S. Sonkaria, V. Khare, 3D printing of antimicrobial agents for food packaging, in: *Food Packaging and Preservation*, Elsevier, 2024, pp. 153–168, <https://doi.org/10.1016/B978-0-323-90044-7.00009-4>.
- [243] C.T. Tracey, A.L. Predeina, E.F. Krivoschapkina, E. Kumacheva, A 3D printing approach to intelligent food packaging, *Trends Food Sci. Technol.* 127 (Sep. 2022) 87–98, <https://doi.org/10.1016/j.tifs.2022.05.003>.
- [244] Md Z. Al Mahmud, M.H. Mobarak, N. Hossain, Emerging trends in biomaterials for sustainable food packaging: a comprehensive review, *Heliyon* 10 (1) (Jan. 2024) e24122, <https://doi.org/10.1016/j.heliyon.2024.e24122>.
- [245] S.H. Shetty, S. Shetty, C. Singh, A. Rao, Supervised machine learning: algorithms and applications, in: *Fundamentals and Methods of Machine and Deep Learning*, Wiley, 2022, pp. 1–16, <https://doi.org/10.1002/9781119821908.ch1>.
- [246] S. Aggarwal, Machine learning algorithms, perspectives, and real - world application: empirical evidence from United States trade data, *Int. J. Sci. Res.* 12 (3) (Mar. 2023) 292–313, <https://doi.org/10.21275/SR23305084601>.
- [247] C.M. Eckhardt, et al., Unsupervised machine learning methods and emerging applications in healthcare, *Knee Surg. Sports Traumatol. Arthrosc.* 31 (2) (Feb. 2023) 376–381, <https://doi.org/10.1007/s00167-022-07233-7>.
- [248] P. Timsina, J. Liu, O. El-Gayar, Y. Shang, Using semi-supervised learning for the creation of medical systematic review: an exploratory analysis, in: 2016 49th Hawaii International Conference on System Sciences (HICSS), IEEE, Jan. 2016, pp. 1195–1203, <https://doi.org/10.1109/HICSS.2016.151>.
- [249] J.-N. Eckardt, M. Bornhäuser, K. Wendt, J.M. Middeke, Semi-supervised learning in cancer diagnostics, *Front. Oncol.* 12 (Jul) (2022), <https://doi.org/10.3389/fonc.2022.960984>.
- [250] M. Al-Hamadani, M. Fadhel, L. Alzubaidi, B. Harangi, Reinforcement learning algorithms and applications in healthcare and robotics: a comprehensive and systematic review, *Sensors* 24 (8) (Apr. 2024) 2461, <https://doi.org/10.3390/s24082461>.
- [251] J. Lee, S.J. Oh, S.H. An, W.-D. Kim, S.-H. Kim, Machine learning-based design strategy for 3D printable bioink: elastic modulus and yield stress determine printability, *Biofabrication* 12 (3) (May 2020) 035018, <https://doi.org/10.1088/1758-5090/ab8707>.
- [252] J.M. Bone, et al., Hierarchical machine learning for high-fidelity 3D printed biopolymers, *ACS Biomater. Sci. Eng.* 6 (12) (Dec. 2020) 7021–7031, <https://doi.org/10.1021/acsbomaterials.0c00755>.
- [253] K. Ruberu, et al., Coupling machine learning with 3D bioprinting to fast track optimisation of extrusion printing, *Appl. Mater. Today* 22 (Mar. 2021) 100914, <https://doi.org/10.1016/j.apmt.2020.100914>.
- [254] D. Wu, C. Xu, Predictive modeling of droplet formation processes in inkjet-based bioprinting, *J. Manuf. Sci. Eng.* 140 (10) (Oct. 2018), <https://doi.org/10.1115/1.4040619>.
- [255] Z. Zhang, X. Zhou, Y. Fang, Z. Xiong, T. Zhang, AI-driven 3D bioprinting for regenerative medicine: from bench to bedside, *Bioact. Mater.* 45 (Mar. 2025) 201–230, <https://doi.org/10.1016/j.bioactmat.2024.11.021>.
- [256] S. Badini, S. Regondi, R. Pugliese, Enhancing mechanical and bioinspired materials through generative AI approaches, *Materials* 6 (Jan. 2025) 100275, <https://doi.org/10.1016/j.nxmate.2024.100275>.
- [257] A. Sedigh, J.E. Tulipan, M.R. Rivlin, R.E. Tomlinson, Utilizing Q-Learning to Generate 3D Vascular Networks for Bioprinting Bone, Oct. 08, 2020, <https://doi.org/10.1101/2020.10.08.331611>.
- [258] G.X. Gu, C.-T. Chen, M.J. Buehler, De novo composite design based on machine learning algorithm, *Extreme Mech. Lett.* 18 (Jan. 2018) 19–28, <https://doi.org/10.1016/j.eml.2017.10.001>.
- [259] D. Ege, S. Serturk, B. Acarkan, A. Ademoglu, Machine learning models to predict the relationship between printing parameters and tensile strength of 3D Poly (lactic acid) scaffolds for tissue engineering applications, *Biomed. Phys. Eng. Express* 9 (6) (Nov. 2023) 065014, <https://doi.org/10.1088/2057-1976/acf581>.
- [260] S. Tian, R. Stevens, B. McInnes, N. Lewinski, Machine assisted experimentation of extrusion-based bioprinting systems, *Micromachines* 12 (7) (Jun. 2021) 780, <https://doi.org/10.3390/mi12070780>.
- [261] J. Chen, Y. Kong, Q. Huang, Analysis of surimi extrusion behavior during 3D printing by modified Computational Fluid Dynamics (CFD) and quick prediction of printability using machine learning based on texture data, *Innovat. Food Sci. Emerg. Technol.* 94 (Jun. 2024) 103698, <https://doi.org/10.1016/j.ifset.2024.103698>.
- [262] B. Muñoz Castro, et al., Machine learning predicts 3D printing performance of over 900 drug delivery systems, *J. Contr. Release* 337 (Sep. 2021) 530–545, <https://doi.org/10.1016/j.jconrel.2021.07.046>.
- [263] M. Rezapour Sarabi, M.M. Alseid, A.A. Karagoz, S. Tasoglu, Machine learning-enabled prediction of 3D-printed microneedle features, *Biosensors* 12 (7) (Jul. 2022) 491, <https://doi.org/10.3390/bios12070491>.

- [264] H. Chen, Y. Liu, S. Balabani, R. Hirayama, J. Huang, Machine learning in predicting printable biomaterial formulations for direct ink writing, *Research* 6 (Jan) (2023), <https://doi.org/10.34133/research.0197>.
- [265] P. Carou-Senna, et al., Predicting pharmaceutical inkjet printing outcomes using machine learning, *Int. J. Pharm.* X 5 (Dec. 2023) 100181, <https://doi.org/10.1016/j.ijpx.2023.100181>.
- [266] S. Ferdousi, R. Advincula, A.P. Sokolov, W. Choi, Y. Jiang, Investigation of 3D printed lightweight hybrid composites via theoretical modeling and machine learning, *Compos. B Eng.* 265 (Oct. 2023) 110958, <https://doi.org/10.1016/j.compositesb.2023.110958>.
- [267] H. Gao, L. Jin, Y. Chen, Q. Chen, X. Liu, Q. Yu, Rheological behavior of 3D printed concrete: influential factors and printability prediction scheme, *J. Build. Eng.* 91 (Aug. 2024) 109626, <https://doi.org/10.1016/j.jobe.2024.109626>.
- [268] M. Elbadawi, T. Gustaffson, S. Gaisford, A.W. Basit, 3D printing tablets: predicting printability and drug dissolution from rheological data, *Int. J. Pharm.* 590 (Nov. 2020) 119868, <https://doi.org/10.1016/j.ijpharm.2020.119868>.
- [269] A. Menon, B. Póczos, A.W. Feinberg, N.R. Washburn, Optimization of silicone 3D printing with hierarchical machine learning, *3D Print. Addit. Manuf.* 6 (4) (Aug. 2019) 181–189, <https://doi.org/10.1089/3dp.2018.0088>.
- [270] I. Rojek, D. Mikołajewski, P. Kotlarz, K. Tyburek, J. Kopowski, E. Dostatni, Traditional artificial neural networks versus deep learning in optimization of material aspects of 3D printing, *Materials* 14 (24) (Dec. 2021) 7625, <https://doi.org/10.3390/ma14247625>.
- [271] I. Rojek, D. Mikołajewski, E. Dostatni, M. Macko, AI-optimized technological aspects of the material used in 3D printing processes for selected medical applications, *Materials* 13 (23) (Nov. 2020) 5437, <https://doi.org/10.3390/ma13235437>.
- [272] C. Yu, et al., Hierarchical multiresolution design of bioinspired structural composites using progressive reinforcement learning, *Adv. Theory Simul.* 5 (11) (Nov. 2022), <https://doi.org/10.1002/adts.202200459>.
- [273] K. Guo, Z. Yang, C.-H. Yu, M.J. Buehler, Artificial intelligence and machine learning in design of mechanical materials, *Mater. Horiz.* 8 (4) (2021) 1153–1172, <https://doi.org/10.1039/D0MH01451F>.
- [274] M. Piovarchi, et al., Closed-loop control of direct ink writing via reinforcement learning, *ACM Trans. Graph.* 41 (4) (Jul. 2022) 1–10, <https://doi.org/10.1145/3528223.3530144>.
- [275] I. Baturynska, O. Semeniuta, K. Martinsen, Optimization of process parameters for powder bed fusion additive manufacturing by combination of machine learning and finite element method: a conceptual framework, *Procedia CIRP* 67 (2018) 227–232, <https://doi.org/10.1016/j.procir.2017.12.204>.
- [276] P. Kumar, P. Gupta, I. Singh, Parametric optimization of FDM using the ANN-based whale optimization algorithm, *AI EDAM (Artif. Intell. Eng. Des. Anal. Manuf.)* 36 (Aug. 2022) e27, <https://doi.org/10.1017/S0890060422000142>.
- [277] F. Ciccone, A. Bacciaglia, A. Ceruti, Optimization with artificial intelligence in additive manufacturing: a systematic review, *J. Braz. Soc. Mech. Sci. Eng.* 45 (6) (Jun. 2023) 303, <https://doi.org/10.1007/s40430-023-04200-2>.
- [278] J. Shi, J. Song, B. Song, W.F. Lu, Multi-objective optimization design through machine learning for drop-on-demand bioprinting, *Engineering* 5 (3) (Jun. 2019) 586–593, <https://doi.org/10.1016/j.eng.2018.12.009>.
- [279] B. Chen, et al., Artificial intelligence-assisted high-throughput screening of printing conditions of hydrogel architectures for accelerated diabetic wound healing, *Adv. Funct. Mater.* 32 (38) (Sep. 2022), <https://doi.org/10.1002/adfm.202201843>.
- [280] Y. Liu, S. Yin, Z. Liu, H. Zhang, A machine learning framework for process optimization in aerosol jet 3D printing, *Flexi. Printed Electronics* 8 (2) (Jun. 2023) 025017, <https://doi.org/10.1088/2058-8585/acd794>.
- [281] S. Rahmani Dabbagh, O. Ozcan, S. Tasoglu, Machine learning-enabled optimization of extrusion-based 3D printing, *Methods* 206 (Oct. 2022) 27–40, <https://doi.org/10.1016/j.ymeth.2022.08.002>.
- [282] Y. Tang, M. Rahmani Dehaghani, G.G. Wang, Review of transfer learning in modeling additive manufacturing processes, *Addit. Manuf.* 61 (Jan. 2023) 103357, <https://doi.org/10.1016/j.addma.2022.103357>.
- [283] J.C. Isichei, S. Khorsandroo, S. Desai, Cybersecurity and privacy in smart bioprinting, *Bioprinting* 36 (Dec. 2023) e00321, <https://doi.org/10.1016/j.bprint.2023.e00321>.