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Review

3D/4D printing of cellulose nanocrystals-based biomaterials: Additives for sustainable applications

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ABSTRACT

Cellulose nanocrystals (CNCs) have gained significant attraction from both industrial and academic sectors, thanks to their biodegradability, non-toxicity, and renewability with remarkable mechanical characteristics. Desirable mechanical characteristics of CNCs include high stiffness, high strength, excellent flexibility, and large surface-to-volume ratio. Additionally, the mechanical properties of CNCs can be tailored through chemical modifications for high-end applications including tissue engineering, actuating, and biomedical. Modern manufacturing methods including 3D/4D printing are highly advantageous for developing sophisticated and intricate geometries. This review highlights the major developments of additive manufactured CNCs, which promote sustainable solutions across a wide range of applications. Additionally, this contribution also presents current challenges and future research directions of CNC-based composites developed through 3D/4D printing techniques for myriad engineering sectors including tissue engineering, wound healing, wearable electronics, robotics, and anti-counterfeiting applications. Overall, this review will greatly help research scientists from chemistry, materials, biomedicine, and other disciplines to comprehend the underlying principles, mechanical properties, and applications of additively manufactured CNC-based structures.

Abbreviations: 3D, Three-dimensional; 4D, Four-dimensional; AA, Acrylic acid; AAm-AAc, Acrylamide-acrylic acid; BC, Bacterial cellulose; BNC, Bacterial nanocellulose; BMC, Ball milled nanocellulose; CMC, Carboxymethyl cellulose; CMWCNT, Carboxylic multiwall carbon nanotube; CNC, Cellulose nanocrystals; CNCMA, Methacrylated cellulose nanocrystals; CNF, Cellulose nanofibril; CNT, Carbon nanotube; DAC, Dialdehyde cellulose nanocrystals; DES, Deep eutectic solvent; dECM, Decellularized extracellular matrix; DIW, Direct ink writing; DLP, Digital light processing; EC, Ethyl cellulose; EDX, Elemental dispersive X-ray; EOL, End-of-life; FDM, Fused deposition modelling; GelMA, Gelatin methacryloyl; GNRs, Gold nanorods; HAp, Hydroxyapatite; IJP, Inkjet printing; KC, Kappa-carrageenan; LCE, Liquid crystalline elastomer; MA, Methacrylate; MAA, Methacrylic acid; MC, Methylcellulose; MLS, Multi-layered sphere; MP, Methoxy pectin; NP, Nanoparticle; NPES, Polyoxyethylene ether; PCL, Polycaprolactone; PCLA, Poly(ε-caprolactone-*co*-lactide); PDA, Poly(dopamine); PDMS, Poly(dimethylsiloxane); PEG, Polyethylene glycol; PEGDA, Poly(ethylene glycol) diacrylate; PI, Polyimide; PHB, Polyhydroxybutyrate; PHBH, Poly(3-hydroxybutyrate-co-3 hydroxyhexanoate); pHEMA, Poly(2-hydroxyethyl methacrylate); PLA, Polylactic acid; PLGA, Poly(lactide-*co*-glycolide); PNVCL, Poly (N-vinyl caprolactam); PAM, Poly (acrylamide); PNIPAM, Poly(N-isopropylacrylamide); PSA, Polymerized stearyl acrylate; PU, Polyurethane; PUA, Polyurethane acrylate; PVA, Polyvinyl alcohol; Py-PNGs, Pyro- and piezoelectric nanogenerators; SA, Sodium alginate; SEM, Scanning electron microscope; SLA, Stereolithography; SLS, Selective laser sintering; SMA, Shape memory alloy; SMP, Shape memory polymer; SMG, Shape memory gel; TA, Tannic acid; TEM, Transmission electron microscopy; TOCN, Tempo-oxidized cellulose nanofibril; TPP, Two-photon polymerization; UV, Ultraviolet; XG, Xanthan gum.

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1. Introduction

The additive manufacturing (AM), also known as the three-dimensional (3D) printing, is an emerging technology, which has gained tremendous attraction in diverse areas including biomedicine, automotive, aerospace, and structural applications [1]. This technology involves the digital designing of 3D objects with precise dimensions through a spatial or layer-by-layer deposition [2]. It eliminates the need for dies, molds, or lithographic masks. These modern manufacturing techniques can produce cost-effective, on-demand, and customized products with high structural complexities by using a variety of materials such as metals, alloys, polymers, and ceramics [3–5]. Owing to these distinct features, the AM is considered as the next-generation manufacturing technology. 3D printing is further classified into five main techniques; (i) Extrusion-based printing, which includes fused deposition modelling (FDM) and direct ink writing (DIW), (ii) Photo-polymerization or vat-photopolymerization that incorporates digital light processing (DLP) and stereolithography (SLA), (iii) Powder-based printing like selective laser sintering (SLS), (iv) Binder jetting, (v) Material jetting or inkjet printing (IJP) [6–9].

With the rapid progress in the AM and the prevalence of eco-friendly, natural, and sustainable materials, the 3D printing of smart materials has gained immense interest around the world and exhibits a huge market potential in near future [10–12]. Fig. 1 shows the market values and shares of AM-based various products in the last decade. In 2013, Prof. H. Jerry introduced the neologism "four-dimensional (4D) printing" during a technology, entertainment, design (TED) talk [13]. This printing is highly suitable to develop smart products for intricate and dynamic environments. It involves the development of 3D-printed structures, which respond to external stimuli such as heat, water, pH, light, moisture, magnetic and electric field in a predictable and controlled manner [14]. Materials that are responsive to stimuli are known as smart materials. These materials possess the ability to change shape on a macro to micro scale under a stimulant environment, as well as add functionalities to the printed materials for various novel applications such as soft robotics [15], biomedical [16–18], and self-deployable structures [19].

4D printing is a fast-growing, challenging, and innovative research area that is attracting academics and industries over the past few years [21–23]. Structures, devices, and various items developed through 4D printing are often considered smart and flexible, as smart materials change their morphologies in a controllable and programmed way over time [24]. Furthermore, this technology offers various advantages over traditional manufacturing routes, such as unrestricted and complex design, faster production, low manufacturing cost and waste, and the one-step manufacturing process which include multiple materials combined into a single object [25–27]. Smart materials include polymers, ceramics, and alloys. Out of these materials, shape memory polymers (SMPs) are promising materials for 4D printing, thanks to their excellent ability to recover their original shape from deformed shapes, under external stimulations [28]. To date, various SMPs like polylactic acid (PLA), polycaprolactone (PCL), polyethylene glycol (PEG), etc. are used for different engineering applications due to their excellent mechanical and functional properties [29].

1.1. 3D/4D printing of sustainable materials

The emergence of synthetic polymers and their composites is highly promising from an AM perspective [30]. Apart from the fact these materials are relatively expensive and the extensive use of synthetic polymers creates harmful effects in the environment, as they are composed of thermosets plastics which do not degrade easily even at their end-of-life (EOL), thus, creating soil, water and air pollution [31–33]. Therefore, there is an urgent need to replace synthetic materials for protecting the environment as well as a potential source for 3D/4D printing materials that can demonstrate excellent shape morphing behaviour under naturally existing stimuli, to maximize the consumption of sustainable materials, which promotes an eco-friendly environment [34]. According to global decarbonization, the development of energy-efficient technologies is need to be in more practice and regularized [35–37]. AM technology consumes a low amount of energy in comparison to traditional manufacturing techniques [38–40]. 4D printing is developed to take more advantage of 3D printing techniques by regulating the printed structures under external stimuli [41–43].

Naturally available polymers such as cellulose, silk, starch, keratin, chitosan, and gelatin are promising materials for various engineering applications such as construction, pharmaceutics, flexible devices, and wearable electronics, due to their environmental sustainability and intrinsic biocompatibility [44–46]. The booming development of 4D-printed biopolymeric materials caught the attention of researchers for

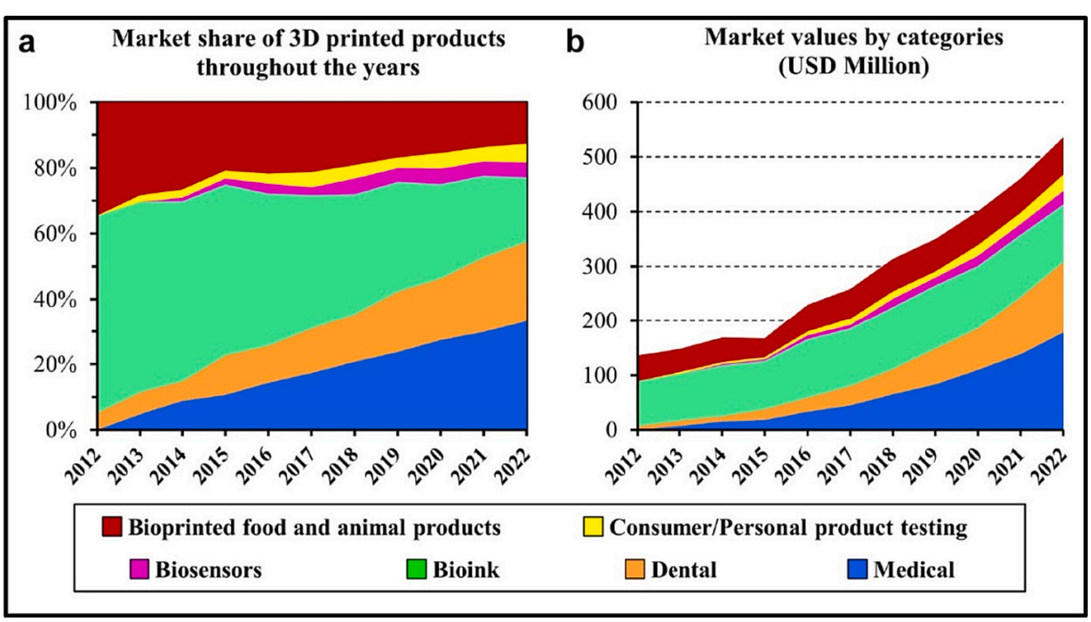

Fig. 1. (a) Market share; (b) Market value of 3D-printed various products across different sectors from 2012 to 2022.
(Adapted from [20], under the Creative Commons Attribution license).

producing highly promising soft bioelectronic devices and intriguing structures for many engineering applications [47–49], as highlighted in Fig. 2. It is worth mentioning that cellulose-based composites have become promising candidate materials for developing and promoting a green environment [50].

Cellulose is a renewable and sustainable polymer widely available in nature and is usually produced from algae, plants, bacteria, and fungi. The world is using cellulose as a chemical raw material for >150 years [51]. Cellulose-based materials have been extensively applied in myriad engineering sectors including the building, biomedical, automobile, textile, electronics, food packaging, and energy industries, thanks to their sustainable, renewable, biocompatible, biodegradable, and toxic-free nature. Different cellulosic materials such as regenerated cellulose, wood pulp, etc. have been extensively applied to develop sustainable composites [52,53]. In the contemporary era, cellulose and its derivatives are employed for formulating hydrogels and stimuli-responsive materials through AM [54]. The rapid progress in the nanotechnology field diverted attention towards sustainable nano-cellulosic materials including cellulose nanocrystals (CNCs), cellulose nanofibrils (CNFs), and bacterial cellulose (BC). These nanomaterials are considered environmentally benign, cost-effective, and sustainable with extraordinary strength-to-weight ratio. In recent years, different combinations of nanocellulose have been employed to develop composites to make them distinctive biomaterials [55].

1.2. Scope of review

Although a significant number of reviews on topics “3D/4D printing” and “cellulose materials” have been published in the last decade, those reviews are usually focused on the understanding of 4D printing [56–59], smart materials [60–62], stimuli mechanisms [63], and their emerging applications. Furthermore, numerous reviews exist on the 3D printing of cellulosic materials, which contains their novel applications [64]. A few review articles have also elucidated the 4D printing of naturally available materials [65,66]. For instance, Muthe et al. [67] recently published a review article on understanding the role of naturally available materials from the 3D/4D printing perspective. Although, Gauss et al. [68] published a review article on cellulose-based biocomposites and comprehensively summarized the recent advances in 3D/4D printing technologies using cellulosic materials. Key aspects related to their functional requirements for the development of smart and responsive structures were also analyzed. However, the literature lacks a detailed information about 3D/4D printing of CNC-based biomaterials. The current review focuses on the utilization of CNCs in 3D/4D printing, and major developments in this field. This contribution also provides systematic discussions on 4D-printed CNC-based structures, and their applications in biomedical, smart flexible electronics, and soft robotics. Thus, the proposed review can be a reference for future research on understanding the basic concepts of 3D/4D printing for CNCs. Finally, this review envisages future directions for promoting smart and sustainable manufacturing by highlighting the key obstacles

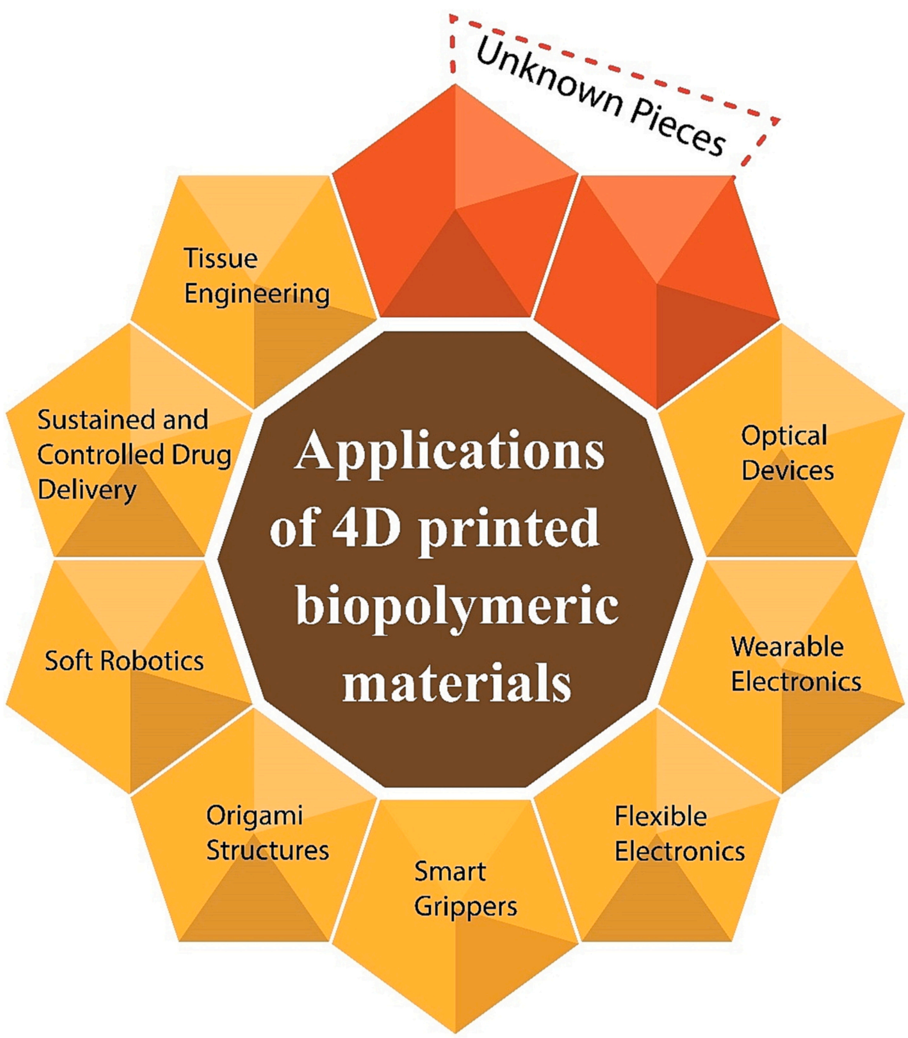

Fig. 2. Pieces of the flower petal revealing potential applications of 4D-printed sustainable polymer materials. Many pieces remain unknown.

and possible remedies for the proposed research field.

2. Cellulosic materials

The total annual biomass production of cellulose is 1.5×10^{12} tonnes, which mainly originates from agricultural residue, wood, algae, tunicate, and bacteria [69,70]. Cellulosic materials are mainly applied to produce paper and pulp. Additionally, the incorporation of cellulose into novel materials like polymers, hydrogels, and nanocomposites has also been evaluated [71]. This section provides a brief overview of cellulose along with an in-depth account about nanocellulose materials, especially CNCs.

Cellulose is a linear carbohydrate polymer of glucose units linked with β-1,4 glycosidic bonds. It is the main structural component of plant cell walls and is considered a polysaccharide due to the sugar monomers. Highly ordered cellulose chains generate cellulose nanocrystalline structures [72]. The combination of cellulose nanocrystalline and disordered cellulose is ingrained in a matrix, which consists of lignin, hemicellulose, and a minuscule amount of pectin, thus, developing a hierarchical structure, as illustrated in Fig. 3. The complex composition and hierarchical structure of plant cell walls permit the development of sustainable products with extraordinary mechanical properties [73–75].

It is a porous polysaccharide with unique properties such as abundant availability, biodegradability, lightweight, low density, high porosity, eco-friendliness, and easy functionalization. As cellulose belongs to the family of natural photonics and recently, it has piqued the interest of researchers, due to its potential in mitigating environmental concerns and can be used as a green alternative to petroleum-based products [78–80].

Cellulose derivatives can be used as alternatives to pure cellulose, thanks to their better dissolution. Furthermore, these materials can be transformed to cellulose through aqueous or ethanolic hydrolysis [81]. Table 1 incorporates different cellulose derivatives, their major sources, and their usual applications. Some cellulose derivatives such as hydroxypropyl methylcellulose, and carboxymethyl cellulose (CMC) exhibit poor water resistance and thus limited applications underwater [82–84]. CNC and CNFs are highly diverse materials, with lengths of 100–600 nm and diameters of ~2 to 20 nm, whereas microfibrillated cellulose (MFC) has a diameter from nano- to micro range, with length typically >1 μm [85–87].

Nanocellulose is nano-scaled material separated from plant cell walls, bacteria, or cotton linters by using high-speed homogenization, acid hydrolysis, and bacterial process [112]. These nanomaterials are extracted from natural cellulosic fibers and contain diameter in nanometre range and length of a few microns. Nanocellulose has been explored widely, thanks to its excellent mechanical properties, biodegradability, reinforcing properties, and low density [113]. Different types of nanocellulose are CNC, CNF, and bacterial nanocellulose (BNC), which are isolated from a variety of cellulose sources through a top-down approach [114–116].

2.1. Cellulose nanocrystals

Cellulose nanocrystals (CNCs) are stiff rod-like nanoparticles (NPs), known as cellulose whiskers, and are extracted from agricultural biomass and plants through a combination of mechanical and chemical treatments [117–119]. CNC has typically the length of 100–3000 nm, diameter of 3–50 nm and aspect ratio vary 5–200, mainly depends on the biological source and isolation methodology [120]. These nanomaterials are the derivatives of a variety of cellulose and are isolated through microbial or enzymatic digestion, nonchemical fragmentation, acid hydrolysis, or other top-down fabrication techniques [121]. Strong acid

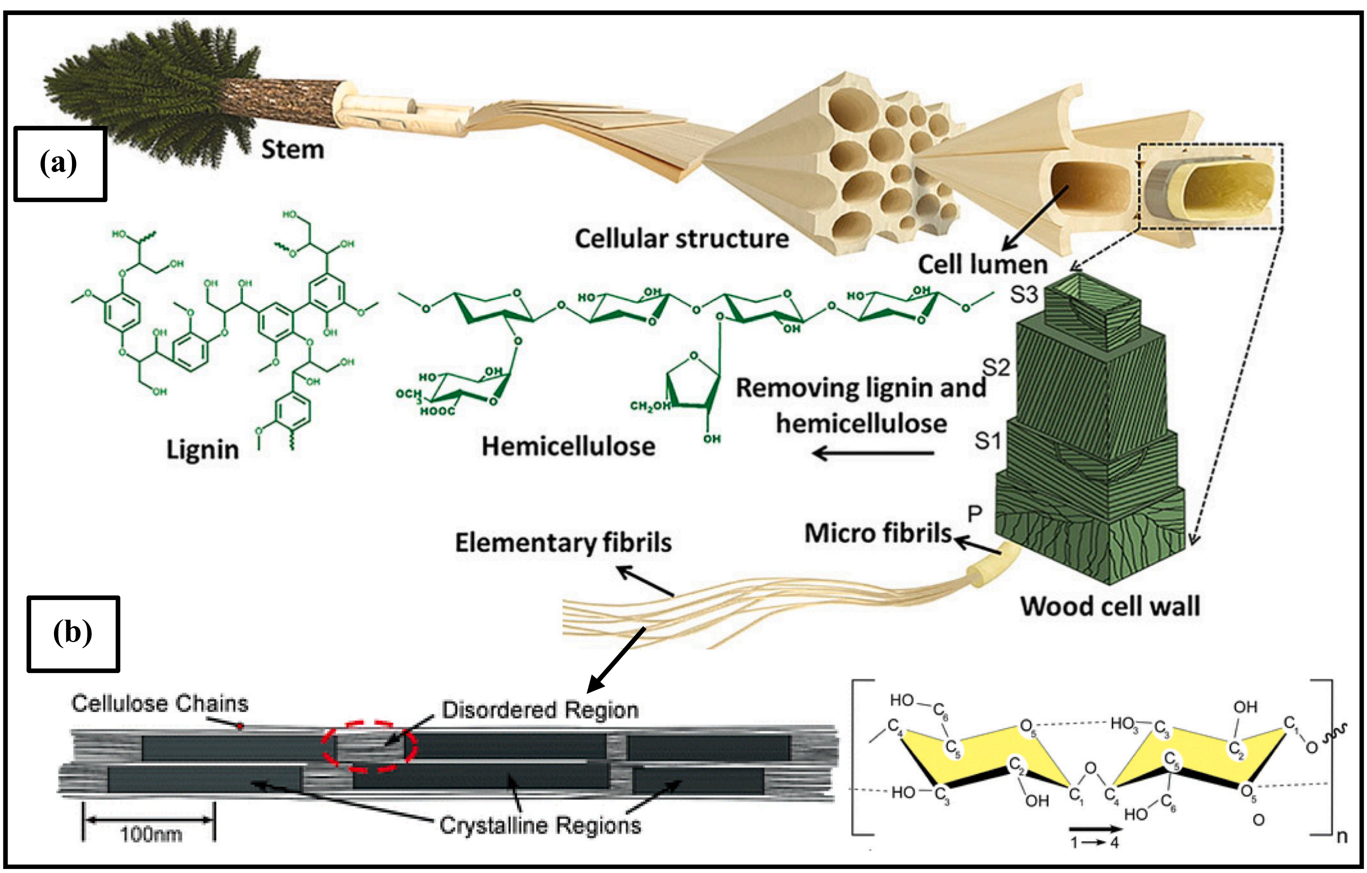

Fig. 3. (a) Schematic representation showing the hierarchical structure of a wood (adapted with permission from [76], copyright 2020, Wiley VCH GmbH); (b) CNCs and amorphous regions (disordered) of fibrils composed of cellulose chain unit (adapted from [77] copyright 2011, the Royal Society of Chemistry).

Table 1
Cellulose derivatives, their major sources, and common applications.

Cellulose derivatives	Subtypes	Major source	Common applications	Ref.
Nanocellulose	BC	*Komagataeibacter xylinus, Achromobacter, Aerobacter, Pseudomonas, and Rhizobium*	Wound healing	[88]
	CNF	Wood and plants	Food industry, biomedical sector and drug deliver	[89,90]
	CNC	Wood or plant pulps	Drug delivery, food industry, and tissue engineering over biosensors and biomedical sector.	[91–93]
Cellulose microfibrils	–	Wood or plant pulps	Wound healing and biomedical devices	[94–96]
Algal cellulose	–	*Posidonia oceanica*, (Cladophora) and (Gelidium elegans)	Tissue engineering	[97]
Tunicate cellulose	–	Sea living animals	Biomedical applications	[98]
Cellulose gel from corn cub	–	Plants	Food and cosmetics	[99]
Cellulose fibers	–	Woods and plants	Biomedical applications	[100–102]
Cellulose ether	Methyl cellulose CMC Ethyl cellulose Hydroxy-propyl methyl cellulose Benzyl cellulose Allylethyl cellulose Benzylhydroxyethyl cellulose	Applying some chemical modification on naturally derived cellulose from various sources such as wood pulp, plants, and various fibers	Food industry, textile industries, self-cleaning materials, batteries, antibacterial materials, porous materials, biomedical applications	[103–107]
Cellulose ester	Sulfate cellulose	Obtained through heterogeneous, homogenous, or quasi-homogenous sulfation procedures	Tissue Engineering	[108]
	Cellulose acetated	Wood pulp	Drug delivery	[109–111]

hydrolysis is the most prominent method applied for extracting CNCs from cellulose. Fig. 4 depicts the needle-like CNCs extracted from the cell plant wall through acid hydrolysis. The yielding of CNCs is dependent on the optimum temperature of 60–70 °C and acid concentration of 56–60 wt%. However, it is inconvenient to extract CNCs through this method, due to the difficulty in the treatment of acid waste [122].

These sustainable nanomaterials can be surface-modified with other functional groups to obtain specific functionalities. Geometrical dimensions of CNCs depend upon the starting cellulosic materials and processing conditions [124]. Table 2 summarizes the extraction of CNC from different natural sources through different methods along with their functional properties and dimensions from the most recent studies.

Cellulose nanocrystals are highly suitable for engineering applications like drug delivery, tissue scaffolds, wound dressings, packaging films, electronics, and wastewater treatment, thanks to its high young's modulus (150 GPa), low density, excellent tensile strength (10 GPa), excellent colloidal stability, high surface area (~250 m^2/g), optical transparency, biocompatibility, and biodegradability [142]. Furthermore, CNCs possess the ability to self-assemble into chiral-nematic crystalline phase. Additionally, CNCs possess surface charge, and amphiphilic properties, and can be used to modify the electrical, optical, and rheological properties of polymers. They do not swell in the humid environment due to hydrophilic surfaces [143–145].

2.1.1. CNC-based composites

CNC-based composites are fabricated by using CNCs and polymers like alginate, chitosan, PLA, PCL, etc. The incorporation of CNCs in polymer matrices significantly improved the thermal resistance, biodegradability, barrier properties, and mechanical properties, without detracting from the materials sustainability. Recently, CNCs have been widely explored as reinforcing elements in smart as well as novel bio-composites [146]. These CNC-based composites have tremendous potential in tissue engineering, drug delivery, wound dressing, bone replacement, bio-sensing, and stimuli-responsive optics, thanks to their superior characteristics [147]. For instance, Voronova et al. [148] prepared polyacrylamide/CNC composites. Results showed that the

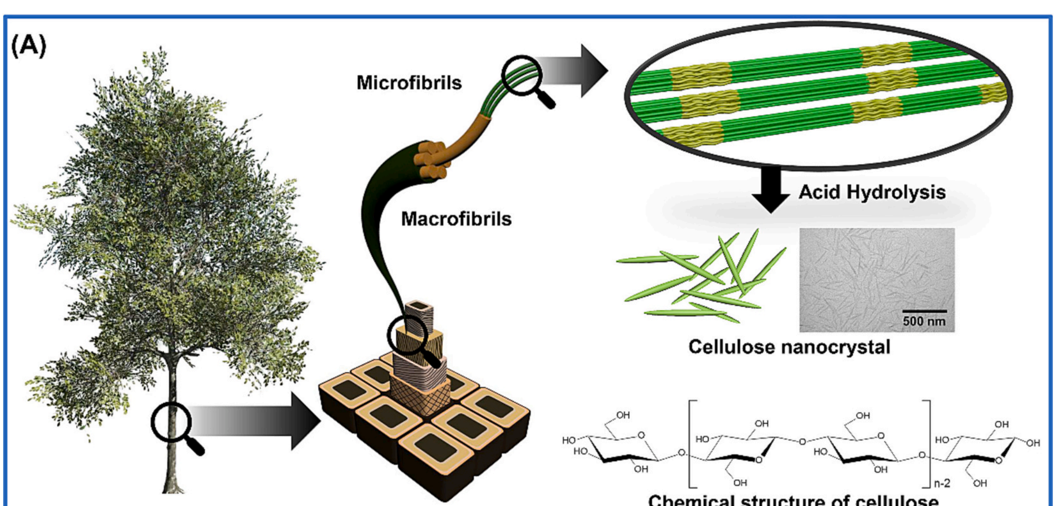

Fig. 4. Schematic representation of CNCs extraction through chemical treatment and TEM image of as-generated CNCs.
(Adapted with permission from [123], copyright 2022, Elsevier Ltd.).

Table 2
Summary of recent works on extraction of CNC from different natural sources through different methods along with their functional properties and dimensions.

Natural resource	Method	Dimensions of CNC			Functional characteristics of prepared CNC	Ref.
		Diameter/ width (nm)	Aspect ratio	Length (nm)		
Rice straw	Sulfuric acid hydrolysis	5.76	–	113	The proposed strategy is highly adapted for producing high-quality CNC from various biomass saccharification without any additional surface treatment.	[125]
Wheat bran	Sulfuric acid hydrolysis	30.58 ± 7.42	25.91 ± 9.93	678.58 ± 92.76	CNC improved the storage stability of protein film.	[126]
Raw Napier grass stems	Sulfuric acid hydrolysis	11.5 ± 2.6	–	156.6 ± 59.6	Triethoxyvinylsilane-modified CNC is considered a potential reinforcing filler for natural rubber-based biocomposites.	[127]
Artemisia annua Stems	Sulfuric acid, phosphoric acid, and hydrochloric acid/ citric acid hydrolysis	4.7–7.2	–	384–504	The prepared CNC have high crystallinity, uniform rod-shaped with remarkable thermal stability and high crystallinity, especially for those CNC obtained from sulfuric acid hydrolysis.	[128]
Kraft pulp	Sulfuric acid hydrolysis	–	–	130–220	CNC average particle size ranged between 160 and 170 nm, and cellulosic solid residues were roughly 270 nm.	[129]
Waste cotton fabrics	Sulfuric acid hydrolysis	18.4 ± 7.8 and 15.1 ± 10.5	15.2 ± 5.9 and 6.3 ± 3.0	249.5 ± 85.6 and 87.9 ± 58.5	The CNC derived from partially dissolved cellulose in solvent treatment demonstrated better reinforcing effects on tensile strength and oxygen barrier properties than CNC derived from completely dissolved cellulose in solvent.	[130]
Waste cotton from the hospital	Ultrasound acid hydrolysis	–	–	10–50	The prepared CNCs had high crystallinity (81.23 %), low particle size (221 nm), and stable thermal properties (140–180 °C) for potential applications in electronics, sensors etc.	[131]
Cotton wool	Sulfuric acid hydrolysis	–	–	–	Carboxylated CNC and chloride anions were used for producing 3D porous polypyrrole supercapacitors with high performance for energy storage applications.	[132]
Coconut gels	Sulfuric acid hydrolysis	~20	–	75–500	The interaction of water molecules with the CNC surface caused substantial structuring of the interface which may affect their surface adsorption and interaction behaviour.	[133]
Kraft pulp	Hydrochloric acid hydrolysis	27.5	17.1	470	Hexadecyl trimethyl ammonium bromide at a low concentration (0.13–0.47 mM) is highly useful for steric barriers to minimize the CNC aggregation.	[134]
Pinewood	Acid hydrolysis	7	–	50	Maleic anhydride improved the dispersion of CNC in high-density polyethylene, which enhanced elongation at maximum tensile strength by up to 20 % of CNC-based composites.	[135]
Wood pulp	Sulfuric acid hydrolysis	–	–	140–217	The low-cost 2D transition-metal dichalcogenides nanosheets/CNC-based composite highly effective for artificial skin, sensor, and multi-responsive photonic films.	[136]
Cotton	Sulfuric acid hydrolysis	2–20 nm	–	~100	The surface modified CNCs improved the tensile strength, modulus and toughness of PLA/CNC biocomposites.	[137]
Cotton pulp fiber	Enzymatic hydrolysis	30–45	–	250–900	The width of ribbon-like CNC reduced with the increase of cellulose concentration.	[138]
Cotton pulp	Sulfuric acid hydrolysis	20–180 (sphere shaped)	4–18 (rod shaped), 10–40 (sphere shaped)	60–160 (rod shaped), 80–300 (sphere shaped)	Two types of high quality CNC were obtained including cellulose nano spheres (short layer distance, lower order degree, and weaker long-range orientation) and cellulose nanorods with anisotropic properties permits their self-organization into chiral nematic liquid crystals.	[139]
Softwood kraft pulp	Sulfuric acid hydrolysis	6	–	140	The developed CNC/waterborne PU photonic composites have enormous scope in intelligent decoration, and anti-counterfeiting applications.	[140]
Rice bran	Sulfuric acid hydrolysis	5.72 ± 1.35	20.79	114.75 ± 37.37	Curcumin-encapsulated Pickering emulsion was successfully prepared using CNCs.	[141]

proposed CNC-based composites demonstrated higher temperature degradation and good re-dispersibility of the freeze-dried than the neat CNC. Thus polyacrylamide/CNC composites have excellent potential to use as compatibilisers and emulsifiers. Researchers have frequently used CNC to strengthen the novel biocomposites, and they have ability to accommodate other natural biopolymeric materials. Recently, Zhao et al. [149] developed CNC/CNF/microscopic cellulosic fines-based collagen biocomposites. Reported results demonstrated that the strength of biocomposites was improved by increasing the concentration of CNF and cellulosic fines. Gois et al. [150] proved that surfactants treated PLA/CNC composites have outstanding degradation rate with low spherulite size. Highly porous structure can also produce through CNC-based composites. For example, polyvinylidene fluoride/CNC composites have remarkable porous structure with the specific surface area of 12.22 $m^2\ g^{-1}$, which allowed an improved adsorption capacity up to 73.04 g/g for engine oil applications [151].

2.1.2. CNC-based hydrogels

CNC-based hydrogels are attractive biomaterials for biomedical applications, thanks to their extraordinary swellability, biocompatibility, and stimuli-responsiveness. In general, CNCs lack the ability to entangle with each other to develop mechanically stable hydrogels, due to their rigid structure and small aspect ratio. Therefore, it is difficult to develop hydrogel by using CNC fillers only, when compared with other polymers [152]. So, mechanically stable CNC-based hydrogels can be acquired by introducing crosslinking networks or by altering surface chemistry [153] which is usually done by incorporating CNCs into polymer networks through physical or chemical methods. The corresponding hydrogels are called physical gels or chemical gels, respectively [154]. Physical gels are bound together through non-covalent bonds like van der Waals forces, polymer chain entanglements, hydrogen bonding, ionic interactions, and crystallite associations. On the other hand, in chemical gels, covalent bonds are formed between polymers through free radical polymerization, graft copolymerization, or radical copolymerization that develop permanent junctions within the polymer network [155].

Currently, several methods including cyclic freeze-thaw processing, homogenized mixing, UV/ion mediated crosslinking, free radical polymerization, and 3D printing have been widely exploited to mix CNC with different types of polymer networks such as PLA, PEG, natural biopolymers, etc. [156]. It also opens pathways for fabricating flexible hydrogels, which exhibit high surface area, high mechanical strength, and tailorable surface chemistry [157]. Recently, CNC-based hydrogels have been extensively applied in different engineering applications [158].

3. 3D/4D printing techniques

AM is a rapidly developed material manufacturing technology for developing sustainable structures through layer-by-layer slices [159]. Compared to conventional fabrication techniques, AM technology permits the fabrication of complex structures. As a result, it has become an effective pathway for achieving high design flexibility and prototyping freedom at high resolution. 3D printing of cellulose-based composites has exhibited its potential for various applications, such as target drug delivery, complex architectures, tissue engineering, dental, automotive, customized food design, etc. [160–162]. The 3D printing technology has been explored for incorporating CNCs into different synthetic polymers as well as some types of natural polymers [163]. These printing techniques include extrusion-based printing (e.g., fused deposition modelling (FDM), direct ink writing (DIW)), vat photopolymerization (e.g., stereolithography (SLA), digital light projection (DLP), two-photon polymerization (TPP)), and inkjet printing (IJP) [164]. Table 3 summarizes the important features, advantages, disadvantages, and common stimuli mechanisms for different 3D printing techniques used to print CNC-based sustainable structures. This section elucidates the additive manufacturing techniques used to print CNC-based composites.

3.1. Extrusion-based printing

Extrusion-based printing is a commonly used process, which enables the programmable assembly of 3D periodic structures. This technique selectively dispenses the extruded material in a layer-by-layer fashion, as illustrated in Fig. 5(c). It can use sustainable cellulosic materials including cellulose derivatives, lignocellulosic materials, and cellulose-based nanomaterials, as feedstock printable materials [183]. Moreover, extrusion-based printing is highly suitable for developing CNC-based structures with propitious anisotropic properties by aligning all CNCs in one direction. These 3D-printed uniaxial-oriented CNC-based composites can be extensively applied to develop actuators and optical devices [184].

Extrusion-based printing is further classified into fused deposition modelling (FDM), direct ink writing (DIW), and micro-extrusion bioprinting techniques [186]. FDM uses thermoplastic filaments of small diameter, which are continuously fed for printing the 3D object in a layer-by-layer manner [187]. However, it is quite challenging to fabricate CNC-based filaments with small diameters, due to the die swelling, which occurs upon the extrusion of viscous liquefied materials from a small nozzle [188]. Additionally, the printability or extrudability of CNC-based materials is vital for producing high-quality structures [189].

On contrary, DIW is preferred over the other 3D printing techniques, thanks to its ability to print a wide range of materials for developing micro-scaled architectures. This approach involves the deposition of ink in a layer-by-layer fashion. It is highly advantageous and conceivable to develop intricate programmable 3D-printed architectures through sequential steps [190]. However, the major challenge associated with the DIW printing is to develop inks with optimum rheological properties. This approach is considered highly suitable for direct paste writing of cellulose-based suspensions. Thus, cellulose suspensions and rheological properties are two crucial factors involved in DIW [191]. The proper printing materials or "inks" should have high viscosity and shear thinning behaviour for reliable extrusion-based printing. To date, metal and polymer-based smart materials are usually employed in extrusion, which requires melting at high temperatures and are relatively expensive materials. Recently, CNCs are applied as a blending modifier or reinforcer to improve the mechanical properties of hydrogel-based inks like alginate, hyaluronic acid, and polyvinyl alcohol (PVA). Additionally, it can be used as a viscosity modifier for improving the ink viscosity [192,193].

Tremendous work has been conducted in searching for hydrogel inks from naturally available materials. In this regard, the development of cellulose-based materials for 3D printing perspective and as natural materials is highly promising [194,195]. In this context, CNCs are investigated extensively due to characteristics such as renewable, biocompatible, and functional reinforcing agents in 3D-printed structures. CNCs also act as rheology modifiers in extrusion-based printing processes such as DIW and FDM when it applies shear and extensional

Table 3
Additive manufacturing, printable sustainable materials, common stimuli's, their advantages, and disadvantages.

AM techniques	Method	Sustainable and other biomaterials	Commonly employed stimuli	Advantages	Disadvantages	Ref.
Extrusion	FDM	Thermoplastic biopolymers (PLA, PCL PLGA, PEG), nanocellulose	Temperature	Less costly and fast printing process	Limited printing resolution High printing cost	[165–167]
	DIW	CNC, starch, gelatin, alginate, maize protein, KC, hydrogels	Temperature, water, and heat	Multi-material printing and low printing waste Room processing temperature	Inferior printing efficiency Poor printing resolution	[168–170]
Vat polymerization	DLP	Metamaterials, elastomers, PEG, PEGDA, and PCL	Temperature and heat	Very fast printing with high resolution on both micro and nanoscales	High materials cost	[171–173]
	SLA	Silk, CNCs, and alginate	Temperature	Complex geometric shapes with high resolution of 20 µm Good surface finish	Unstable photopolymers over time Expensive feedstock materials	[174–176]
Laser-assisted printing	SLS	Ceramics, alloys, thermoplastic biopolymers	Temperature and water	Complex shapes with no support are required Large-scale scaffolds	Non-uniform printed shape Poor surface quality Expensive powder and laser system	[177–179]
Inkjet printing	IJP	Thermoplastic biopolymers (alginate, silk fibroin, PEG) nanocellulose, hydrogels	Temperature, pH	Multiple printing abilities Complex shapes High throughput and efficiency	Low resolution for multi-material layers	[180–182]

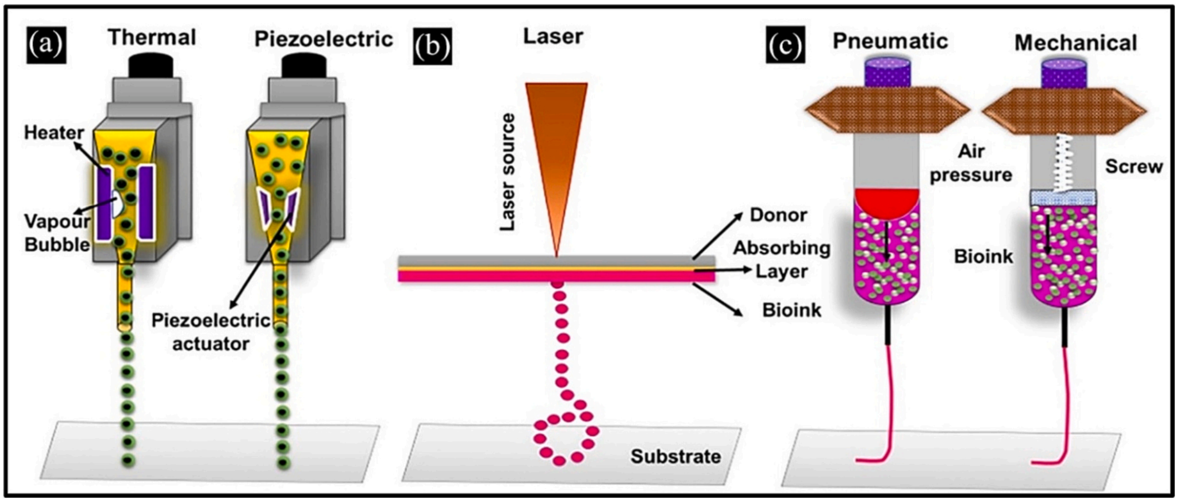

Fig. 5. Schematic depiction of various types of printing; (a) Inkjet printing; (b) Laser-assisted printing; (c) Extrusion printing. (Adapted from [185], under the Creative Commons Attribution license).

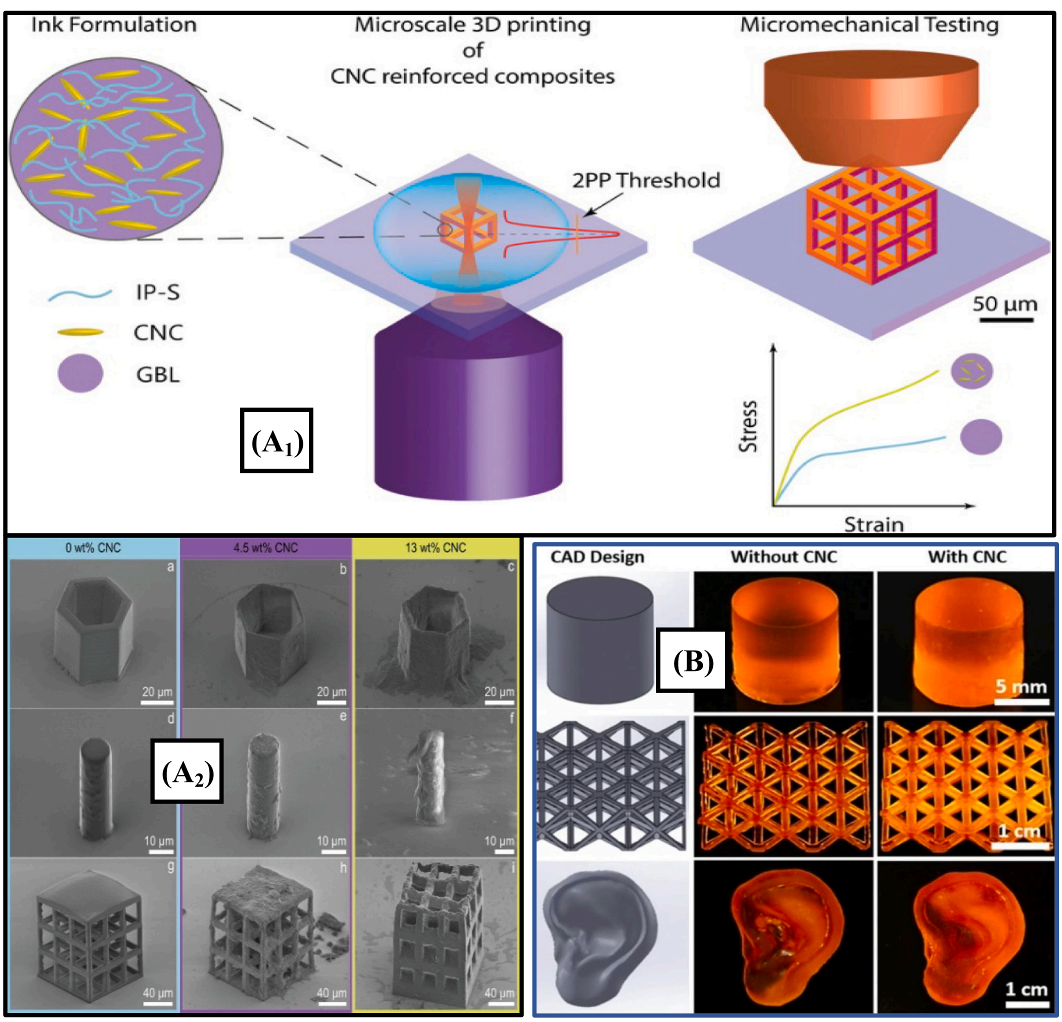

Fig. 6. (A_1) Schematic illustration of ink formulation, which is used to develop CNC-based composites using TPP printing; (A_2) 3D-printed micro-scale structures developed by varying CNC contents (adapted from [206], copyright 2022, Wiley-VCH GmbH); (B) CNC/PEGDA-based 3D-printed disk, octet truss lattice structure, and an ear model printed by using DLP technique (adapted from [205], copyright 2019, Springer Nature B.V.).

stresses to the suspension [196–198].

3.2. Vat photopolymerization

Apart from extrusion-based printing, vat photopolymerization such as stereolithography (SLA) and Digital light projection (DLP) are widely applied to print CNC-based composites containing photopolymer resins. These techniques can design and fabricate bone scaffolds, robots, and pharmaceutical devices by using CNC fillers and photoreactive resins as the prime constituents [199]. The incorporation of these nanomaterials improves the thermal stability and mechanical properties of the printed products. SLA and DLP, as vat photopolymerization techniques, use light sources such as gamma, X-rays, UV, and electron beams for the polymerization of liquid resins [200].

In SLA, a liquid photopolymer is cured selectively through light-activated polymerization. SLA printing is highly suitable for printing intricate geometric parts [201]. Additionally, this technique exhibits high manufacturing accuracy, small layer thickness, and excellent surface quality. The mechanical properties of optically curable resins can be improved by incorporating CNC fillers [202]. DLP is a suitable 3D printing process, which is capable of developing complex and precise micro/nano-scaled structures at a high speed [203]. DLP prints materials from a single exposure of light and offers higher efficiency, compared to other AM processes [204]. Some authors have developed CNC-based composites by using DLP printing. For instance, Li et al. [205] developed intricate structure by incorporating CNCs into photocurable resin, as depicted in Fig. 6(B). The results revealed that the mechanical properties of CNC/PEGDA-based composites were significantly improved due to the addition of CNCs. Furthermore, curing thickness also helped in tuning the mechanical and swelling properties of DLP-printed composites. It can be further exploited to develop 3D-printed complex structures for biomedical applications.

Two-photon polymerization (TPP) is an attractive micro-scale 3D printing process, which permits voxel-by-voxel printing with ultrafine resolution. In this approach, polymerization is confined to the laser focal spot, which results in the printing of 3D structures inside a film of monomer without changing the base material. It is a highly suitable technique to develop intricate micro-structures such as micro-needles and micro-scaffolds [207]. CNC-based inks can also be applied to develop small-scale architectures. For instance, Groetsch et al. [206] developed non-cytotoxic CNC-based ink for printing intricate 3D structures using a TPP process, as illustrated in Fig. 6(A_1–A_2). The results revealed that CNC contents improved the stiffness and mechanical properties. Thus, this printing approach can be used to improve the performance of micro-scale products like micro-scaffolds or to tune energy absorption devices.

3.3. Inkjet printing

Inkjet printing (IJP) is a non-contact, cost-effective, and fast 3D printing process, which uses multiple nozzles for replicating images onto the substrate, through the precise ejection of ink by using acoustic or thermal forces onto a predetermined path [208], as illustrated in Fig. 5 (a). Inkjet-based 3D printers exhibit high resolution, speed, and precision, that makes it a suitable platform for developing functional 3D architectures [209]. Table 4 summarizes different features of AM techniques including IJP, which are used to print CNC-based sustainable composites. This technique offers high resolution and prints highly precise structures [210].

3.4. 4D printing

One of the major drawbacks of 3D printing is the development of static structures that lack shape-morphing properties. With an increasing focus on the development of multifunctional and sophisticated smart structures, which can adapt to changes under external surroundings [234]. Scientists have found a novel manufacturing method known as the 4D printing, which can overcome the issues associated with 3D printing. 4D printing uses time as the 4th dimension and is the advanced version of 3D printing for achieving the necessary functionalities of 3D-printed structures such as a change in shape, colour, and properties under external stimuli [235]. 4D printing also relies on a highly complex and powerful mathematical modelling of the desired structure, similar to the 3D printing. A brief comparison between 3D and 4D Printing technologies is summarized in Fig. 7.

4D printing uses smart materials, which are programmed to offer changes in their properties in a controlled fashion upon an external stimulus. Mostly, extrusion and vat photopolymerization-based techniques are generally employed for the 4D printing. To date, various stimuli responsive materials, such as shape memory gels (SMGs) including hydrogels, SMPs, shape memory composites, shape memory alloys (SMAs) and liquid crystalline elastomers (LCEs) have been intensively used as promising feedstocks of 4D printing [236]. Among them, SMPs, SMGs and LCEs have demonstrated remarkable programmable anisotropy for variety of 4D behaviour such as self-actuating, large-amplitude, self-healing, self-sensing, self-diagnostic, shape-morphing effects, and long-lifetime anisotropic motions upon stimuli [237]. These stimuli responsive materials have capability to capture safe

Table 4
Features of different types of 3D/4D printing techniques.

	AM techniques			
Aspects	Extrusion	Laser	Inkjet	Ref.
Resolution (μm)	40–1200	40–100	20–100	[211–213]
Printing time	Medium	High	Low	[214–216]
Contact	Contact	Non-contact	Non-contact	[217]
Viscosity (m·Pas)	30–6 × 10^7	1–300	3.5–12	[166]
Dispensing speed	10 μm–50 mm/s	200–1600 mm/s	1–10,000 droplets per second	[185]
Dispensing from	Filament	Droplets/continuous deposition	Droplets	[218–220]
Dispensing mechanism	Pneumatic or mechanical	Laser	Thermal and piezoelectric	[221–223]
Gelation techniques	Chemical, photo-cross linking, temperature, shear thinning	Photo-cross linking, chemical	Chemical, photo-cross linking	[224–226]
Cost	Medium	High	Low	[227–229]
Popular companies/ countries	Germany, Spain,	Germany, Japan, UK	Israel, USA, Japan	[230–233]
Popular 3D Printers	EnvisionTEC and RegenHU, Markforged, Triditive, Anisoprint ProM IS 500, Stratasys F900, CreatBot PEEK-300, 3DGence Industry F421	EOS GmbH, DMG Mori, SLM Solutions, Trumpf, Aconity3D, Materialise NV (MTLS), Proto Labs Inc. (PRLB), 3D Systems Corp. (DDD)	Zebra Technologies, nano Dimension, ChemCubed, Domino printing, Hanita Coatings, Epson	

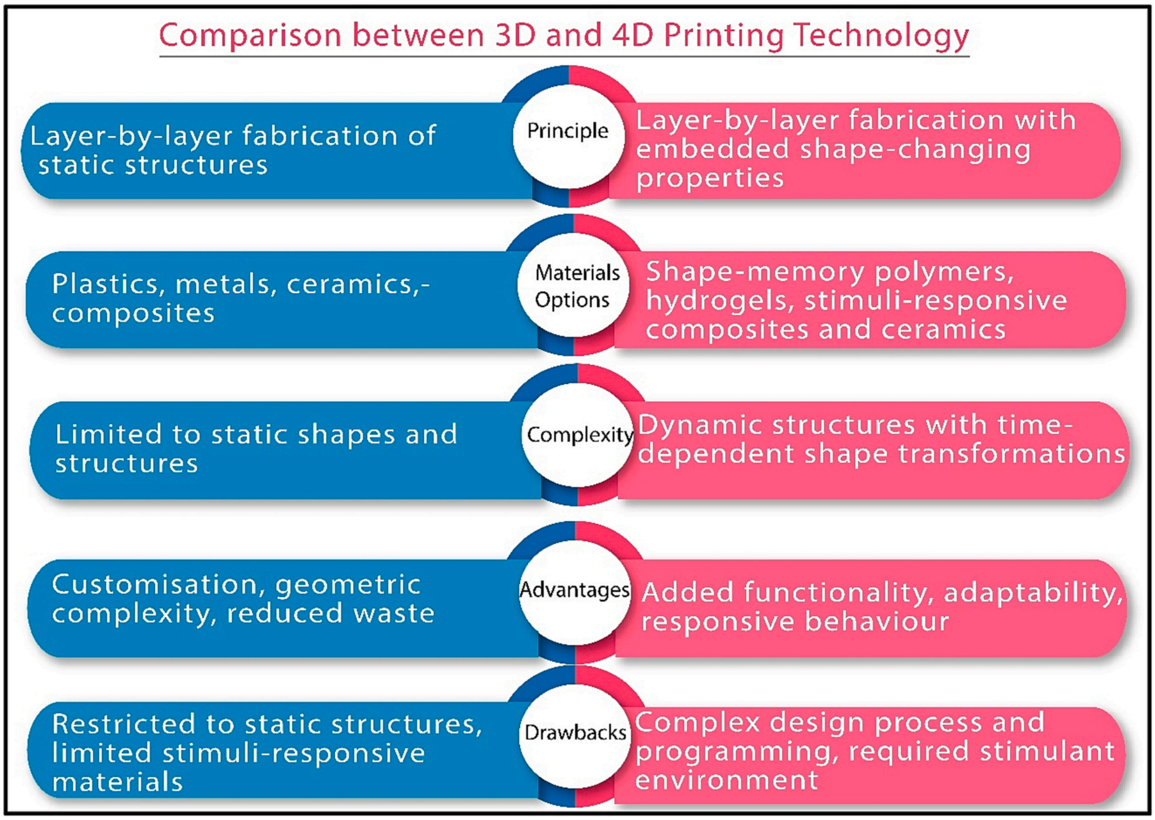

Fig. 7. A brief comparison between 3D and 4D Printing technologies (Figure drawn with the help of [247]).

response in their memory under external stimuli such as temperature, light, pH, ultrasound, humidity, magnetic field, or chemical substances [238–240]. Some temperature-triggered SMPs such as PU demonstrate dual-shape morphing effects [241]. These SMPs have crystallites or oriented polymeric chains that trigger their shape-changing behaviour associated with a transition temperature and this is happened at the molecular level [242–246].

Generally, SMPs and SMGs showed noteworthy changes in their physical properties, such as stiffness, shape or volume, under stimulant environment [248]. While, SMA sensed environmental stimuli, as a result their structure showed martensitic phase transformation autonomously under thermoelastic or stress-induced-based stimulus for demonstrating unique "sensing" and "actuation" properties [249]. SMAs are being continuously explored in 4D printing technology for their promising role in micro-actuators, electromagnetic shielding interface devices and self-deployable structures [250]. Table 5 provide comprehensively overviewed of 4D printing technology with focused towards smart materials, potential shape morphing behaviour for wide range of applications.

Shape morphing behaviour and programming in 4D printing is demonstrated as shape memory effects (SME) such as one-way SME, two-way SME and multiple way SME. In one way SME, the printed structure or devices must be reprogrammed after each recovery cycle. Two-way SME allows reversible printed structures with two different shapes under specific time, and stimuli. Multiway SMEs allow an intermediate shape between temporary and original shapes under controlled cycle of stimulus and programming [251].

4D printing can be employed for both single-material and multi-material-based smart structures such as uniform distribution, gradient distribution, and special patterns by integrating stimuli-responsive materials with their functionalities [252,253]. Thus, 4D-printed multi-materials smart structures regarded as advanced materials for all subject domains in chemistry, biomedical, materials, and computer science. Moreover, 4D-printed multi-material structures offer more complex pre-programmed actuating deformation for wide spectrum of applications from macro to centimeter to micro scale range for advanced actuators

Table 5
Brief overview of 4D printing technology using extrusion, inkjet, and laser-based 3D techniques for various smart materials and their potential applications.

Smart materials	Sub-types	Commonly employed stimulus	4D behaviour	Potential applications	Ref.
SMGs	Alginate, PEGDA, PNIPAM, cellulose	Moisture, solvent, temperature	Shrinkage, swelling, self-healing	Tissue engineering, drug delivery, actuators	[254]
SMPs	Soybean oil epoxidized acrylate, PLA, PVA, and PEG	Solvent, temperature, light, electric and magnetic field	Bending, twisting, folding, self-adaptive	Soft robotics, optical devices, energy dissipations, tissue engineering, anti-counterfeiting, and drug delivery	[255]
LCEs	–	Temperature, UV light, electric and magnetic field	Bending, stretching, folding	Soft robotics, smart structures, optical sensors, and smart displays	[256]
SMAs	Ni-Ti alloy (Ti rich), Ni—Ti alloy (Ni rich), Cu-Al-Ni alloy	Temperature, light, electric and magnetic field	Martensitic phase transformation, bending, twisting, folding	Micro-actuators, optical devices, self-deployable structures, electromagnetic smart devices	[257,258]

applications.

3.4.1. CNC-based stimuli-responsive materials for 4D printing

4D printing uses the same 3D printing techniques for the development of smart and dynamic structures. This technology has potentials in achieving high resolution, thus, developing complex parts with high accuracies such as fast response grippers, soft actuators, biomimetic, and other biomedical devices [259–262]. The utilization of CNC-based sustainable materials in the 3D printing sector entirely depends on its functionalization techniques, which are usually performed through dissolution regeneration avoiding water solubility and later used as reinforcement or modifiers in AM [263].

Feng et al. [264] prepared CNC/PEGDA-based chiral nematic structures with reversible multiple-stimuli-responsive functions for a potential colorimetric (humidity) sensor. The plasticization of PEGDA and the hydrogen-bonding interactions among CNCs, and the composite coatings responded to various environmental signals. Insights of this study demonstrated that distinguishable colours were observed when coating immersed in ethanol and methanol mixed solvents. Moreover, a clear red shift up to 262 nm was seen when the coatings were placed in increased relative humidity from 30 to 98 %. According to Tang et al. [265], stimuli-responsive properties of CNC/hydrogel systems such as self-healing can be regulated and controlled through temperature demonstrating a thermally triggered actuator.

3D-printed CNC-based composites also demonstrate shape-changing behaviour under external stimuli, due to alignment during printing and the hygroscopic nature of the NPs, which causes predetermined bending, twisting, and curling noteworthy characteristics for 4D printing. These 4D-printed CNC materials are effectively used in the fabrication of lightweight and highly intriguing structures, and flexible sensors for applications such as wearable electronics [266–268]. One of the most traditional and well-investigated stimuli applied to tune the behaviour of smart sustainable polymers is heat. Thermo-responsive materials contain both hydrophilic and hydrophobic segments, which exist in the form of a collapsed globule or an extended chain. A polymer collapse or chain extension depends upon upper critical solution temperature or low critical solution temperature, respectively. CNCs with distinct properties can be combined with thermo-responsive polymers to develop physically adaptable smart structures by using 3D printing techniques [269–271]. For instance, Belyaeva et al. [272] employed thermo-responsive poly(n-isopropylacrylamide) (PNIPAM)/CNC-based hydrogel to evaluate the mechanical properties of fibrillar structure developed through a DIW printing. The results revealed that the resulting gel exhibited stimuli-responsive characteristics and shear-thinning behaviour. PNIPAM/CNC-based hydrogel changed their behaviour from translucent to opaque, during the transition from 25 °C to 37 °C, as illustrated in Fig. 8(A). Additionally, this temperature transition increased the storage modulus by 2–3 times. It was also found that the transition temperature can be controlled by varying the concentrations of CNC and PNIPAM.

Similarly, Zhou et al. [273] impregnated shape memory polyurethane (PU) with CNC and investigated the shape fixation ratio and the shape recovery rate of PU/CNC-based structure developed through an FDM technique, as illustrated in Fig. 8(B). The results revealed that an appropriate proportion of CNCs and ball-milled nanocellulose (BMC) improved the mechanical strength. Additionally, the authors observed that these PU/CNC-based composites exhibited 99.2 % shape fixity and 98.9 % shape recovery rate. Furthermore, these 3D-printed composites also presented tunable glass transition temperature (Tg), which is close to human body temperature. Thus, allowing them to be used in smart

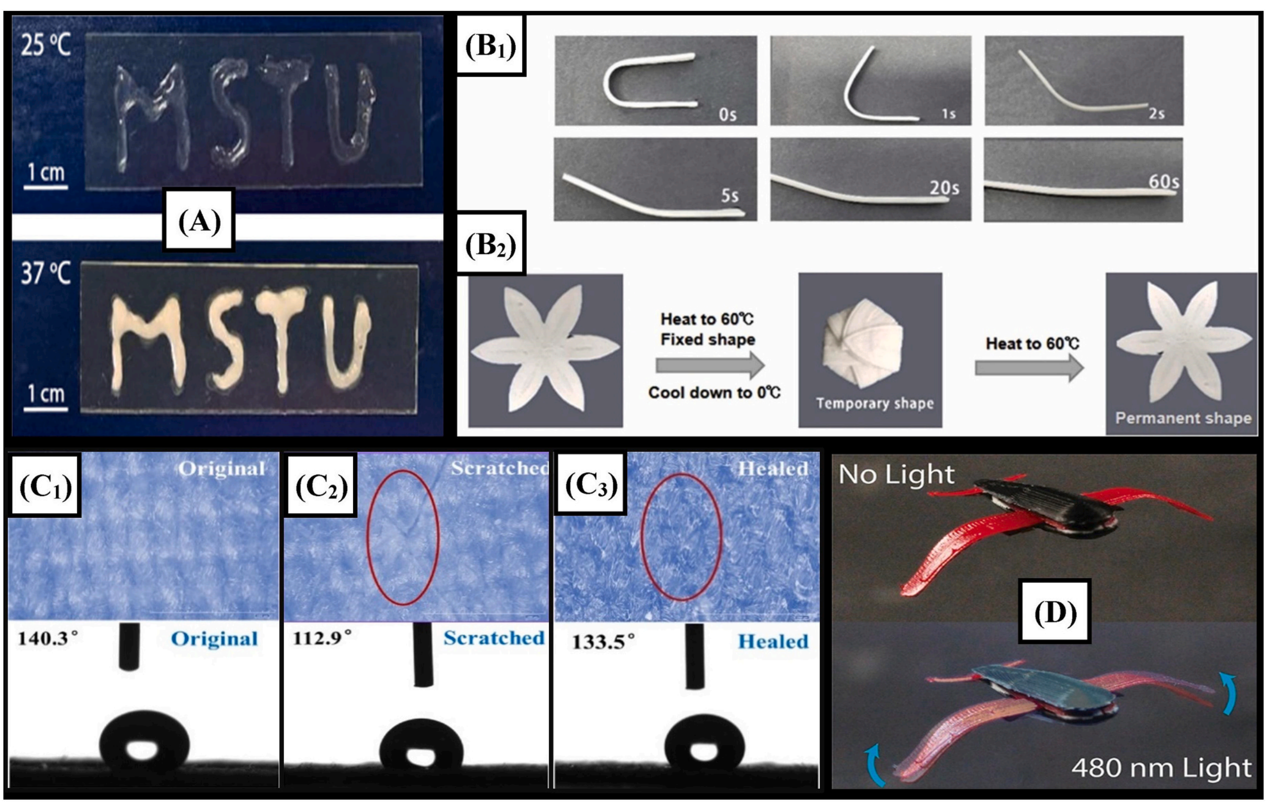

Fig. 8. (A) CNC/PNIPAM-based hydrogel extruded letter, which changed its behaviour from translucent (25 °C) to opaque (37 °C) (adapted with permission from [272], copyright 2022, Elsevier Inc.); (B) Digital images of the shape recovery behaviour (B_1) Bending of the sample at 60 s, (B_2) Petal shape under heating and cooling (adapted with permission from [273], copyright 2022, Elsevier B.V.); (C_1–C_3) Microscopic ultra-depth-of-field and water contact angle photos of different fabircs; (C_1) Original finished; (C_2) Scratched finished; (C_3) Self-healed finished (adapted with permission from [274], copyright 2023, Wiley Periodicals LLC.); (D) CNC/LCE-based photo-responsive actuator developed through the DIW (adapted with permission from [275], copyright 2022, American Chemical Society).

biomedical devices.

Light is a non-contact stimulus, which permits remote actuation with precise temporal and spatial control. Photo-responsive CNC-based nanocomposites exhibit shape-changing features such as reversible bending and dynamic softening, when exposed to light stimulus. The introduction of CNC into polymer matrices is used to print soft robots, self-standing structures, and other biomedical devices [276,277]. For instance, Müller et al. [275] introduced CNC-based sustainable fillers into a liquid crystalline elastomer (LCE) to develop photo-responsive 3D architecture through DIW printing, as illustrated in Fig. 8(D). The results revealed that the excellent covalent bonding between NPs and matrix promoted rapid shape recovery after actuation. Additionally, this 3D-printed structure with tailored and dynamic stiffness presented an excellent potential for developing different actuating and customized medical devices. In another work, Zhou et al. [274] studied photo-responsive CNC/fluorinated polyacrylate containing coumarin. Therein, scanning electron microscope (SEM) and elemental dispersive X-ray analysis (EDX) results showed that the photo-responsive CNC had excellent self-healing characteristics when the prepared emulsion was uniformly coated on the fabric surface, as depicted in Fig. 8(C). Furthermore, the finished fabric had good oleophobicity, hydrophobicity, and surface repair performance.

Polymers with H-bonding interactions generally possess hygroscopic nature. CNCs contain hydrophilic hydroxyl group, however, they are not sensitive to moisture, due to their strong hydrogen bonding and high crystallinity. Therefore, CNCs are promising candidates for developing moisture-driven architectures [278]. Moisture-responsive CNC-based composites can be developed moisture-driven architectures by combining CNCs with other moisture-sensitive materials like PVA, PEG, etc. These materials are highly suitable for fabricating sustainable smart structures, which can be reprogrammed to reshape and perform tasks by adapting their morphologies for the specific application [279]. For instance, Cecchini et al. [280] employed 4D printing technology to develop environmentally-driven humidity responsive bioinspired soft robot for the realization of an artificial seed, as presented in Fig. 9(A). The authors fabricated a bilayer hygroscopic structure, in which a PCL-based passive layer was developed through FDM printing, and the electrospun polyethylene oxide (PEO)/CNC-based active fiber layer was deposited on the top of the substrate. The results revealed that 4D-printed hygroscopic robot is highly suitable for environmental soil exploration, thanks to their comparable biomechanical performances and geometrical dimensions of natural and artificial seeds.

In another study, Gevorkian et al. [281] developed a single-layer nanocolloidal hydrogel actuator by using extrusion-based printing, which exhibited shape transformation under water stimulus, due to structural anisotropy. Fig. 9(B) depicts the swelling-induced shape morphing behaviour of CNC/gelatin methacryloyl (GelMA)-based hydrogel and different 3D shapes can be yielded by changing the CNC orientation.

Thanks to the presence of a large number of hydroxyl groups on nanocellulose surface, functional fillers can easily be incorporated onto the surface, thus, endowing it with electro- or magneto-responsive properties [284]. The introduction of these functional fillers such as carbon nanotubes (electro-active particles) and magnetic nanoparticles can help in producing controllable CNC-based smart structures for different engineering applications [285]. Electro-active CNC-based materials can be applied as energy conversion devices for sensors, robots, artificial muscles, and controlled drug release devices. For instance, Bi et al. [283] developed electric-driven carboxylic multiwall carbon nanotubes (CMWCNTs)/CNC-based conductive device through an FDM technique (as illustrated in Fig. 9(D)), which exhibited excellent shape-memory and self-healing ability.

Multi-responsive materials consist of multiple integrated responsive functional groups, which permit the simultaneous response to different two or more stimuli. These materials have a wider scope of applications, compared with single-responsive materials. Multi-responsive CNC-based 4D-printed products can be developed by designing multilayer functional architectures. Consequently, multiple single response functions are concentrated in one actuator, focusing on diverse external stimuli [286]. Various researchers explored the mechanical performance of 4D-printed composites by reinforcing the polymers with CNCs. For instance, Qu et al. [282] explored a multi-responsive, flexible, bilayer acrylamide-acrylic acid (AAm-AAc)/CNC-based hydrogel network through 4D printing. The different layers were combined through reversible coordination of Fe^{3+} to Fe^{2+} which was also responsible for shape morphing behaviour through stretching and unstretching of layers, as presented in Fig. 9(C). The 4D construct was immersed in sodium lactate and exposed to ultraviolet (UV) light, until maximum deformation was achieved. This 4D-printed structure exhibited good biocompatibility and excellent mechanical properties. Additionally, the results showed that a 4D-printed bilayer hydrogel stent demonstrated excellent curvature and flexibility, which is highly suitable to treat enteroatmospheric fistulas (EAFs) in the intestine.

Table 6 incorporates the development of different CNC-based smart structures such as wearable electronics, tissue engineering, soft robots, metamaterials, and self-adaptable structures, etc. through 4D printing techniques.

4. Applications of 3D/4D printed CNC-based smart materials

The combination of CNCs and other biopolymers like gelatin, chitosan, and alginate have been extensively investigated for formulating composites and hydrogels for 3D printing, thus, expanding the use of CNCs in different applications, as illustrated in Fig. 10. Table 7 summarizes the different applications of CNC-based sustainable structures fabricated by using AM techniques. Additionally, CNCs have also been applied to develop smart hydrogels and composites, which demonstrate their change in shape, functions, and colours, thus, meeting ideal 4D printing materials requirements [293]. Nowadays, 4D printing of CNCs has also demonstrated its applications such as wearable and flexible electronics and ionotropic devices. 4D printing has the potential to use in various optical applications for its ability to form lyotropic liquid crystals and has good mechanical properties and interference colour changes under external stimuli [294–297]. This section incorporates the applications of additively manufactured CNC-based materials.

4.1. Biomedical

With the booming development of stimuli-responsive materials for 4D printing technology an unprecedented prospect have been seen in biomedical field. These 4D-printed smart structures have multi-layered, unique texture, and complex microstructures for novel applications such as in precision medicine [327]. Recently, the 3D/4D printing technologies for the fabrication of CNC-based biomaterials with 3D-shaped architectures have drawn great attention in wound healing, tissue regeneration, and regenerative medicine, especially biomineralization agents and mechanical reinforcement, thanks to their biodegradability, non-toxicity, biocompatibility, as well as excellent ion deposition, printability, self-assembly behaviour, distinct geometry, and ability to develop the bone scaffolds interface [328–330]. Valiant efforts have been made by researchers to develop CNC-based scaffolds through 3D printing [158]. Moreover, 3D-printed CNC structures has huge prospect in various biomedical formulations including bioimplants, regenerative tissues, self-folding tubes, patches, and cell-laden or cell-absent structures. For instance, Bakht et al. [331] proposed a facile strategy for high-resolution cell-laden construction within an extracellular matrix mimetic fibrillar support using CNC-based natural material through 3D bioprinting. Insights of this study revealed that a highly transparent biomimetic nano-scaled fibrillar matrix was achieved through CNC fluid gel which further showed exceptionally arbitrary geometries and self-assembly with biocompatible calcium ions. Thus, the proposed methodology is considered an efficient platform for automated biofabrication

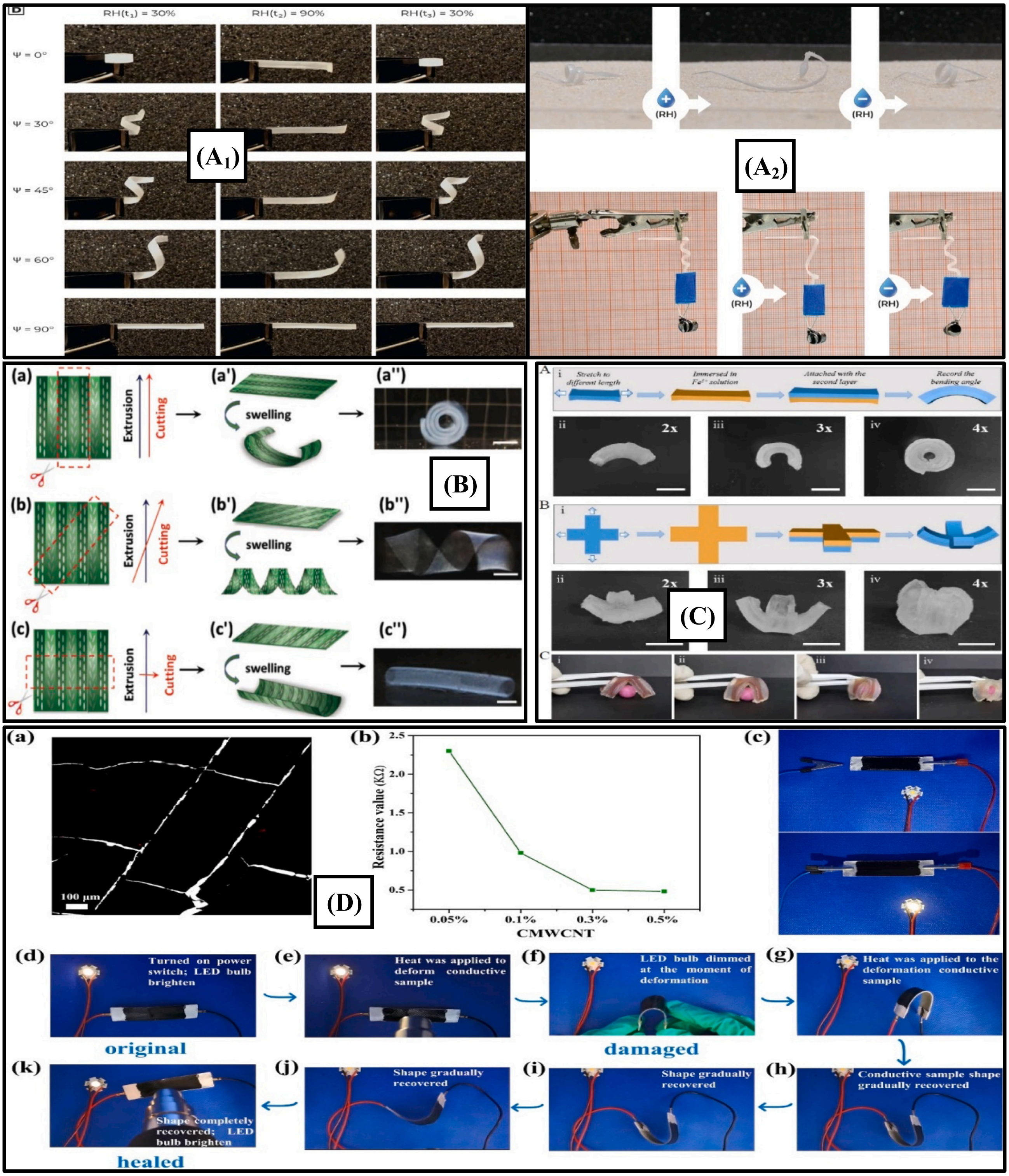

Fig. 9. (A_1) Reversible behaviour of bilayer structures under different humidities; (A_2) Self-lifting performance of artificial seed-like robot under humidity stimulus (adapted from [280], under the Creative Commons Attribution license); (B) Relationship between swelling induced CNC/GelMA-based hydrogel shape and orientation of CNC w.r.t the long sheet axis (adapted with permission from [281], copyright 2021, Wiley-VCH GmbH); (C) Shape-morphing behaviour of bilayer hydrogel strips, (ii–iv) Images of hydrogel strips illustrating curled structures with various bending degrees programmed by different pre-stretched lengths of the first layer, (b) Images of cross-shaped hydrogels exhibiting curled structures with different bending degrees, (c) Images of hydrogel grabbing a bead in its cross-shaped bilayer (adapted from [282], under the Creative Commons Attribution license); (D) Shape-memory assisted self-healing nature of conductive device; (a) Image showing stressed bending part of the device; (b) Relationship between concentrations of CNTs and change in resistance; (c–k) Lightening and electric healing process of the conductive device (adapted with permission from [283],copyright 2021, Elsevier B.V.).

Table 6
CNC-based smart structures fabricated by combining AM techniques and external stimuli for different applications.

AM technique	Smart biomaterials	Stimuli	4D behaviour	Applications	Ref.
FDM/electrospinning	PCL/PEO/CNC	Humidity	Swelling	Bioinspired soft robot	[280]
Extrusion	AAm-AAc/CNC	Light and solution	Stretching and bending	Clinical applications	[282]
FDM	PLA/CNC	Water	Swelling	Bone scaffolds	[287]
Extrusion	CNC/polyester	Heat	Bending	Bioinspired structures	[288]
DIW	CNC	Light	Stretching and bending	Dynamic dampers and energy absorbers	[277]
Extrusion	CNC/PU/BMC	Temperature	Bending	Smart devices	[273]
DIW	CNC/PNIPAM	Temperature	Functional property change	Biomedical applications	[272]
DIW	CNC/AA	Temperature	Self-healing	Wearable sensors	[289]
DIW	CNC/LCE	Light	Bending and folding	Smart sensors and actuators	[275]
DIW	CNC/GelMA	Water	Swelling and contraction	Smart actuators	[281]
FDM	CNC/PLA	Temperature	Folding	Wearable electronics	[290]
DIW	CNC/PUA	Light	Shrinking	Energy absorbing structures	[276]
Extrusion	CNC/PCLA-PEG-PCLA	Temperature	Bending	Biomedical scaffolds	[291]
DIW	CNC/PNIPAM	Humidity	Bending and twisting	Self-supporting intricate architectures	[292]
FDM	CNC/TPU/PCL	Electric	Bending	Biomimetic skin and conductive devices	[283]

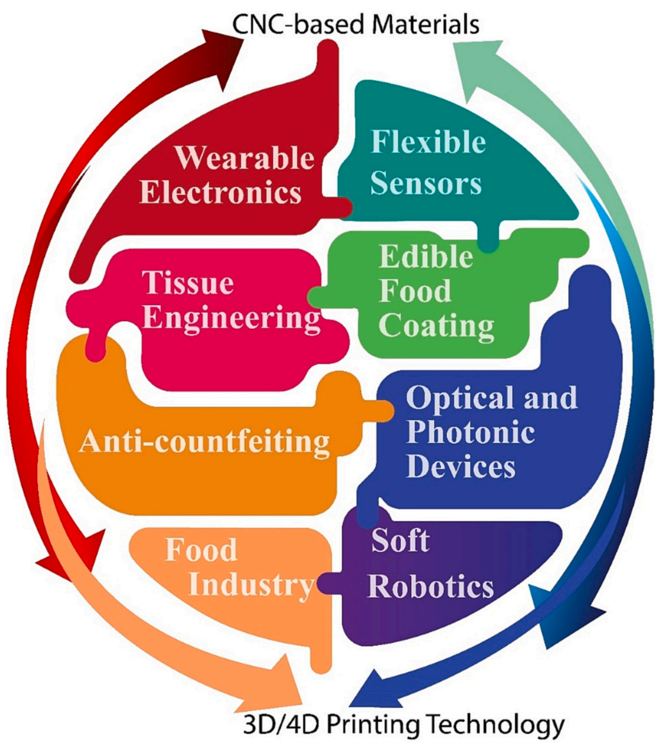

Fig. 10. Applications of CNC-based structures fabricated by using 3D/4D printing.

such as liver tissues and organ models.

Sultan et al. [308] developed CNC/gelatin/sodium alginate (SA)-based biomedical hydrogel scaffolds through a DIW printing, as illustrated in Fig. 11(A). The results indicated that the nano-scaled pore wall thickness and the orientation of 3D-printed hydrogel nanostructured scaffolds exhibited excellent cell growth, interaction, and proliferation during tissue regeneration.

4D printing using CNC-based sustainable materials under human-related stimuli such as pH, and body temperature further improves the cost-effectiveness, sustainability, excellent match with human tissues, on-demand, and rapid fabrication as well as versatility for dynamic structures [332–334], thus, opening a new paradigm for novel biomedical applications. For instance, Cui et al. [291] introduced CNCs into polyethylene glycol-aliphatic polyester block copolymers to develop thermo-sensitive 3D hydrogel constructs through an extrusion-based printing for bioprinting applications, as illustrated in Fig. 11(B). These CNC-enhanced copolymers developed hydrogel exhibited

Table 7
A summary of different CNC-based sustainable composites developed through 3D printing for various applications.

AM technique	Smart biomaterials	Applications	Ref.
Extrusion	PSA/CNC	Optical/photonic technologies	[298]
DIW	CNC/AAm	Chiral structures	[299]
FDM	PCL/CNC/PU	Surface protection coatings	[300]
DIW	CNC	Sustainable smart structures	[301]
DIW	CNC/silver nanowires	Wearable ion sensors	[302]
SLA	CNC/PEGDA	Soft tissue engineering	[303]
DIW	CNC/XG	Liver tissue engineering	[304]
Extrusion	Gelatin/alginate/CNC	Bone grafting	[305]
FDM	CNC/PHBH	Personalized medical device	[306]
IJP	CNC	Origami tube structures	[307]
DIW	CNC/apple/pureed	Self-supporting edible structures	[93]
DIW	CNC/gelatin/SA	Tissue regeneration	[308]
DIW	CNC	Smart textiles, photonics, and high-performance architectures	[309]
SLA	CNC/MA	Tissue engineering	[310]
SLA	CNC-g-MA	Biological tissue engineering	[311]
SLA	CNC/MAA	Energy storage devices	[312]
Extrusion	CNC coating onto PEGDA	Tissue engineering and regenerative medicine	[313]
Extrusion	CNC/KC/MC	Cell proliferation, tissue regeneration and wound dressing	[314]
DIW	CNC/MP	Medical tissue engineering	[315]
DIW	CNC/PI	Thermal insulation	[316]
FDM	CNC/PLA	Dental applications	[316]
DLP	CNC/PEGDA	Biomedical applications	[205]
DLP	CNC/CMC	Pharmaceutics and tissue engineering	[203]
DIW	CNC/Py-PNGs	Healthcare sensors	[317]
DIW	CNC/alginate/gelatin	Tissue engineering	[158]
DIW	TOCN/alginate/dECM	Cartilage tissue engineering	[318]
DIW	Gelatin/DAC	Tissue repairing	[319]
Extrusion	TA/CNC	Wearable sensors	[320]
Extrusion	PLA/PHB/CNC	Different engineering applications	[321]
DIW	PAM/CNC/GNRs	Optical devices	[322]
SLA	CNCMA/pHEMA	Self-expandable stent	[323]
DIW	CNC/silicone/carbon black	Personalized insole design with embedded piezoresistive sensors	[324]
Droplet-based printing	Alginate/CNC/$CaCl_2$	Personalized oral protein delivery	[325]
TPP	CNC	Tissue scaffolds	[206]
IJP	CNC/NPES/organosilane	Anti-counterfeiting applications	[326]

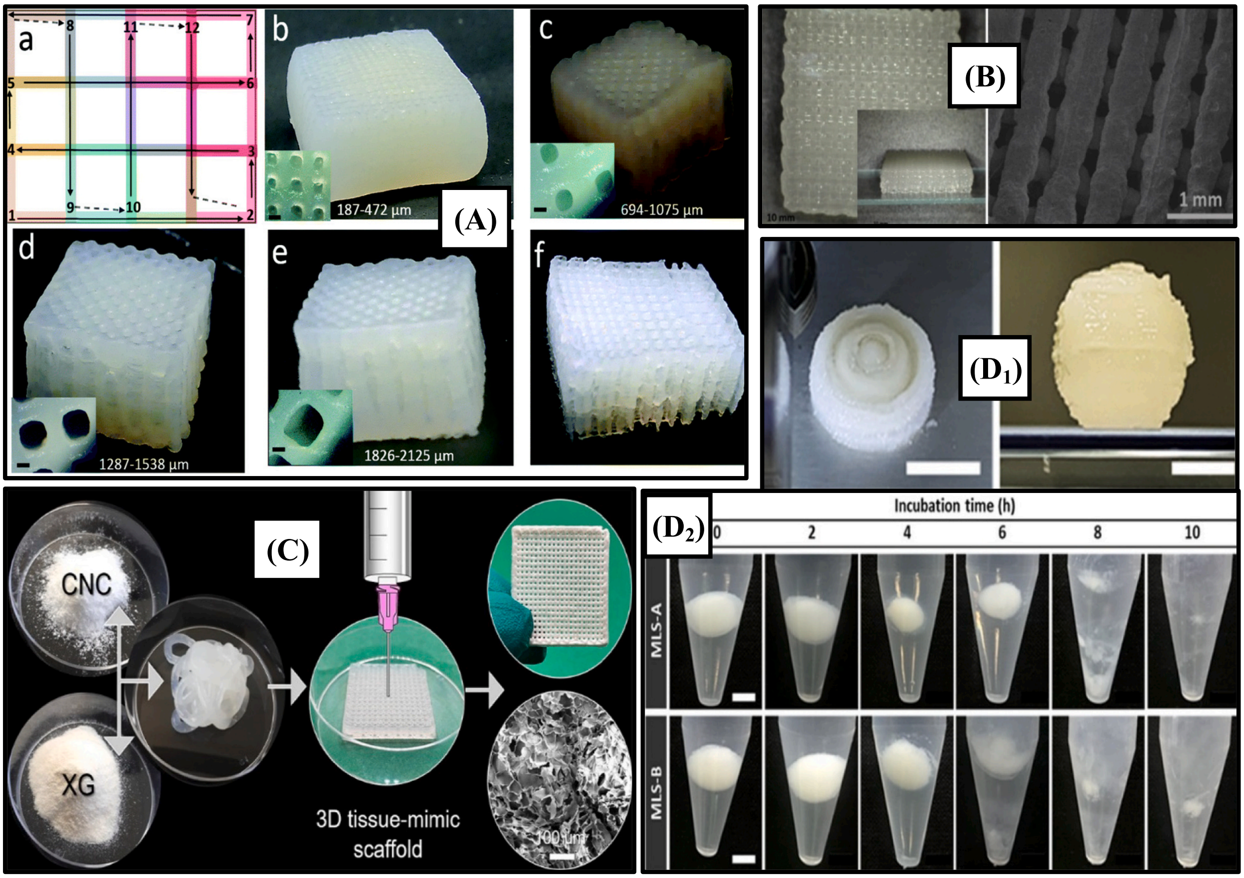

Fig. 11. (A) Schematic representation of nozzle movements applied for developing 3D-printed hydrogels. CNC/gelatin/SA-based scaffolds with different pore sizes (adapted from [308], under the Creative Commons Attribution license); (B) Optical and microscopic images of 3D-printed PCLA-PEG-PCLA/CNC-based bioscaffolds (adapted from [291], under Creative Commons Attribution license); (C) Schematic diagram depicting steps involved to develop 3D-printed scaffold and its morphology (adapted from [304],under the Creative Commons CC-BY license); (D_1) Alginate/CNC/$CaCl_2$-based MLS fabricated by using 3D printing, (D_2) Morphological changes of MLS in simulated gastrointestinal fluid (adapted from [325],copyright 2021 Elsevier Ltd.).

excellent mechanical properties, which maintained its shape with high precision and resolution. Additionally, the incorporation of CNCs significantly improved the printability of hydrogel-based scaffolds.

In another novel work, Baniasadi et al. [304] used xanthan gum (XG)/CNC bioinks for developing different 3D geometries of soft tissue scaffolds by using the DIW technique. The addition of CNC fillers significantly improved the overall printability of bioinks. Furthermore, the printed samples showed fair resolution and high fidelity without any deformation after printing, as presented in Fig. 11(C). Additionally, these CNC-based composite scaffolds demonstrated excellent swelling of approximately 11 g/g, which helped in oxygen and nutrient transportation.

Due to biodegradability, biocompatibility, renewability, and non-toxic properties, CNC-based biomaterials are excellent candidates for drug delivery applications in which 3D-printed structures have been applied for pharmaceutics. For instance, Yoon et al. [325] used CNCs and calcium chloride as support nanomaterials for enhancing the rheological properties and printability. They fabricated alginate-based multi-layered spheres (MLS) by using droplet-based 3D printing, as illustrated in Fig. 11(D). The results revealed that these bovine serum albumin-loaded spheres exhibited sustained and controlled release in the intestine. Thus, this platform can be used for personalized oral delivery with tailorable release patterns.

CNC-based auxetic structures show lateral expansion during elongation upon insertion and can be used for fabricating biodegradable biomedical devices. For instance, Pruksawan et al. [323] used CNC as a multifunctional macro-crosslinking agent to produce stretchable and tough hydrogel for developing oesophageal self-expandable stent, as depicted in Fig. 12(A). The results revealed that methacrylated cellulose nanocrystals (CNCMA)/poly(2-hydroxyethyl methacrylate) (pHEMA)-based soft auxetic structure printed through the SLA technique exhibited excellent stretchability and toughness.

CNC-based biomaterials have also shown tremendous perspectives in developing smart medical sensors, due to their ability to exhibit high piezoelectric responses [335]. For instance, Maity et al. [317] developed pyro- and piezoelectric nanogenerators (Py-PNGs)/CNC-based cardio-respiratory monitoring sensors for personal healthcare, as illustrated in Fig. 12(B). DIW-printed architecture exhibited excellent durability and sensitivity. Therefore, real-time cardiorespiratory monitoring through smart sensors offers fascinating and notable information in biomedical diagnosis, and the world is stepping towards the development of novel biomedical devices and human-machine interfaces. This distinct approach can be used to print structures from renewable energy sources for electric energy generation and temperature sensor applications. In another study, Binelli et al. [324] proposed a 3D printing platform for developing silicone/CNC/carbon black-based soft wearables with integrated piezoresistive sensors, as illustrated in Fig. 12(C). The results revealed that the placement of sensors into personalized insole shoe helped in monitoring the shear and normal pressure of human gait.

4.2. Soft robotics

Nowadays, soft robotics is growing manifold, where the robots are fundamentally soft and elastically deformable [336]. The construction of soft robots comprises a motor-less robot driven from smart materials typically responsive polymers which can change their shapes under a

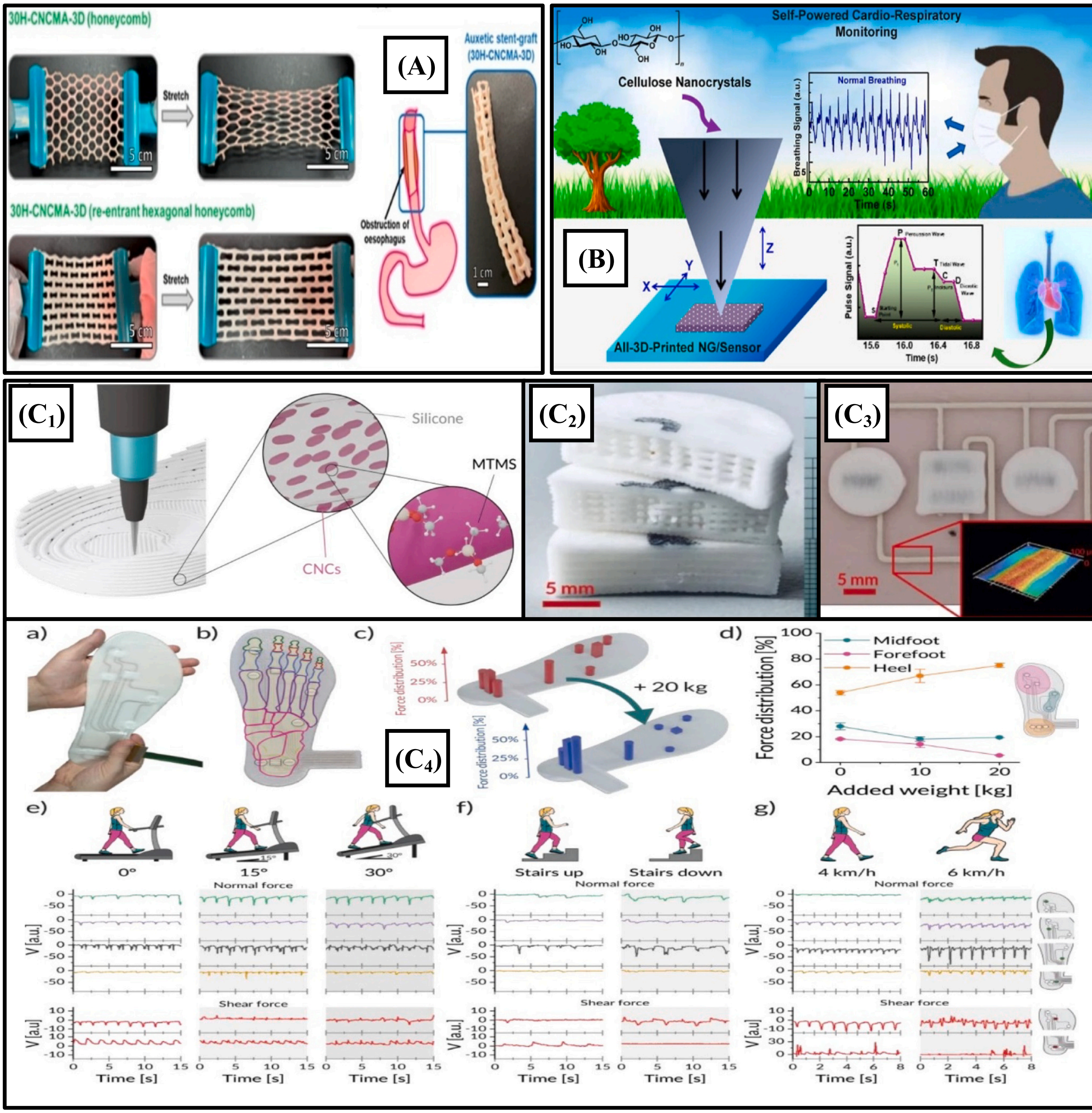

Fig. 12. (A) CNCMA/pHEMA-based honeycomb self-expandable stent developed through SLA for obstruction of oesophagus (adapted with permission from [323], copyright 2022 Wiley-VCH GmbH); (B) Schematic representation of the 3D-printed CNC/Py-PNGs developed through DIW for healthcare systems (adapted with permission from [317], copyright 2023, American Chemical Society); (C_1) Schematic illustration of CNC/silicone-based ink for DIW printing; (C_2) Grid-type architecture with varying infill densities; (C_3) 3D reconstruction of printed silver connector on a substrate; (C_4) 3D-printed personalized insole with integrated sensors and their response for different physical activities (adapted from [324], under the Creative Commons license).

stimulus. Soft robotics pave the foundations for various novel applications such as actuators, soft grippers, and many biomedical devices [337–339]. CNC-based stimuli-responsive materials have been extensively applied to develop smart and complex grippers, actuators, and robots for different engineering applications [275]. For instance, Fourmann et al. [292] printed self-supporting complex structures of PNIPAM/CNC-based hydrogels by using a DIW technique. The angles and textures of complex architectures, as well as programmable self-shape actuation of hydrogel ink were possible, due to the alignment of CNC-based ink. Furthermore, hydrogel structures demonstrated shape-morphing behaviour such as bending or twisting upon a hydration stimulus, as presented in Fig. 13(A). Such complex 3D structures with programmable actuation and anisotropic swelling properties can also be used in wound healing applications.

In another study, Müller et al. [277] developed intricate structures using a polyurethane acrylate (PUA) matrix with up to 15 wt% CNC reinforcement through a DIW printing. Azobenzene photochromes were grafted onto the CNC surfaces for developing photo-induced responses and printed materials presented a shape memory behaviour when it reacted to visible-light irradiation with 30–50 % reversible softening, as presented in Fig. 13(B). Additionally, the dynamic mechanical responses of 3D complex structures under environmental stimuli were due to

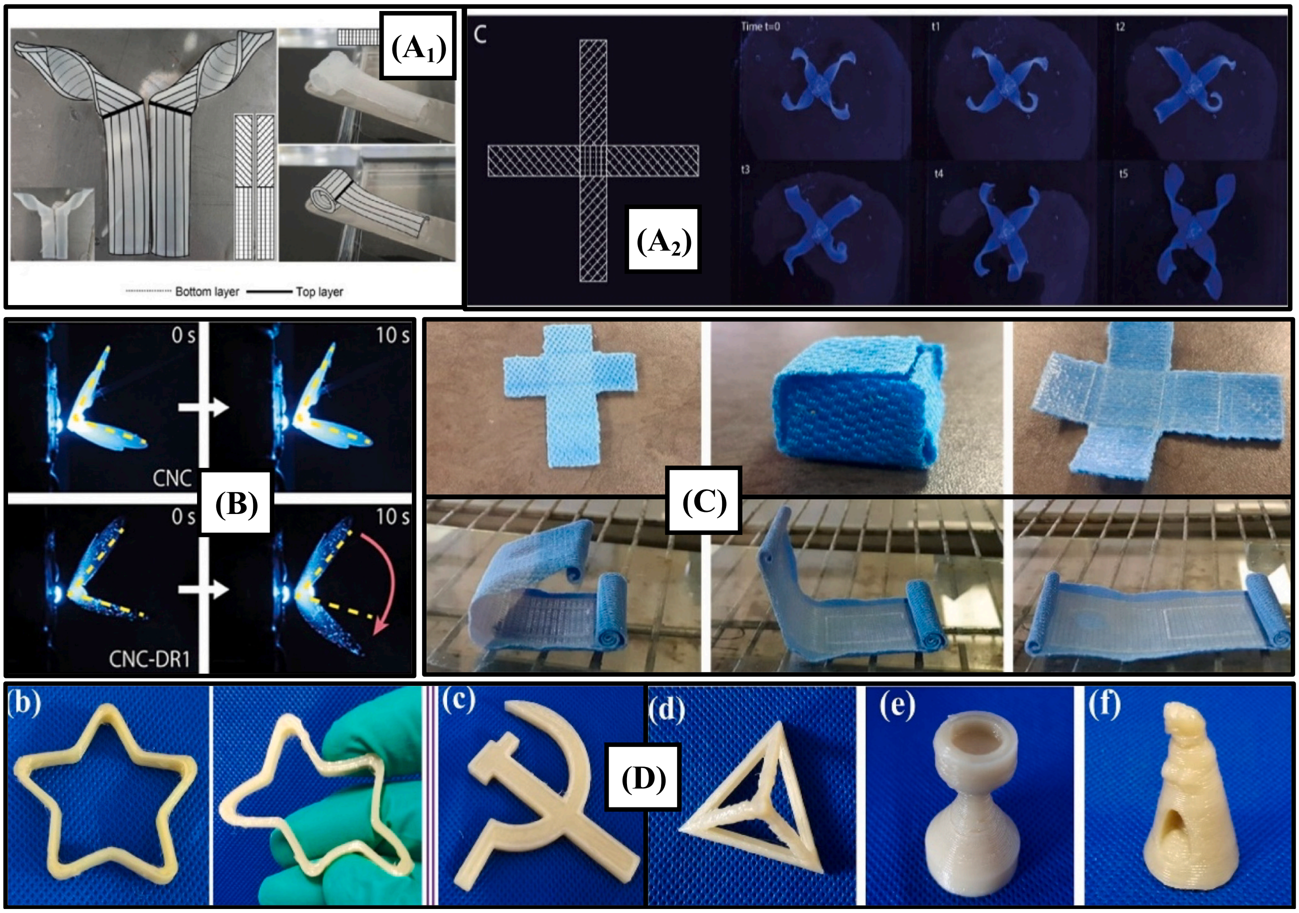

Fig. 13. (A$_1$) CNC/CNF/PNIPAM hydrogels-based 3D bilayer structure with swelling and anisotropic actuation behaviours, (A$_2$) Two bilayer strips of nanocellulose-PNIPAM hydrogels (20 wt% CNC) produced by 3D printing leading to synthetic structures that twist (adapted from [292], under Creative Commons license); (B) Shape memory effect of printed butterfly w.r.t time under illumination stimulus (adapted with permission from [277], under the Creative Commons CC-BY license); (C) CNC/PLA-based thermo-responsive folded shape memory object for wearable electronics (adapted from [290], under Creative Commons Attribution license); (D) (b) 3D-printed pentagram polygon model under loading; 3D-printed of (c) emblem model; (d) Four-node ball; (e) Chess piece; (f) Cartoon character (adapted with permission from [283], copyright 2021, Elsevier B.V.).

phototunable energy absorption. Thus, the printed structures with CNC reinforcement are promising for those areas in which dynamic responses to environmental changes are required.

4.3. Wearable electronics

Wearable electronics are considered built-in sensors, which are widely used for tracking the positions as well as movements of various body parts [340]. Recently, versatile smart wearable electronics such as a smartwatch, chips on body parts for joint movements, glucose level tracking, and blood pressure monitoring have been employed [341]. However, their effectiveness through traditional techniques is quite limited and now, wearable electronics are developed by using 3D printing of sustainable biomaterials especially CNCs, which allows better control over the exciting characteristics such as super flexibility, high stretchability, ultra-thinness, and lightweight feature for healthcare systems [342]. For instance, Bi et al. [283] printed TPU/PCL-based composites by using modified CNC as a crosslinking agent. The results revealed that these composites exhibited excellent printability and flexibility. Furthermore, CNC-based 3D-printed composites possessed excellent lap shear strength (12 MPa), high thermal stability, efficient self-healing ability, and good shape memory behaviour. Fig. 13(D) incorporates different 3D-printed models developed by using TPU/PCL/CNC-based composites. These composites are excellent candidates for developing wearable electronics, thanks to their excellent self-healing, shape-memory, and rheological properties.

4D printing technology has further groomed and revolutionized the wearable electronics sectors due to its multifarious peculiarities including excellent shape-deformation behaviour under body stimuli such as pH, humidity, and temperature, which accurately predict the body response [343–345]. For instance, Agbakoba et al. [290] developed 3D-printed CNC/PLA-based foldable cube and grid fabrics with excellent shape memory behaviour, as illustrated in Fig. 13(C). Additionally, the results revealed that specimens exhibited excellent tensile strength, modulus, and elongation at break. Such a heat-triggered SMP fabric can be used for wearable electronics.

4.4. Anti-counterfeiting

Novel applications of CNC-based stimuli-responsive materials in anti-counterfeiting are due to their response under pH, light, heat, and electrical stimuli, which is also promising for other ground-breaking applications [346–348]. For instance, Cheng et al. [349] printed flexible photonic hydrogel chromatic patterns by using CNC-based inks. The various developed patterns such as QR codes, hydrogel colour cards, and Tai-chi models were visible under polarized light, as presented in Fig. 14 (A). Additionally, the brightness of hydrogels can be controlled by varying the number of layers and the printing angle. This 3D-printed photonic hydrogels with chromatic patterns visible in polarized light has potential use in information hiding, security making, and anti-

Fig. 14. (A) Photonic hydrogels under polarized light with (a–c) Tai-chi pattern and (d–f) QR code (adapted with permission from [349], copyright 2022, Elsevier B. V.); (B) (a–j) Anti-counterfeiting patterns printed by using through different printing parameters. WHU patterns under natural light in (a) and polarized light in (b-d) (adapted with permission from [326], copyright 2021, Royal society of chemistry); (C) Steps involved for transforming raw food into CNC-based self-supporting edible structures developed through 3D printing (adapted with permission from [93], copyright 2022, Elsevier Ltd.); (D) CNC-reinforced photo-responsive architectures, which can be used for energy absorption applications (adapted with permission from [276], copyright 2020 WILEY-VCH); (E) (a) 3D-printed and crosslinked NFC/alginate/$CaCO_3$ flat and twisted sheets at various angles using gluconolactone (Gdl), crosslinked induced with $CaCl_2$; (b) 3D-printed and Gdl-crosslinked tube stability test; (c) 3D-printed and Gdl-crosslinked tube compressed with the tweezer and released over a period of 60 times; (d) $CaCO_3$ NPs/Gdl-crosslinked models (adapted from [351], under the Creative Commons CC BY license).

counterfeiting applications. Additionally, surface functionalization of CNCs with luminescent inks such as quantum dots, fluorophores, or carbon NPs offers tremendous potential in sensing, bio-imaging, nanoparticle tracking, and anti-counterfeiting applications [350].

In another study, Li et al. [326] successfully printed optical anti-counterfeiting patterns by using surface functionalized CNC/organosilane/polyoxyethylene ether (NPES) ink, which was developed by sequential modification of CNC with NPES and organosilane. The results revealed that the ink showed good flowability under shearing force and transformed into a gel-like phase. Additionally, the optical properties and texture of 3D-printed patterns were controlled by tailoring the printing parameters. Moreover, these patterns were transparent under light, but, exhibited vivid interference colour, as illustrated in Fig. 14 (B). Thus, depicting anti-counterfeiting characteristics between crossed polarizers.

4.5. Miscellaneous applications

Besides, wearable electronics, soft robotics, anti-counterfeiting, and biomedical applications, various stand-alone studies have used CNCs through 3D/4D printing technology for producing a wide range of edible food coatings, apparel architectures, and self-supporting and complex structures, thanks to their biocompatibility, non-toxicity, and biodegradability [352–354]. In recent years, AM technology has been extensively applied in the food industry to develop edible food printing by using extrusion-based printing techniques especially, DIW. Edible 3D printing has the potential to offer appealing meals with personalized nutrition profiles [355]. CNC can be used to incorporate shear thinning properties and viscosity of printable inks [356]. For instance, Armstrong et al. [93] employed CNCs as a renewable and safe rheological modifier, which was capable of enabling DIW printing of different foodstuffs including puree, tomato puree, spinach, and applesauce, and finally, freeze-dried to obtain the self-supporting edible structure, as illustrated in Fig. 14(C). The results from this research revealed that CNC-laden inks offered the capability to develop multi-material structures with integrated packaging. Additionally, CNCs were excellent rheological modifiers, which promoted shear thinning behaviour in the edible feedstocks.

CNC-based biomaterials can also be used to develop load-absorbing personalized devices and self-adapting structures [357]. For instance, Müller et al. [276] developed a CNC-reinforced (30 wt%) photo-responsive intricate energy absorption device by using multi-material printing, as illustrated in Fig. 14(D). The results revealed the spatially controlled illumination permitted tunable compression, Young's modulus, stiffness, and other mechanical properties of 3D-printed structures. In another study, Lackner et al. [351] printed various nano/micro-scaled 3D structures with internal radial chiral. Reconfiguration of the chiral arrangements was done in the CNC-based suspensions through exploited shear forces in the extrusion-based 3D printing. Furthermore, the resolution of the 3D printing technique was excellent in terms of the voxel size and from tunable nano/micro scale (in dry/wet structure) in chiral molecules. Reactive monomers were incorporated into the chiral inks to produce a polymer network and arrest the chiral structures. Different 3D-printed architected materials with structural orders spanning multiple length scales, as presented in Fig. 14(E).

In summary, 3D printing helps to fabricate 3D structures by using CNC-based sustainable materials for numerous applications not limited to tissue engineering, wound healing, bio-sensors, actuators, edible foods, and optical devices. Additionally, the combination of CNC-based stimuli-responsive materials and 3D/4D printing has further helped to develop intricate and tunable structures with complex responses suitable for biomedical, actuating, and sensing applications.

5. Summary and future perspectives

Sustainability considered as use of renewable raw resources in place of petroleum-based components [358]. 3D/4D printing used minimal material consumption for preserving sustainable environment [359]. Thus, potential renewable feedstock of 3D printing, plant fibers typically contain lignin, hemicellulose, and cellulose, waxes, pectin, and water-soluble organic compounds (up to 20 %). AM of CNC-based renewable and green materials is an important research arena with enormous potentials. The rise of 3D printing and CNC-based materials team up and permits harnessing the green electronics, smart sensors, biodegradable medical scaffolds, drug delivery systems, edible coatings, and optical devices [360,361]. This review focuses on the importance of developing complex structures by using CNC-based sustainable and stimuli-responsive materials. Moreover, CNC-based materials have extreme sensitivity to particular stimuli, their self-healing and self-repairing capabilities thriving them as a futuristic and sustainable 4D printing technology [362].

Additionally, these toxic-free materials offer biocompatibility, biodegradability, and shape-morphing characteristics. The major future direction is the development of resource-efficient and eco-friendly sustainable structures for myriad applications by using modern manufacturing methods. Fig. 15 depicts the futuristic view of additively manufactured CNC-based green materials. However, there is a need to address some challenges that are hindering the progress of additively manufactured CNC-based structures.

The use of CNCs for developing inks for 3D/4D printing is limited, due to their solubility in toxic mixtures of solvent. Such solvent traces can be removed by using advanced purification techniques or by synthesizing CNCs with controlled shapes and dimensions. 3D printing of CNC-based stimuli-responsive materials opens up unlimited prospects to develop functional materials and devices, which exhibit intricate shapes such as soft robotics and biomimetic devices [363]. Despite their ability to fit in traumatized tissues, due to their shape-morphing nature, additively manufactured CNC-based structures still lack sufficient control over programmed shapes and precise responses, which limits the development of micro-scaled smart grippers and actuators.

The alignment of anisotropic NPs during the deposition of ink directly influences the mechanical properties and microstructure of extrusion-based architectures [364]. Although, the CNC orientation in diluted suspensions has been well understood through numerical and analytical models. However, there is a need to fully explore the dynamics of CNC orientation in the bioinks developed for DIW printing. Additionally, the development of 3D structures with CNC-based inks is in the nascent stage, and CNCs have been mostly used to print macro-scale products like tissue scaffolds and bio-mimetic shape morphing systems [365]. However, there is a need to develop CNC-based inks for micro-printing, as it is more challenging to fabricate micro-scaled structures by using nanocellulosic materials. It will allow the research to explore new functional materials and tunable properties.

The development of CNC-based materials has made inspiring designs for 3D/4D printing. To provide novel and sophisticated structure designs, particularly for CNC-based smart responsive materials, various computation tools as well as machine learning algorithms can also be employed for solving a variety of real-world engineering and industrial problems mainly in the micro and nanoscale realms [366].

This is worth mentioning that synergism of CNC in various biopolymers such as PLA, PVA, PBAT, polyvinyl acetate etc. is regarded as an industrial application of CNC through simplistic 3D printing process [367]. Thus, biopolymer/CNC materials through 3D printing will compete for high-performance requirements of strength with high dimensional accuracy in the hunt for a sustainable environment [368]. Nowadays, various emerging 2D materials have enormous prospects in 3D printing. In this regard, the synergistic effect of CNC with 2D materials can further improve its binding properties for the achieving of well-dispersed colloidal suspensions. For instance, CNC can be attached to CNTs and dispersed in an aqueous solution eliminating agglomerations and highly liquid crystalline alignment CNC/CNTs bonding within a polymer network for achieving the remarkable mechanical properties of CNC-based 3D-printed structures [186].

CNC-based hydrogel ink can be used as a potential material for uniform printing. Various hydrogels such as GelMA, PNIPAM, SA, and PEGDA are potentially being applied for CNC to improve its shear thing behaviour, self-healing properties, and various mechanical and physical properties [369]. This is crucial for printing various smart structures including gradient porous scaffolds and many smart actuators [281]. Moreover, numerous surface treatments such as dopamine coating improved the binding capability of other bioactive materials such as the growth factor of isolated stem cells and the hydrophilicity of 3D-printed CNC scaffolds for opening new avenues in tissue engineering applications [370]. The surface-modified CNCs acted as macro-cross-linkers, which improves the structural integrity and mechanical stability of

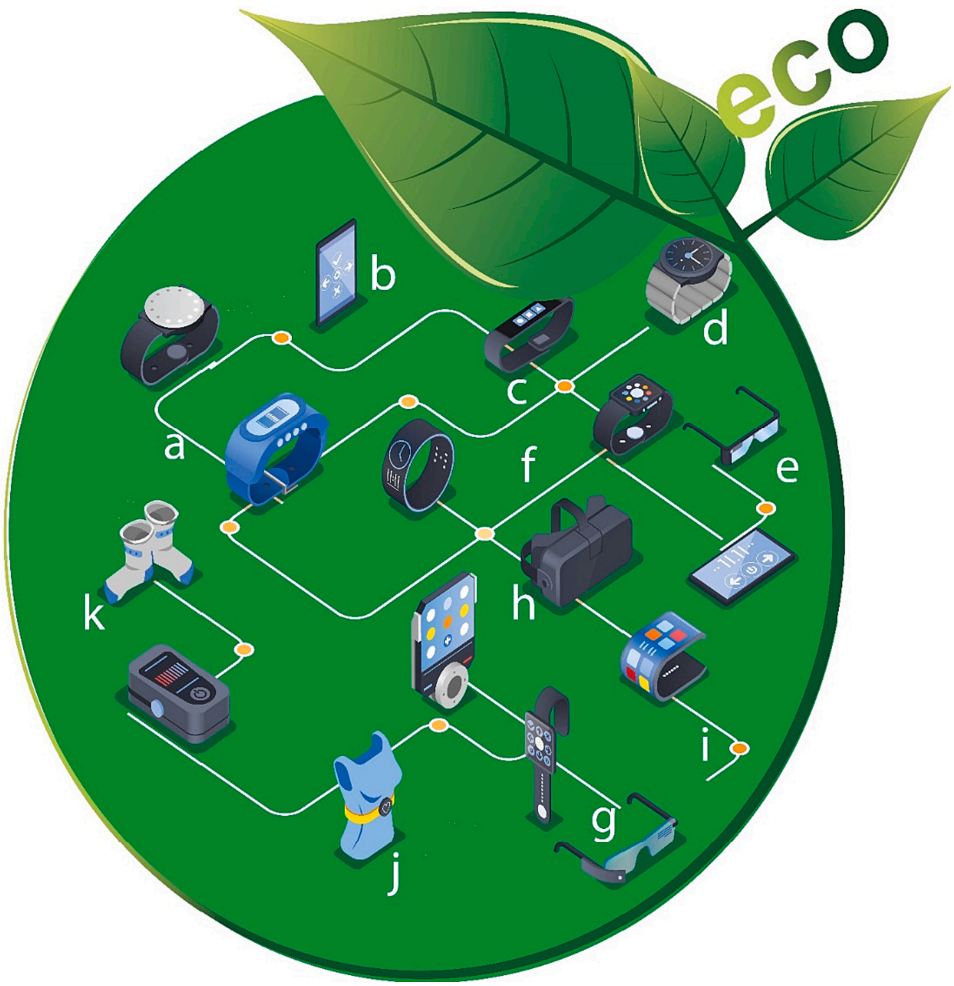

Fig. 15. Futuristic view of CNC-based materials in achieving the novels concept of "green electronics " by using 3D/4D printing technology such as (a) Fitness tracker, (b) Entertainment, (c) Fitness tracker, (d) Smart watch, (e) High polarized goggles, (f) Highly promising working environment, (g) Augmented reality, (h) VR technology, (i) Smart screen, (j) Smart cloths, (k) Sports technology.

3D-printed CNC/hydrogel structures with tannable self-healing properties. Also, functionalization techniques such as benzophenone moieties, azobenzene or grafting coumarin on the CNC particle's surface, improved its localized photostiffening. This unique function promotes cinnamate moieties on the CNC surface for successfully used in 3D printing of CNC-based nanocomposites [371].

Tunable colour metasurfaces that show changes in their optical properties under an external stimulus are also considered promising potential areas [372]. CNC-based photonic structures manufactured through 3D printing for anti-counterfeiting applications is at the infant stage. There is a need to develop CNC films with the ability to conceal information under light by enhancing stokes scattering. It has been demonstrated that by removing the solvent from a suspension of the crystals, CNCs can form a chiral nematic nanostructure and the developed solid film can maintain the chiral nematic structure and demonstrate excellent structural colour [373]. However, it is found that these materials cannot be reversibly dispersed in most solvents after forming solid films, due to their strong hydrogen bonding, which leads to unsatisfactory recyclability and processability.

Finally, CNC-based stimuli-responsive biomaterials combine the advantages of both external stimuli and CNCs to develop 3D smart structures for biomedical, soft robotics, electronics, edible food printing, and anti-counterfeiting applications. However, there are some limitations associated with 3D printing of CNC-based biomaterials including insufficient response accuracy and long response time. To date, CNC-based stimuli-responsive materials are limited to laboratory experiments. There is a need to promote practical applications by overcoming the limitations associated with these materials. CNC-based stimuli-responsive biomaterials are anticipated to make significant progress in practical applications, in the coming years.

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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.

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