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REVIEW

Sustainable prospects of lignocellulosic wood and natural fiber-based materials in 3D and 4D printing

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Abstract

The sustainable options for 3D/4D printing, utilizing lignocellulosic materials derived from wood and natural fibers, have gained significant attention in the pursuit of building a greener and more environmentally friendly world. As environmental concerns continue to grow, there is an increasing focus on greener materials and manufacturing processes. The utilization of these environmentally friendly alternatives as substitutes for synthetic fiber filled polymer matrices in 3D/4D printing is driven by the objective of enhancing the material properties of printed items while reducing material costs. 3D/4D printing, also known as additive manufacturing, represents a promising frontier in environmentally friendly manufacturing. Recent progress and advancements in 3D/4D printing technology have expanded its capabilities beyond prototyping to the rapid fabrication of finished goods. This review explores the characteristics, processing techniques, mechanical and physical properties, applications, and future possibilities of sustainable 3D/4D printed products developed from wood and natural fibers. Architectural design and polymer selection have the potential to yield materials with improved functionality, mechanical characteristics, porosity, and stability. Additionally, the multifunctional polymer-based 3D/4D printing product development has enabled the production of biomedical devices, electrical products, and aerospace-related items. The challenges associated with utilizing these products for large-scale production and other aspects of sustainable 3D/4D printing will also be discussed, along with recommendations for future solutions. Overall, this work offers valuable insights that can guide future research, development, and implementation of lignocellulosic-based 3D/4D printed composites. By exploring the potential of these sustainable materials in 3D/4D printing, it contributes to the advancement of environmentally friendly manufacturing practices and promotes the adoption of greener alternatives across various industries.

Keywords Sustainable 3D/4D printing · Lignocellulosic materials · Wood and vegetable fiber · Functional properties · Processing techniques · Applications and future perspectives

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Abbreviations

ABS	Acrylonitrile butadiene styrene
AM	Additive manufacturing
ASTM	American Society for Testing and Materials

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BTC	Bend-twist coupling
BJ	Binder jetting
CAD	Computer-aided design
CFD	Computational fluid dynamics
CTE	Coefficients of thermal expansion
DED	Directed energy deposition
DMLS	Direct metal laser sintering
DSC	Differential scanning calorimetry
EBM	Electron beam melting
FDM	Fused deposition modeling
FFF	Fused filament fabrication
HBC	Hygromorph BioComposite
MJ	Material jetting
MJF	Multi-jet fusion
NFRC	Natural fiber-reinforced composite
PLA	Polylactic acid
PU	Polyurethane
PBF	Powder bed fusion
PA	Polyamides
PET	Polyethylene terephthalate
PEEK	Polyetheretherketone
PHB	Poly(3-hydroxybutyrate)
SLM	Selective laser melting
SL	Sheet lamination
SLA	Stereolithography
SLS	Selective laser sintering
TGA	Thermo gravimetric analysis
2PP	Two-photon polymerization
UV	Ultraviolet
VP	Vat photopolymerization
WF	Wood flour
3DP	Three-dimensional printing
4DP	Four-dimensional printing

1 Introduction

Advanced manufacturing of 3D and 4D printed products has emerged as a cutting-edge innovation with the potential to disrupt a variety of sectors, particularly production, health care, architecture, and design. The precision and efficiency with which complex 3D/4D objects can be created have captured the attention of researchers, engineers, and entrepreneurs [1]. However, the widespread adoption of 3D/4D printing has raised concerns about its environmental impact, particularly regarding the sourcing and processing of materials such as polymers. Therefore, to make 3D and 4D printed products more sustainable, there is increasing attention on using lignocellulosic materials, such as natural fibers and wood-based substances, to replace traditional non-biodegradable materials [2]. Natural fibers are increasingly becoming popular as fillers and reinforcement materials in plastic products due to their high specific stiffness, processing flexibility and reduced volumetric cost. Compared to other natural fibers, woods are gaining popularity because of the presence of lignocellulosic polymeric compounds like cellulose, hemicellulose, and lignins, which contribute to their biodegradable properties [3]. Wood is a biological material used in engineering applications [4], characterized by an intricate hierarchical organic composition consisting of cellulose fibrils interwoven inside a matrix composed of hemicelluloses and lignin [5]. The incorporation of biobased lignocellulosic materials in composites also enhances their mechanical, electrical, acoustic insulating, and thermal qualities, as well as their fracture resistance [6]. In contrast, traditional non-biodegradable polymeric compounds used in 3D/4D printing technologies often fail to achieve the sustainable development objectives, such as reducing greenhouse gas emissions, minimizing environmental pollution, promoting renewable energy, and supporting a sustainable circular economy, set aside by the United Nations, it is essential to take appropriate measures [7]. To address these concerns, researchers and innovators are exploring sustainable alternatives for 3D/4D printing materials [7–9]. One promising avenue of investigation involves exploring lignocellulosic wood and natural fiber-based materials as the sustainable alternatives to conventional petroleum-based plastics [5, 10]. Lignocellulosic materials, derived from plant biomass, offer numerous benefits, including renewability, biodegradability, and a minimal environmental impact, making them an appealing choice for sustainable 3D/4D printing products [10]. In this regard, Soleimanzadeh et al. developed a sustainable biodegradable 4D printed robot [7]. The developed robot offers a notable functional advantage to the field of soft robotics, particularly in tasks that necessitate intelligent shape modification in response to environmental factors such as light, pH, and heat. This capability allows the robot to adapt and interact with its surroundings in a dynamic and responsive manner [7]. The study also highlights the feasibility of directly fabricating complex and functional soft robotics through employing 3D and 4D printing of different soft and multi-materials. In another study, Agrawal et al. developed natural fiber-reinforced acrylonitrile–butadiene–styrene (ABS) filaments tailored for 3D and 4D printing applications [8]. The fibers incorporated included banana, orange, cinnamon, and neem leaf fibers [8]. The study focused on evaluating the mechanical, rheological, and shape memory properties of the resulting composites. The inclusion of fibers enhanced the shape memory performance of ABS while also reducing its viscosity, making it more suitable for advanced printing techniques [8].

Moreover, there is a worldwide increase in both the production and consumption of natural fibers [11]. The production of natural fiber in the world is expected to be 32.7 metric tons by 2024, which was nearly one million metric tons less in 2023 [12]. To address environmental concerns and promoting greener composite material development, the utilization of 3D/4D printed materials based on plant fibers, woody materials, and biopolymers holds significant potential. It is projected that the annual growth rate of biodegradable material consumption will reach approximately 13% [13]. In Fig. 1a, b, the recent research progress of 3D/4D printed wood lignocellulose or natural fiber reinforced composite is presented. The data has been collected by generating this code in the Web of Science database ((“3D printing” or “4D printing” or “Additive manufacturing”) AND (“wood” or “wood lignocellulose” or “wood composite” or “natural fiber composite” or “natural fiber reinforced composite”)) and selecting year (2013–01-01 to 2023–12–31). This investigation found that a total of 614 technical research articles have been published in this domain. According to Shah, biobased fibers have the potential to represent up to 28% of the reinforcing materials market by 2020 [14]. The projected global production of natural fibers for the year 2022 is expected to reach 33.3 million tons, showing an increase of nearly 200,000 tons compared to the previous year [15]. This figure exceeds the forecast made by the Discover Natural Fibres Initiative (DNFI) in June 2023 by more than 1 million tons [15]. Furthermore, the production of wood and wood fiber has also witnessed an upward trend. Another study claimed that the use of 3D printing technology has demonstrated its effectiveness in reducing construction waste by up to 60%, decreasing manufacturing time by 70%, and significantly reducing labor expenses by 80% [13]. Therefore, both 3D and 4D technologies are becoming popular significantly. Additive manufacturing (AM) technology, also known as 3D/4D/5D printing, is a comparatively new technique that involves the computerized design of 3D/4D/5D components with precise measurements, using spatial or layer-by-layer deposition [16]. It reduces the necessity for traditional tools such as lithographic masks, molds, and dies. This modern manufacturing method enables the production of structurally complex objects on demand, at affordable cost, and in an extensive range of configurations and materials [17]. These unique characteristics make additive manufacturing a leading-edge technology. Recently, 4D printing has acquired substantial attention from academia and industry due to its

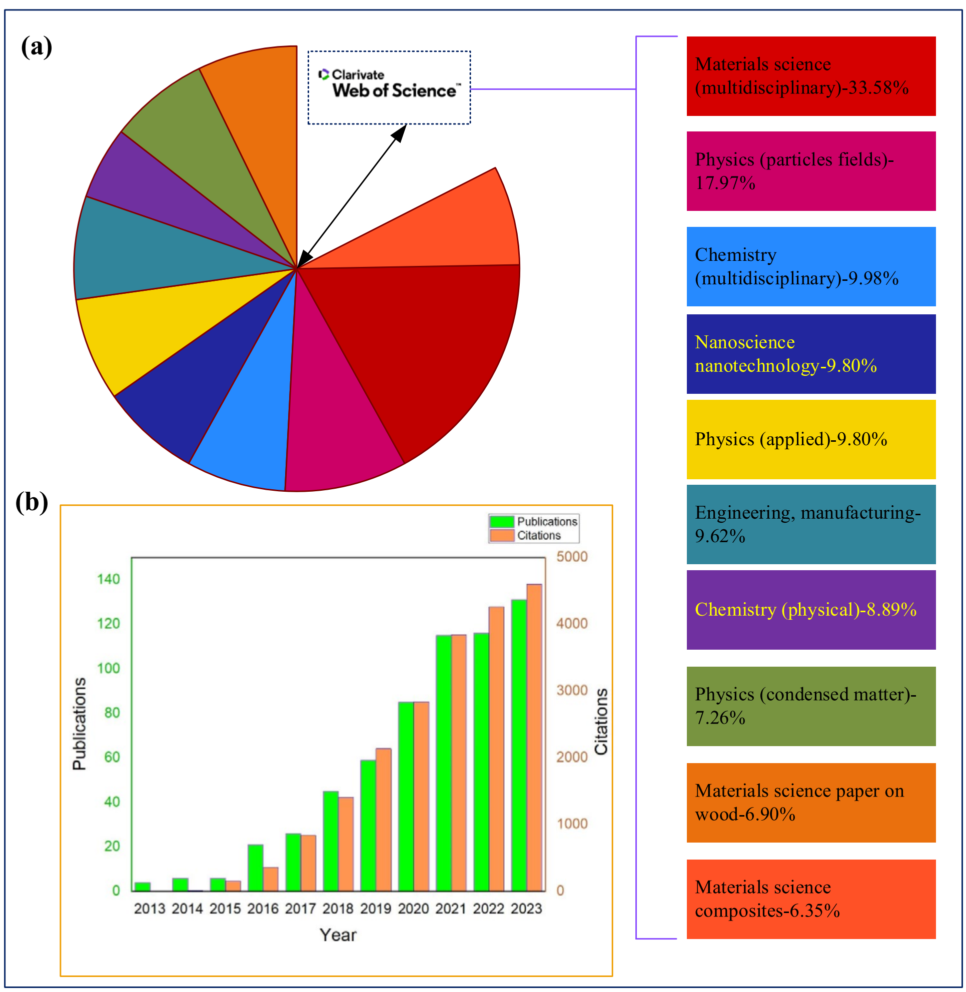

Fig. 1 **a** Recent research progress on different disciplines in 3D/4D printed woody lignocellulose and natural fiber reinforced composite materials; **b** bar chart developed from ten years of publications, and citation numbers data collected from the Web of Science

rapid development, stringent requirements, and innovative potential [18]. 4D printing entails the utilization of intelligent materials capable of deliberately and precisely altering their shapes or morphologies as time progresses. This adaptability makes structures, devices, and other objects created through 4D printing appear smart and responsive [19]. Materials such as polymers, ceramics, and metals find application in 4D printing, creating smart structures and delivering desired responses [20]. Among these, shape memory polymers (SMPs) stand out as highly appealing for 4D printing, owing to their extraordinary capability to revert back to their original shape from deformed states when exposed to external triggers. Similar to 3D/4D printing, 5D printing is another method of additive manufacturing used primarily to produce curved or concave-shaped layers with superior accuracy and precision. However, the main difference between these methods is that 5D printing allows the printing head and bed to move at any angle [21]. In Fig. 2a, the various types of manufacturing in 3D and 4D printing are demonstrated. In addition, in Fig. 2b, a comparative study between the 3D and 4D printing is presented. Figure 2c depicts the revolution of 3D, 4D, and even 5D printing.

This work incorporates a detailed analysis of recent studies that evaluate the mechanical, thermal, and surface properties

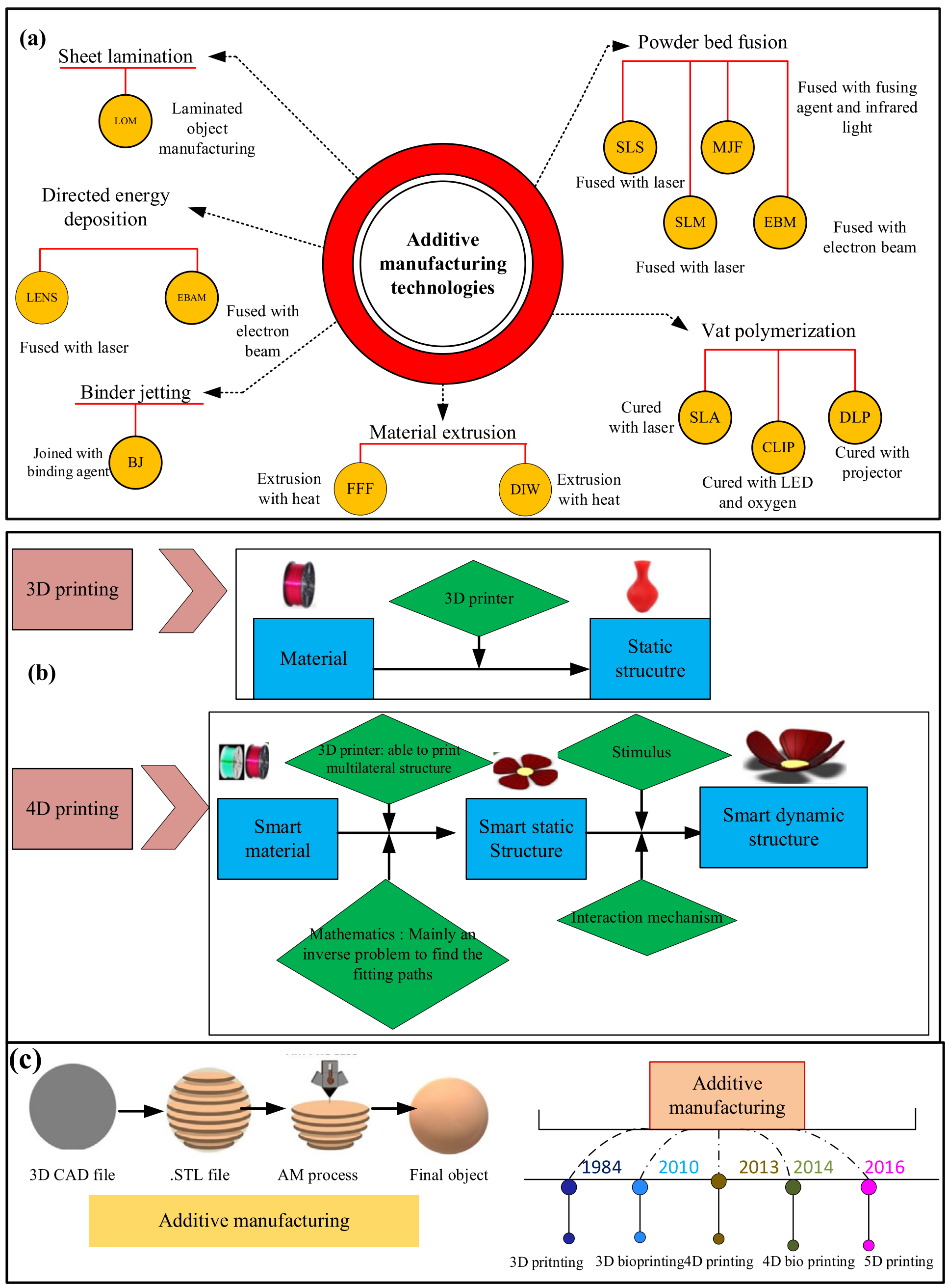

Fig. 2 **a** Manufacturing processes and types of 3D and 4D printing method [22], **b** differences between the 3D and 4D printing [16], and **c** diagram depicting the flow and development of the AM process throughout time [23]

of 3D and 4D printed materials derived from lignocellulosic wood and natural fiber-based materials. Although there are numerous studies on 3D/4D printing, the uniqueness of this research lies in its valuable insights into the performance and suitability of such materials for a wide range of advanced applications. It showcases state-of-the-art innovation, particularly focusing on lignocellulosic wood and natural fiber-oriented 3D/4D printing materials.

The introduction of this work focuses on the needs and different aspects of lignocellulosic wood and natural fiber-oriented 3D/4D printing materials, highlighting their advantages (Sect. 1). Section 2 provides a comprehensive review of lignocellulosic woods and natural fibers, including their specifications. The materials utilized in the production of various lignocellulosic wood and natural fiber-based 3D/4D printing materials are discussed in Sect. 3. Section 4 delves into the different methods employed for sustainable 3D/4D composite product manufacturing, including material extrusion, vat polymerization, and powder bed fusion. This section also includes a comparison of these methods, along with discussions on their challenges and possible solutions. In Sect. 5, the potential of nanotechnology for the development of advanced and smart lignocellulosic wood and natural fiber-oriented 3D/4D printing materials is addressed. Section 6 focuses on the thermomechanical and physical properties that determine the specific usability of sustainable composites. The subsequent section (Sect. 7) explores various applications, such as structural, biomedical, aeronautical, and electronics, demonstrating the versatility and wide-ranging possibilities offered by these 3D/4D printed materials. Section 8 discusses the challenges associated with lignocellulosic wood and natural fiber-oriented 3D/4D printing products, including sourcing, processing, scaling up of production, and offers potential solutions to overcome these obstacles. Finally, this work is concluded by outlining the many potential avenues for future research related to sustainable 3D and 4D printed products that are not yet widely explored (Sect. 9). Additionally, the review highlights potential applications in fields such as architecture, aviation, product design, biomedical engineering, defense, microfluidics, and sustainable consumer goods. The discussion emphasizes the economic importance, opportunities, future perspectives, and the environmental benefits presented by these materials. By consolidating existing knowledge and research in this rapidly evolving field, this comprehensive work aims to provide valuable insights to researchers, practitioners, and policymakers. It explores the potential and constraints of lignocellulosic wood and natural fiber-based materials in 3D and 4D printing, advocating for an environmentally friendly approach to additive manufacturing and contributing to a greener, more sustainable future.

2 Specification of lignocellulosic wood or natural fiber used for sustainable 3D/4D printing technology

Lignocellulosic fibers, mostly sourced from plant and wood resources, consist of cellulose, hemicellulose, and lignin. These fibers are well recognized as biodegradable substances and find use in many industries, including pulp, paper, clothing textile, fiberboards, and nonwoven sectors [24–27]. Fibers could be categorized into two broad items like wood and nonwood fibers; however, nonwood fibers are mostly utilized. Nonwood fibers are also termed as the vegetable or plant fibers as well. Conversely, wood fibers are generally produced from the wood logs or chips. Wood fibers are a category of lignocellulosic fibers that originate from renewable materials, especially obtained from diverse tree species [28]. The variety, richness, and sustainable nature of these entities are well acknowledged. The use of wood fibers has been observed for years throughout a diverse range of applications, owing to their distinctive features. Wood fibers have exceptional mechanical strength, making them well-suited for reinforcing applications in composite materials [28]. These materials demonstrate favorable characteristics such as increased tensile strength, rigidity, and dimensional stability. The aforementioned characteristics render wood fibers very advantageous even for sectors, including building, automobile, and furniture production. Wood fibers have garnered significant interest as a prospective material for AM within the realms of green 3D and 4D printing products [29]. By integrating wood fibers into printed filaments or powders, it becomes feasible to fabricate items that exhibit wood-like appearance, texture, and even possess wood-like qualities.

Rosenthal et al. [30] produced 3D printed wood through utilizing liquid deposition modeling. In the dry mass, they increased the wood content up to 89% which is a very significant content [30], and they found significant physical properties with the certain wood size. In the same study, the observed reduction in size resulting from the process of drying ranged from 17.3 to 20.0%. The density of the material falls within the range of 0.33 to 0.48 g/cm^3. The bending strength (MOR) and modulus of elasticity (MOE) exhibited a diverse range of values, with MOR ranges within 2.3 to 7.4 MPa and MOE ranging from 284.8 to 733.1 MPa [30]. However, the density, MOR, and MOE exhibited an upward trend as the viscosity of dissolved methylcellulose increased, but they showed a downward trend with increasing particle size [30]. The AM technology presents opportunities for the creation of personalized timber prototypes, architectural models, and ornamental items [31]. Nevertheless, there are inherent difficulties associated with the processing and printing of wood fibers, mostly stemming from their proclivity to undergo degradation, combustion, or obstruction during the printing procedure. In Fig. 3a, it demonstrates a schematic process of 3D printed composite material from natural fiber. In Fig. 3b, the molecular-level characteristics of wood are presented to understand its macroscopic properties. However, in Fig. 3c, different 3D-printed bioinspired structures are shown.

Natural fibers are derived from a diverse range of materials found in the natural environment, including plants, animals, and minerals. Throughout millennia, humans have utilized these resources due to their abundance, adaptability, and environmentally sustainable attributes. Natural fibers offer a wide array of advantageous characteristics that make them highly valued in various industrial sectors [35]. Plant-based natural fibers are particularly prominent and find numerous applications. Examples of some commonly used natural fibers include cotton, hemp, flax, jute, sisal, bamboo, and kenaf [35]. These fibers are obtained from stems, woods, leaves, or seeds of various plant species. They exhibit properties such as satisfactory tensile strength (20–1672 MPa)

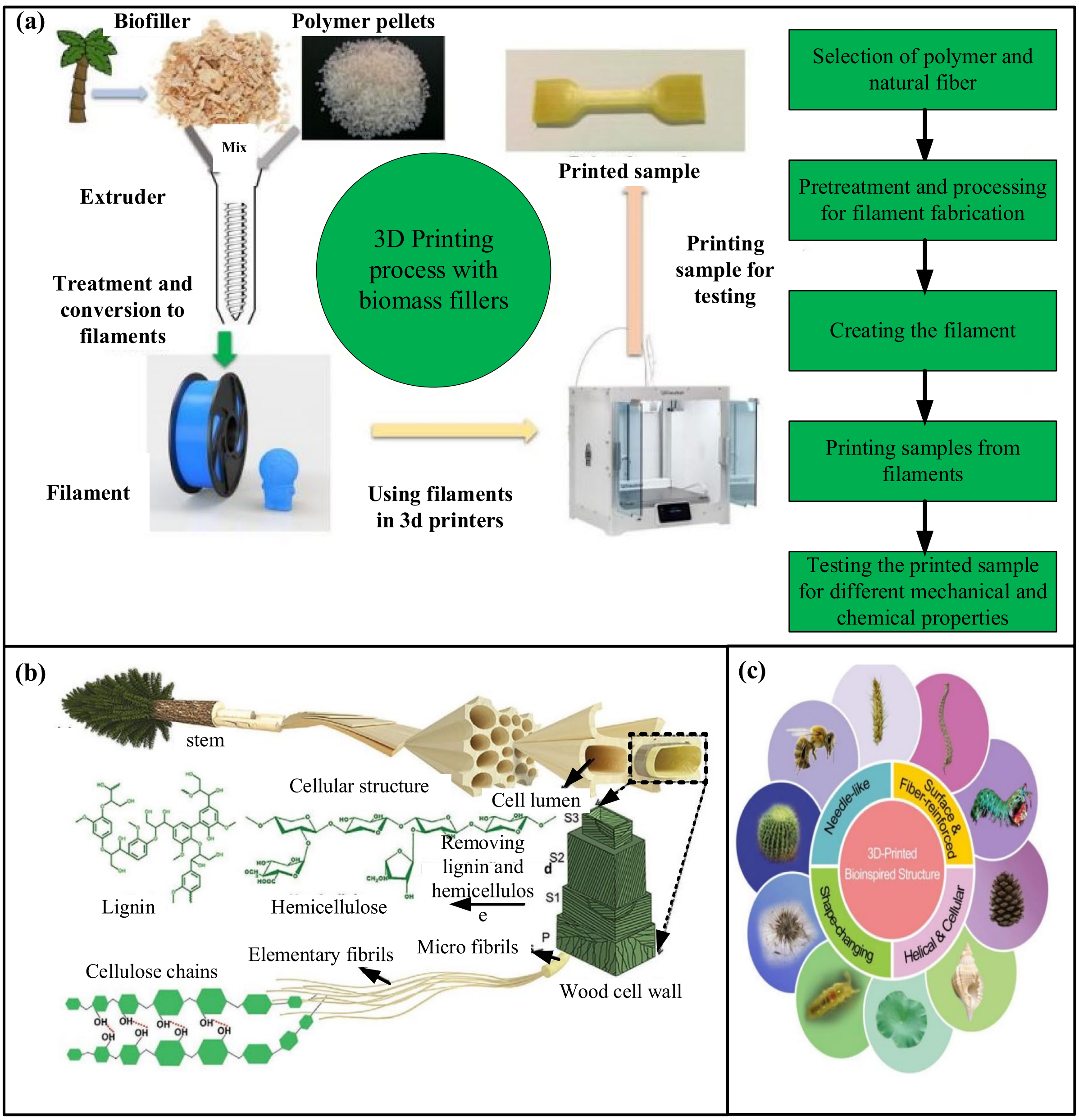

Fig. 3 **a** The process of 3D printing with natural fibers [32], **b** a graphical representation of the structure of hierarchy of wood, from the macroscopic to the molecular level [33], and **c** bioinspired and functional structures of 3D printed products [34]

[36], effective air permeability, efficient moisture absorption, and biodegradability [37, 38]. Plant-derived fibers are extensively employed in the textile industry for manufacturing garments, home textiles, and industrial materials. They are also utilized for producing paper, packaging materials, and composite materials used in the construction and automotive sectors [37, 39]. Nevertheless, it is important to acknowledge that natural fibers have certain limitations. The quality of these materials may exhibit less uniformity, and they can be susceptible to deterioration when subjected to environmental factors for instance moisture, ultraviolet light, or pests. Additionally, natural fibers processing and manufacture need specialized procedures to enhance their properties and address inherent constraints. Recently, interest towards utilization of natural fibers in the field of 3D and 4D printing is increasing [40]. By integrating natural fibers into printed materials, it becomes possible to create products with distinctive visual qualities, enhanced mechanical characteristics, and heightened ecological sustainability [41, 42]. This opens up opportunities for customizing goods, architectural models, and prototypes while incorporating a natural and environmentally conscious element. Duigou et al. reported about the possibility of developing sustainable 4D printed composites which enables the production of biocomposites that undergo shape-changing behavior using continuous flax fibers [43]. This study assesses the feasibility of utilizing 4D printing techniques for the customization of Hygromorph BioComposite (HBC), hence enabling the development of environmentally friendly metamaterials capable of altering shape in a sequential manner [43].

Lignocellulosic fibers derived from wood and natural fibers are both considered as the renewable raw materials. Natural fibers mostly consist of cellulose derived from various sources, with the most prevalent ones being from the plant world. When making a choice between lignocellulosic wood or natural fiber for the purpose of 3D/4D printing, it is crucial to take into account many elements in order to get the most favorable outcomes. The selection of fiber or wood material should be conducted with careful consideration of the particular criteria associated with the intended use. Various types of natural fibers, including hemp, pulb, flax, jute, and bamboo, cotton, exhibit unique mechanical characteristics, sustainability attributes, and biocompatibility. For example, this study used wood pulp fibers incorporated into a PLA matrix and found an improvement in the interfacial adhesion between the fibers and the matrix [44].

In Fig. 4a–d, different sources of natural fibers and fiber/matrix system employed to fabricate sustainable composite materials in recent times are presented. In a similar vein, several wood species, including birch, pine, and oak, display disparities in relation to their strength, density, and moisture content. The decision on the selection should be predicated by the intended attributes and characteristics of the printed product. Additionally, size of the fibers or wood particles play a fundamental part in ensuring the success of the 3D/4D printing procedure. The dimensions and proportions of the fibers have the potential to influence the printability, structural integrity, and surface polish of the resultant printed item. The optimization of fiber or wood particle length, diameter, and homogeneity is crucial in the context of the particular printing technique and intended product. Therefore, a thorough evaluation of the intrinsic qualities of the chosen lignocellulosic wood or natural fiber is required. Apart from this, the matrix can play a significant role since it can lead to anisotropic material properties in the 3D printed composites [45, 46]. In Fig. 4a–c, fiber, fiber-matrix, and bundle interfaces of natural fiber and natural fiber-based composite are presented. The comprehension and selection of fibers or wood materials possessing appropriate characteristics will significantly contribute to the attainment of the intended performance, sustainability, and overall quality of the printed goods. This will be achieved by carefully selecting and specifying the suitable lignocellulosic wood or natural fiber for the printing process, considering factors such as material type, dimensions, and inherent properties. In this regard, a novel methodology for designing and producing the wind blades has been unveiled by researchers [47]. This approach utilizes 4D printing to replicate the structure of the leaf, leading to notable enhancements in strain energy, stress intensity, stiffness, static strength, fatigue life, and reversible bend-twist coupling (BTC), all while eliminating the need for actuators and sensors [47]. Performance evaluations, computational fluid dynamics (CFD) analyses, and wind tunnel tests have demonstrated the suitability of this environmentally sustainable, multipurpose blade for wind turbines [47].

Tao et al. [49] revealed a recent development in the field of 3D printing, whereby WF (Wood fiber) and PLA filled composite materials were used using fused deposition modeling. In contrast to pure PLA filament, the introduction of WF altered the materials fracture surface microstructure, increased the composite's resistance against initial deformation, marginally decreased its initial thermal degradation temperature, and had no discernible impact on its melting temperature, according to a recent study [49]. In another investigation, Fruleux et al. [50] conducted a research on hydromorph biocomposites, wood fibers reinforced with PLA with a wood volume fraction of 40% (content of wood 0% and 10%, size 0.5 mm to 3.0 mm). These materials exhibit the capacity for self-shaping via the introduction of a bioinspired architecture consisting of multiple layers, as well as the swelling generated by moisture in the naturally formed vegetable fibers. This unique combination allows for precise control over the resulting movements. The introduction of functional porosities in a 3D-printed

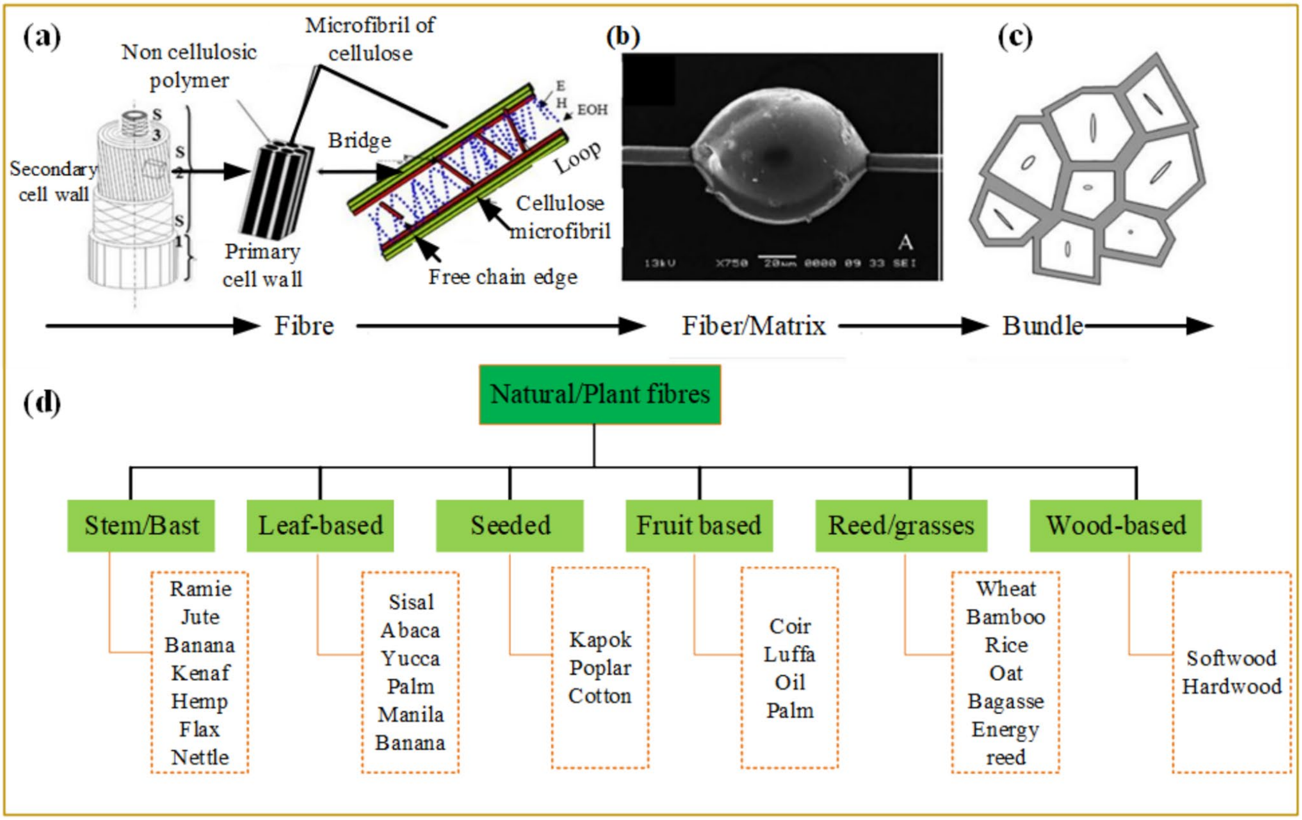

Fig. 4 The multiple scales necessary to analyze the **a** the interfaces of internal fiber cell-wall, **b** fiber/polymer interface, and **c** the interface between fiber to fiber inside the bundles of natural fiber composites system are described [2], and **d** classification of various vegetable fibers [48]

biocomposite, reinforced with wood fibers, enhances the reactivity of moisture-induced actuation while maintaining the integrity of other functional properties [50]. This improvement leads to a substantial eight-fold acceleration in the speed of shape-change transformation and enables the customization of morphing capabilities in biocomposites [50]. The nanoparticles' usage for developing 3D/4D printed materials has acquired considerable popularity recently. In this context, Kumar [51] performed a study that centers on the creation and characterization of a composite material composed of WF reinforced PLA with the incorporation of nano-SiO_2 particles. The findings reveal that there is a positive correlation between temperature and both the recovery ratio and the rate of shape memory recovery [51]. The incorporation of 2 wt% of nano-SiO_2 results in a significant enhancement in micro-hardness, MOR, and impact strength, with increases of 35%, 25.4%, and 32.6%, correspondingly [51]. Incorporating 5 wt% of WF into the material resulted in significant enhancements in micro-hardness, MOR, and overall strength. Specifically, the addition of WF 5 wt% led to a 17% rise in micro-hardness, a 4.4% increase in MOR, and a 15% increase in overall strength [51]. Conversely, the incorporation of WF and nano-SiO_2 results in a reduction in tensile strength [51]. Furthermore, shape recovery rate is positively correlated with the rise in WF loading, whereas it is negatively correlated with the increased loading of nano-SiO_2. The thermal degradation temperature is enhanced by the augmentation of WF and nano-SiO_2 content in thermal analysis [51]. In contrast, the incorporation of WF and nano-SiO_2 has been seen to result in a reduction in the heat conductivity of the PLA composite. The utilization of wood fiber biocomposites in 3D printing presents challenges such as nozzle clogging, filament breakage, uneven melt flow, rougher surface quality, and increased porosity compared to pure polymer components. These limitations are influenced by both the wood fiber percentage and size. Typically, wood fiber fractions ranging within 5–20 wt% are employed, with 20 wt% already causing stick–slip phenomena during extrusion with a nozzle of 1 mm [2, 52]. The study also revealed that the tensile strength and modulus of 3D printed components decrease as the fiber fractions exceed 10 wt%, primarily due to an increase in porosity [2]. In 3D printing, nozzle exits usually range from 0.2 to 0.8 mm, requiring particulates or fibers with a diameter smaller than that of nozzle [53]. Recycled fibers are used in particle forms or the flour, rather than fibers with high aspect ratios, which are used to produce 3D printing filaments to minimize blockages [52]. However, the irregular shape and comparable wood particles size do not enhance the tensile properties of the polymer matrix. The potential application of larger nozzle diameters in 3D printing, especially in industries like construction, building, and automobile manufacturing, offers an exciting opportunity for reinforcing materials with wood fibers that have a higher aspect ratio. This approach can potentially enhance the structural integrity and mechanical properties of larger printed components [52]. When it comes to natural fibers, their mechanical qualities, like stiffness and strength, are not the same everywhere. They depend on their hierarchical substructure and molecular composition, which includes lignin, pectins, cellulose, and hemicelluloses [2].

3 3D/4D printed materials developed through integrating lignocellulosic materials

Integrating lignocellulosic materials into the process of 3D/4D printing has presented novel opportunities for the advancement of sustainable and environmentally conscious manufacturing practices. Lignocellulosic materials, such as wood and agricultural leftovers, possess distinctive characteristics such as fibrous structure, low cost, biodegradability, compatible with wide range of binders, that render them well-suited for AM. The conversion of these materials into printable forms, such as filaments or fibers, enables the production of 3D printed objects that exhibit improved environmental performance [54]. Conversely, the AM technique is also termed as 4D printing enables the structures or objects fabrication from materials whose properties, functionality, or form can gradually transform in response to environmental stimuli comprising heat, moisture, light, or other environmental influences [55]. For example, this research discusses a 4D printed smart valve designed to control the flow of hot and cold liquids in harsh environments, capable of self-assembly [55]. In contrast to conventional 3D printing methods that yield fixed objects, 4D printing incorporates the concept of time, enabling printed materials to autonomously assemble or transform into pre-established configurations or shapes. Integrating intelligent materials like hydrogels or SMP into 3D-printed components makes these structures dynamic. This allows the structures to execute predetermined operations or endure programmed changes subsequent to their creation. The intrinsic attributes of lignocellulosic materials, including their renewable quality, diminished carbon footprint, and capacity for waste usage, are in accordance with the increasing need for sustainable manufacturing methodologies.

3D printing uses plastic, ceramics, and metal to build 3D structures. However, certain materials' low response to external stimuli limits their 4D printing potential. In Fig. 5a, b, the schematic process of 3D and 4D printing and different 4D printed materials with their stimulus responses is provided. However, Fig. 5a, c exhibits the 3D printed wood composite with PLA filament, showcasing its application. In recent times, the use of 3D printers has facilitated the production of a wider range of objects with practical features via the modification of processing factors, including nozzle properties, printing settings, and temperature. Consequently, selecting appropriate materials for 4D printing is crucial. Polymer-based composite materials, in particular, benefit from reinforcement through the incorporation of particles, fibers, or nanoparticles. In a recent research study conducted by Sharma et al. [56], a fusion material comprising of wood and biodegradable polylactic acid (PLA) was reinforced with silica nanomaterial. The study revealed that loading of nanoalumina and silica led to improved stability against thermal degradation and a decrease in the rate of maximum shape recovery in the wood PLA composite. Moreover, the incorporation of 2 wt% nanoalumina showed a 40% rise in hardness, a 25% rise in compressive strength, and a 3.3% elevation in bending strength [56]. Conversely, the inclusion of 2 wt% of nanosilica significantly enhanced hardness, compressive strength and bending strength, demonstrating improvements of 60%, 55%, and 10%, correspondingly [56]. Trey et al. [57] introduced a promising technique for producing 3D lattices composed of structured nanomaterials. This method involves utilizing wood templates to facilitate the controlled development of inorganic material within the organized wood cell walls. The wood templates were impregnated with transition metal ion precursors like iron, cobalt, and manganese under a pressure of 0.5 MPa, which were subsequently converted into magnetic particles such as $MnFe_2O_4$, Fe_3O_4, and $CoFe_2O_4$ using alkaline solutions [57]. The study demonstrated the successful fabrication of cost-effective and ecofriendly lightweight wood composites with ferromagnetic properties through this methodology. Functionalization of the wood framework was achieved by incorporating a significant weight percentage of particles ranging from 15 to 20, as confirmed by thermo gravimetric analysis (TGA) observations [57].

In recent times, notable advancements have yielded a variety of intelligent materials suitable for the purposes of functional 3D or 4D printing. These materials exhibit self-assembly characteristics in response to external stimuli like temperature, ultraviolet (UV) radiation, self-degradation, or water absorption. The latest iteration of 4D printers utilizes the process of water absorption to facilitate the execution of 4D printing tasks [60]. To fabricate bi-material structures, the researchers use two distinct materials characterized by varying porosities and water absorption capacities. Both structures are characterized by a porous surface that is capable of absorbing water, and a stiff layer that is impermeable to water on the opposite side.

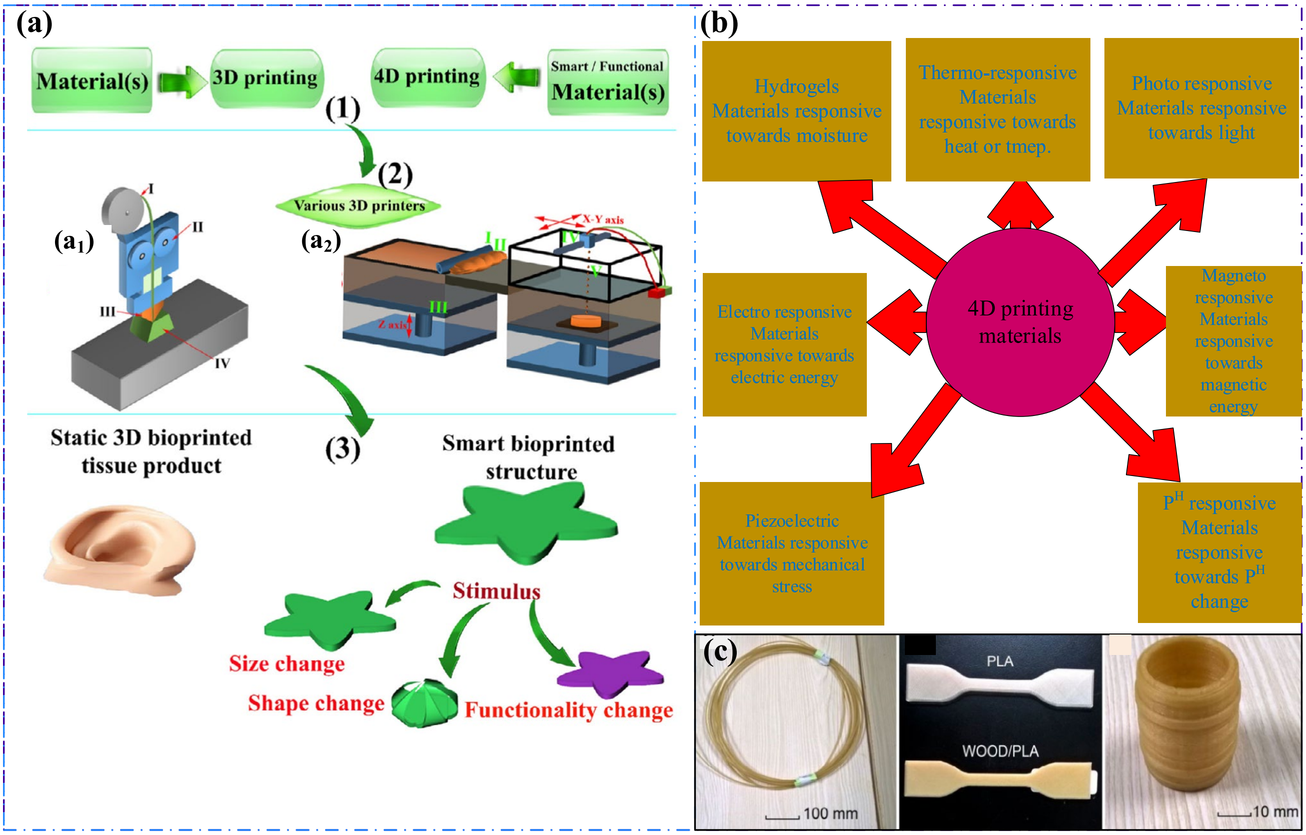

Fig. 5 **a** Generalized 3D and 4D printing schematics, covering (1) materials and (2) technological procedures [58]; **b** materials used in 4D printing are shown schematically [59], **c** different 3D printed products and the test samples [49]

4 Processing techniques and material formulations for sustainable 3D/4D printing technology through integrating lignocellulosic materials

The AM or 3D printing methods can be broadly categorized into seven main groups: material extrusion, material jetting (MJ), vat-based photopolymerization (VP), directed energy deposition (DED), powder bed fusion (PBF), sheet lamination (SL), and binder jetting (BJ) [61]. However, among these methods, material extrusion, PBF, and VP achieved significant attention in the different industries for fabricating wood and natural fiber-based parts and their applications. In Fig. 2c, schematic processes have been presented to illustrate the overall manufacturing process for the FDM printing method. This manufacturing process is also similar to other 3D printing methods. Firstly, a CAD (computer-aided design) model of the printed part needs to be designed in the CAD software. After that, the CAD file is required to save as an STL (Standard Tessellation Language or Standard Triangle Language) file. Later this STL file is inserted into slicing software. In this slicing software, the required manufacturing or printing parameters are mainly predefined, and the slicing software will provide a GCODE. Finally, this GCODE inserts into the machine, feeding the 3D printing materials (Filament), and the printing process starts. Between material insertion and final 3D printing parts, there may have some pre and post-processing's based on material type and characteristics [61, 62].

4.1 Material extrusion

The 3D printing method, also known as FDM or fused filament fabrication (FFF), falls under the category of material extrusion in AM and has gained significant attention in multiple industries and academia. This emerging technique may provide unique mechanical performance, vast applications in rapid prototyping, enable to produce the parts at low manufacturing cost, and require less manufacturing time [63]. Moreover, most of the printed parts of this technology are biodegradable, recyclable, and environmentally friendly [32]. Thus, this technology is remarkably employed in biomedical, automotive, aerospace, electronics, and, most crucially, fabricating composite materials. In the FDM manufacturing process, thermoplastic polymeric materials are mainly used as feed materials. However, it is also significantly employed for fabricating natural fiber-reinforced composite (NFRC) materials incorporating natural fillers [64]. These are some common thermoplastic filaments like PLA, polyurethane (PU), acrylonitrile butadiene styrene (ABS), polyether-ether-ketone, polypropylene (PP), polyethylene terephthalate (PET), and polyamides (PA) used in FDM method [65]. To fabricate the natural fiber-based composite materials through FDM technology, first, a thermoplastic material is combined with a natural filler in a particular proportion and extruded through the twin or single extruder and fabricate into a filament. Finally, this fabricated composite filament is fed to the FDM machine and connected to the nozzle, where the printing temperatures of this composite filament are needed to predefine or set above the glass transition temperature [66]. In Fig. 6a–c, flow process has been presented for 3D printed PLA-wood fiber composite (from filament fabrication to final composite products).

Adding natural fillers with thermoplastic materials may enhance the mechanical performance of the FDM-printed composite parts. For example, this study analyzed the effect of different proportions of wood content combined with PLA on tensile properties [52]. The study revealed a significant increase in tensile strength when incorporating wood content at a level of 10%. However, again adding wood content (50%) may negatively affect or decrease the mechanical properties. The observed increase in tensile strength with the addition of wood content may be attributed to the enhanced stiffness of the wood-PLA composite. However, it is important to note that as the wood content increased, the composite also exhibited increased brittleness. In Fig. 6d, e, different manufacturing defects of wood-PLA composite are shown.

Manufacturing setting in FDM printing technique may greatly impact the performance of printed items created

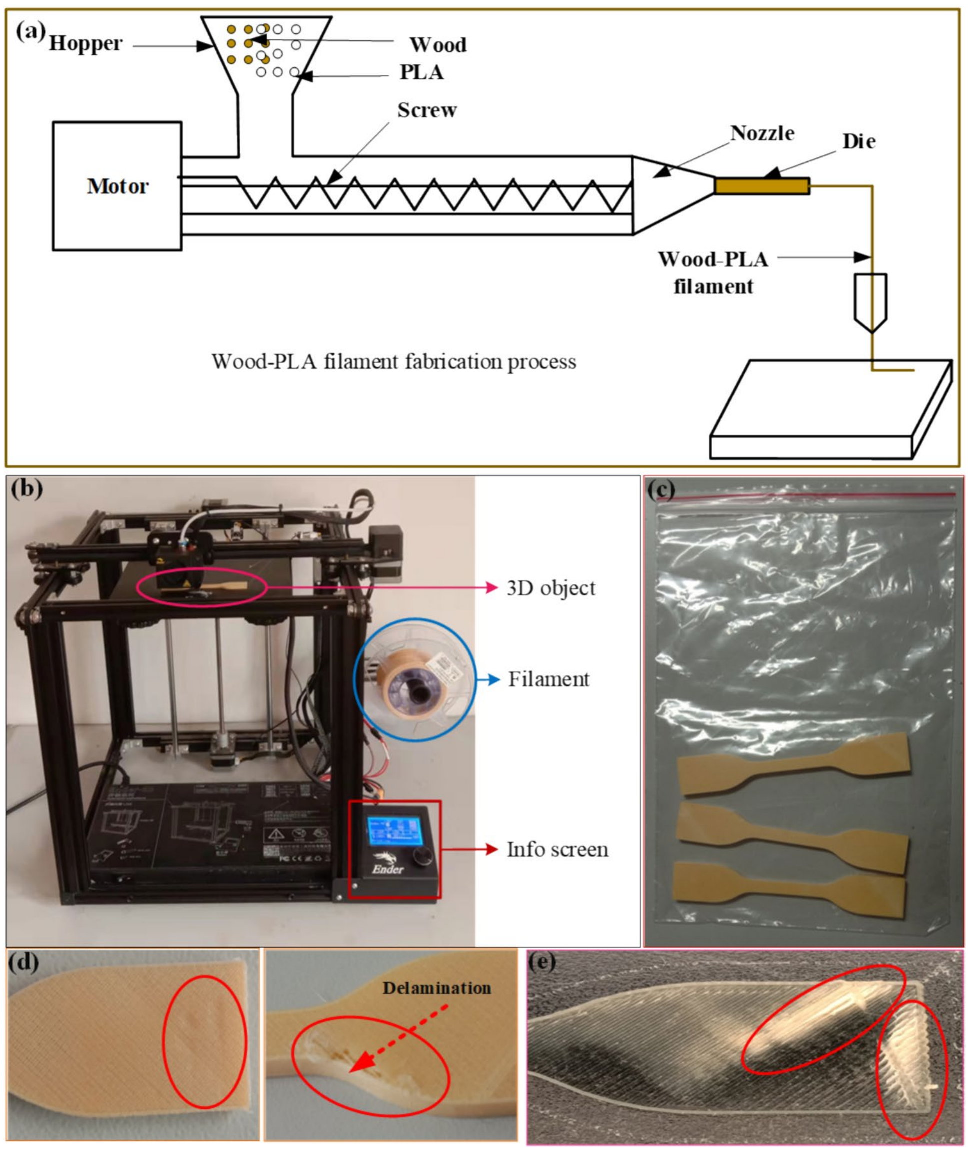

Fig. 6 **a** Fabrication process of a wood-PLA filament by single screw extruder [67]; **b, c** wood-PLA composite manufacturing in FDM via 3D printing machine; **d, e** different manufacturing defects during wood-PLA composite fabrication [68]

from pure thermoplastic or NFRC-based materials [67]. These factors greatly affect print quality and functionality. Layer height, print speed, infill density, and nozzle temperature are crucial to FDM-printed items performance and must be optimized and controlled [67]. For example, one of the recent study by Hua et al. investigated the impact of printing parameters like printing temperature, printing speed, and layer height on the basalt fiber polyamide composite and found that the tensile and compressive properties of this NFRC material were significantly affected by the considered parameters [67]. Similarly, Yang et al. also analyzed the effect of printing speed on wood fiber/PLA composite and obtained that with the increment of printing speed (30 to 70 mm/s), the compressive strength and modulus of the fabricated composite parts drastically reduced by 34.3% and 14.6%, respectively [69]. The possible reason would be as the printing speed increased, the density of the fabricated composite parts reduced and impacted the overall mechanical performance [69]. Moreover, this study highlighted the crucial effectiveness of the raster angle and build orientation on the flexural properties on wood PLA composite [70].

In FDM printing technology, most of the natural fiber composite filaments are fabricated from plant or vegetable-type fibers [49, 52, 61, 63, 65, 66, 69–71]. In plant or vegetable-type fiber, the main constituent polymers are cellulose, hemicellulose, pectin, and lignin. However, natural fibers may also contain some impurities. Although it is challenging to completely remove these from the fiber surface, they can negatively impact the bonding between the fiber and matrix. Therefore, an alkaline pretreatment (NaOH process) is employed to eliminate the impurities and improve the mechanical performance of the natural fiber-reinforced products [72, 73]. Table 1 presents some NFRC (natural fiber reinforced composite) materials used through FDM and DIW printing technologies, along with material composition and prospective applications of the fabricated parts.

Direct ink writing (DIW) is another well-known additive manufacturing (AM) technology, a type of material extrusion where elastomeric ink is extruded through a nozzle in a layer-by-layer process [76]. Similar to FDM or FFF, DIW can also be a suitable fabrication technique for NFRC materials. For example, this study designed and fabricated NFRC parts via DIW, incorporating a silicon matrix with different compositions (15 to 20%) of hemp fibers. The outcomes revealed that, compared to molded samples, these DIW-fabricated parts provided improved mechanical properties However, in DIW, manufacturing parameters such as extrusion speed and material composition rates can play a

Table 1 Material composition, fiber content (%), considering parameters, different study findings, and NFRC material applications

Material composition	Natural fiber content (%)	Considering parameters	Main findings	Applications	Ref
Polyamide (L-PA) + basalt, FDM	15%	Layer height, printing temperature, and printing speed	Mechanical properties improved	Complicated structures	[67]
PLA + wood, FDM	40%	Printing speed	Mechanical properties decreased	Aeronautics, civil engineering, art, and medical fields	[69]
PLA + wood + PHA (polyhydroxy acid), FDM	30% recycled wood	Printing temperature	Printing temperature optimized		[66]
PLA + wood, FDM		Raster angle and build orientation	Flexural properties enhanced	Engineering application	[70]
PLA + wood, FDM	5%	Effect of wood content	Material fracture surface improved	Wooden barrel	[49]
PLA + pineapple leaf, FDM	20%	Printing temperature and printing speed	Tensile properties improved	Applicable as renewable materials	[74]
ABS + oil palm fiber, FDM	5%	Effectiveness of oil palm fiber	Tensile properties slightly improved	Bioproducts	[72]
PLA + kenaf, FDM	2.5%	Effectiveness of pretreatment	Interfacial bonding improved	Aerospace, automotive, and construction industries	[73]
ABS + oil palm, FDM	3%	Layer thickness, orientation, printing speed, infill density	Tensile and flexural properties enhanced	Sustainable application purpose	[71]
PLA + wood, FDM	11% recycled wood	Infill patterns	Flexural properties investigated	Industrial applications	[75]
Silicone + hemp, DIW	15%	Composition of materials	Tensile properties improved	Soft robotics	[76]
Silicone + flax, DIW	0.2–0.5%	Particle ratio, extrusion rate, printing speed	Tensile properties improved	Soft robotics and biomedical devices	[77]

crucial role in the mechanical properties and quality of the printed NFRC materials [76–78]. Additionally, treatments and surface modifications can further enhance the mechanical properties and desired functions of the final components [76, 77]. Some recent studies of DIW-based NFRCs and their potential applications are discussed in Table 1.

4.2 Vat polymerization

Similar to material extrusion 3D printing, VP is also a layer-to-layer process remarkably employed in different industries ranging from biomedical to composite industries [79, 80]. VP was first demonstrated in 1986 by Chuck Hull for polymeric materials (Thermoset). In this method, a vat contains some photopolymer resin which is mainly cured by UV or laser source in a layer-to-layer process to fabricate the 3D parts. However, the main benefits of this technique are that it can significantly improve the mechanical properties of 3D printed parts due to better printing accuracy, has a comparatively low manufacturing cost, and can provide better printing resolutions [81]. The VP method is categorized into different types based on the type of curing source employed. These include SLA, digital light processing (DLP), and two-photon polymerization (2PP) [82]. However, among these, SLA and DLP are achieved significant attention in academia and industrial applications [83].

4.2.1 Stereolithography (SLA)

The SLA method, initially patented by C.W. Hull in 1986, falls under the category of VP 3D printing techniques [84]. As defined by the American Society for Testing and Materials (ASTM), SLA involves the use of one or more lasers to selectively cure liquid-state photopolymers, resulting in the hardening of the material and the fabrication of 3D parts through a layer-by-layer process [85]. Although SLA and FDM both have almost identical manufacturing benefits, however, compared to FDM, SLA is more economical and sustainable [84]. SLA also can provide a better surface finish and higher printing accuracy and enable the fabrication of a wide range of materials [86]. Thus, these emerging methods are employed in several science and technological fields, for example, medicine, bioprinting, tissue engineering, naval, aeronautical, and automotive [87]. In Fig. 7a, a schematic process of SLA is presented. However, Fig. 7b–d illustrates 3D printed wood composite parts (at different percentages) by the SLA 3D printing method and their mechanical performances, like stress–strain curves and tensile strength.

Considering the aforementioned benefits, this technique (SLA) is also used to produce natural fiber-reinforced composites [84, 87, 91, 92]. The incorporation of natural fiber into a polymer matrix in a specific ratio can lead to notable improvements in mechanical properties. For instance, a recent report emphasized that adding 0.4 wt% of softwood craft lignin to a polymer matrix resulted in a substantial enhancement of approximately 60% in the tensile strength of a wood composite printed using the SLA technique [93]. In a similar vein, another study examined the impact of incorporating bamboo fiber particles into composite parts fabricated using the SLA method [92]. The study revealed that the addition of 5% microfiber (average length 192 µm and width 21.1 µm) bamboo particles in the reinforced composites led to a substantial improvement in tensile and flexural properties, increasing them by approximately 3–6 times compared to pure resin [92].

In a SLA 3D printing method, manufacturing parameters can crucially affect the mechanical properties, for instance, particle size (natural or artificial fibers), type of polymer matrix, modification or pretreatment in the natural fiber, perforation ratio, etc. [84, 87, 91, 92]. Indeed, particle size is a crucial parameter in the fabrication of composite materials using the mentioned method. Imperfections in particle size can pose a significant challenge, particularly in achieving optimal properties for the composite materials in question [84]. Additionally, the same study found better tensile strength (50 MPa) when the particle size of the fibrous straw was 45 µm, within the range of 45–250 µm that they studied. However, the outcomes fluctuated and did not show a linear trend with the increase in particle size [84]. Therefore, careful attention must be paid to particle size control during the manufacturing process to ensure the desired performance of the composite materials. A recent study manufactured 3D printed composite parts and conducted analysis on the impact of the size of glass fiber particles (long and short—35.16 µm), and found that long fiber-based composites need processing and the mechanical performance is less than short fiber [94]. Table 2 showcases several applications of SLA-printed natural fiber-reinforced composite parts, including details such as the material composition, natural fiber content (%), and significant manufacturing parameters.

4.2.2 Digital light processing (DLP)

DLP is also a well-known 3D printing technique, able to print high-resolution parts at a higher printing speed than other AM methods. DLP 3D printing method almost has a similar schematic process compared to SLA; however, only the exception is in printing accuracy and resolutions (better than SLA). Thus, this printing method is widely adopted, particularly in biomedical industries, to fabricate scaffolds, tissue regeneration, and bioimplantations [98]. Apart from this, DLP employs a digital projector to flash the image and cure all the resin simultaneously in a layer-by-layer process. This allows it to complete the fabrication process quickly and handle viscous resins effectively. In contrast, SLA utilizes an ultraviolet laser to produce objects point

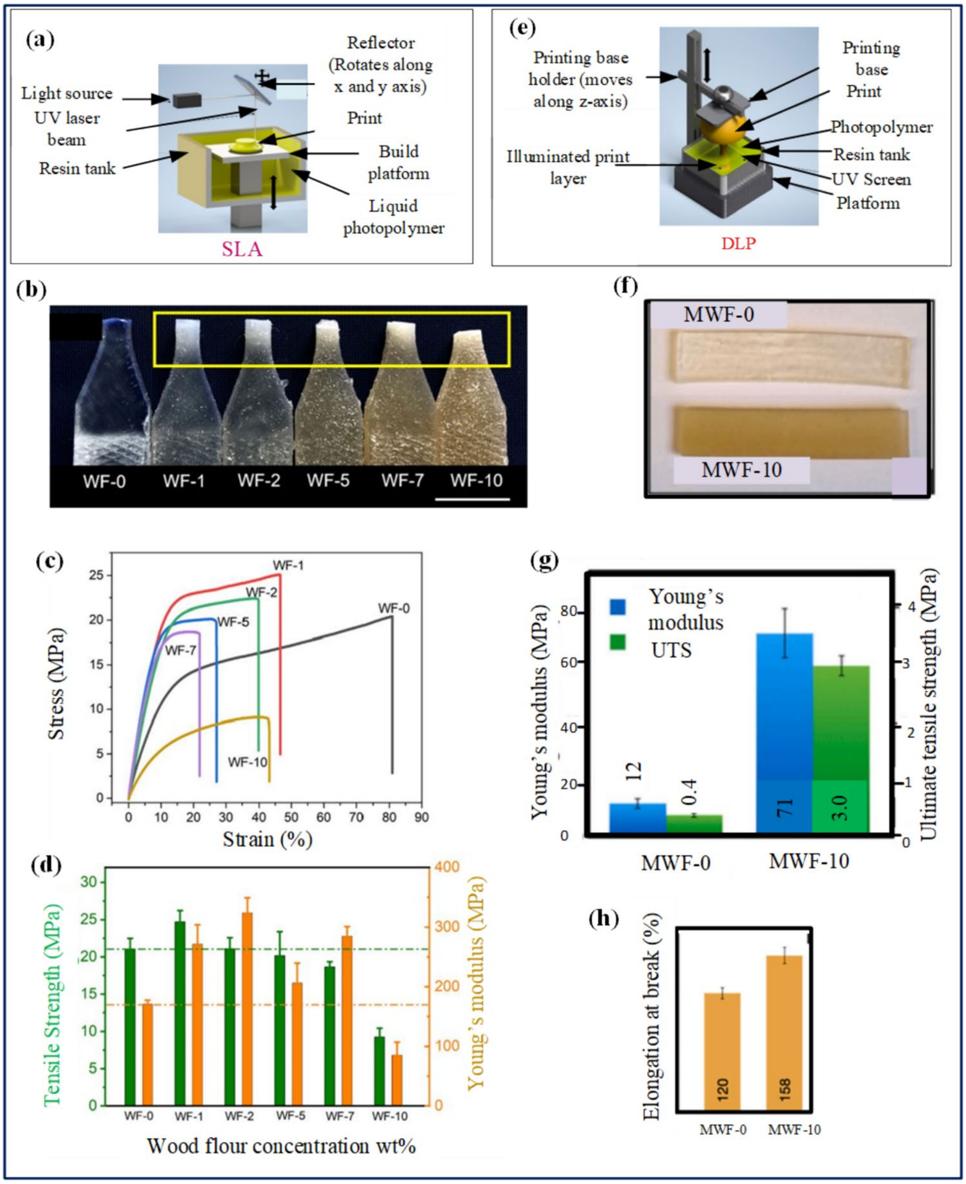

Fig. 7 **a, b** SLA and DLP 3D printing manufacturing process [88]; **c** Poplar WF dog-bone specimens were fabricated using the SLA 3D printing method with varying content; **d, e** mechanical properties of wood composite printed through SLA at different WF concentration [89]; **f, g** DLP fabricated wood composite at 0–10% WF concentration and their mechanical properties [90]

by point, precisely curing the resin in specific areas and accommodating higher viscosity resins. Considering these differences, DLP is faster and more precise since it can produce entire layers simultaneously, enabling it to avoid the challenges associated with higher viscosity resins [99, 100]. A schematic process of the DLP 3D printing method, along with the fabrication of parts using wood lignocellulose and their mechanical performances, is illustrated in Fig. 7e–h. In addition to the mentioned benefits, DLP also offers other advantages. The mechanical properties of DLP-printed parts can be enhanced or customized by a straightforward process, such as altering the resin composition [101]. Moreover, the DLP printing method typically involves less complexity in terms of layering and areas, making it a more streamlined and efficient process [97]. Therefore, this simple 3DP method is vastly used in multiple applications to fabricate and enhance the mechanical properties without compromising any benefits or quality of the 3D printed parts [101]. Tables 1 and 2 show that for composite fabrication via FDM and SLA, most of the natural fiber is used from vegetable-type fibers. In contrast, for DLP, most of the fiber is from animal-type fibers, especially silk fiber. Silk fiber-based composite incorporated with polymer materials manufactured through DLP caught significant attention in bioimplantations, wound healings, enzyme immobilization matrix, and tissue engineering owing to their inherent and favorable biomechanical properties such as biocompatibility and better cell adhesion [96, 97]. Despite these positive effects of silk composite, only the drawback is it may require some pre-processing, such as coupling reactions and amino acid modifications, which is a challenging and time-consuming task [97].

Table 2 Applications, material composition, significant manufacturing parameters of natural fiber reinforced composite materials via SLA and DLP 3DP method

Material composition, 3DP method	Natural fiber content (%)	Considered parameters	Main findings	Applications	Ref
Wood + resin, SLA	10%	Wood content	Tensile properties improved	Engineering applications	[89]
Cork + resin, SLA	5%	Influence of particle size	Mechanical and thermal properties improved	Sustainability purpose	[87]
Recycled cotton fibers + VisiJet-SL, SLA		Impact of perforation ratio and air gap	Sound absorption capability improved	Acoustic absorber	[91]
White, rice straw + resin, SLA	5%	Effect of different fiber sizes	Thermal conductivity decreased	Building materials (boards)	[84]
Bamboo + vinyl monomer, VP	1, 3, 5%	Impact of modified and unmodified micro bamboo fiber in the mechanical properties	Interfacial bonding in fiber and matrix improved	Building and electrical industries	[92]
Silk fibroin + polyethylene glycol acrylate, DLP	0, 0.25, 0.5, 1, and 2%	Impact of particle size	Cell proliferation and cell spreading improved	Tissue engineering and biomedical applications, scaffold	[95]
Silk + gelatin and methacrylic anhydride, DLP	1, 2.5, 5, and 10%	Silk and methacrylic anhydride percentage	Cell adhesion, biocompatibility improved	Biomedical and tissue engineering	[96]
Silk + glycidyl methacrylate, DLP	10, 20, 30%	Degree of methacrylate on silk fibroin modified by various glycidyl methacrylate percentage	Cell proliferation improved	Cartilage tissue engineering	[97]

4.3 Powder bed fusion (PBF)

PBF is a layer-by-layer AM technique that is widely employed in various industries, such as automotive, aerospace, biomedical, and energy harvesting. This technique is highly valued for its ability to produce intricate geometries and its potential for numerous impactful applications [102]. In addition, the fabricated parts can provide better mechanical properties like strength and long life cycles with low manufacturing costs and time [103]. In the powder bed fusion method, feedstock materials are metal, ceramic, alloy, polymer, and composite powder. For fabricating the AM 3D parts, first, the powders are fed into a build platform, and later the laser source mainly are used to melt or fuse the material and make the desired shape of the parts [104]. In PBF, the classification of this AM process can be based on the fabrication method and the type of laser source used. This categorization includes direct metal laser sintering (DMLS), selective laser sintering (SLS), selective laser melting (SLM), electron beam melting (EBM), and multi-jet fusion (MJF) [102]. However, among these methods, SLS is well-accepted for fabricating polymeric composite or natural fiber-reinforced polymer composite [105].

4.3.1 Selective laser sintering (SLS)

Selective laser sintering is a layer-to-layer technique of AM, where the pre-heated powders are spread into the build platform in a layer, and a laser is used to fuse or melt (partially or entirely) the powder material, creating the desired three-dimensional object [106]. SLS can manufacture intricate structures and low-volume parts without any complexity compared to other AM techniques and traditional manufacturing methods. In addition, during fabrication, the printed parts do not require any auxiliary support, and the uninterred material can be reused and recyclable, which is the more sustainable and eco-friendly approach to industrial scale [107]. Therefore, this method has drawn significant consideration in the automobile, aerospace, biomedical, and construction industries and become a hotspot in academia [108, 109]. This method's schematic process and applications are depicted in Fig. 8a–i with its potential applications. However, in terms of polymer composite or natural fiber-reinforced polymer composite this AM method exhibits notable advantages in delivering components with high dimension accuracy and stability at a low manufacturing cost [105]. This technology is also convenient for fabricating various materials like polymers, metals, and composite. Including these suitabilities, the main benefit of this method is that it can enhance longevity, robust corrosion resistance, and better mechanical properties of the natural fiber-reinforced parts than conventionally manufactured biocomposites [108]. In the SLS manufacturing method, manufacturing parameters and the characteristics of natural fiber have a crucial role, which can increase and decrease mechanical properties.

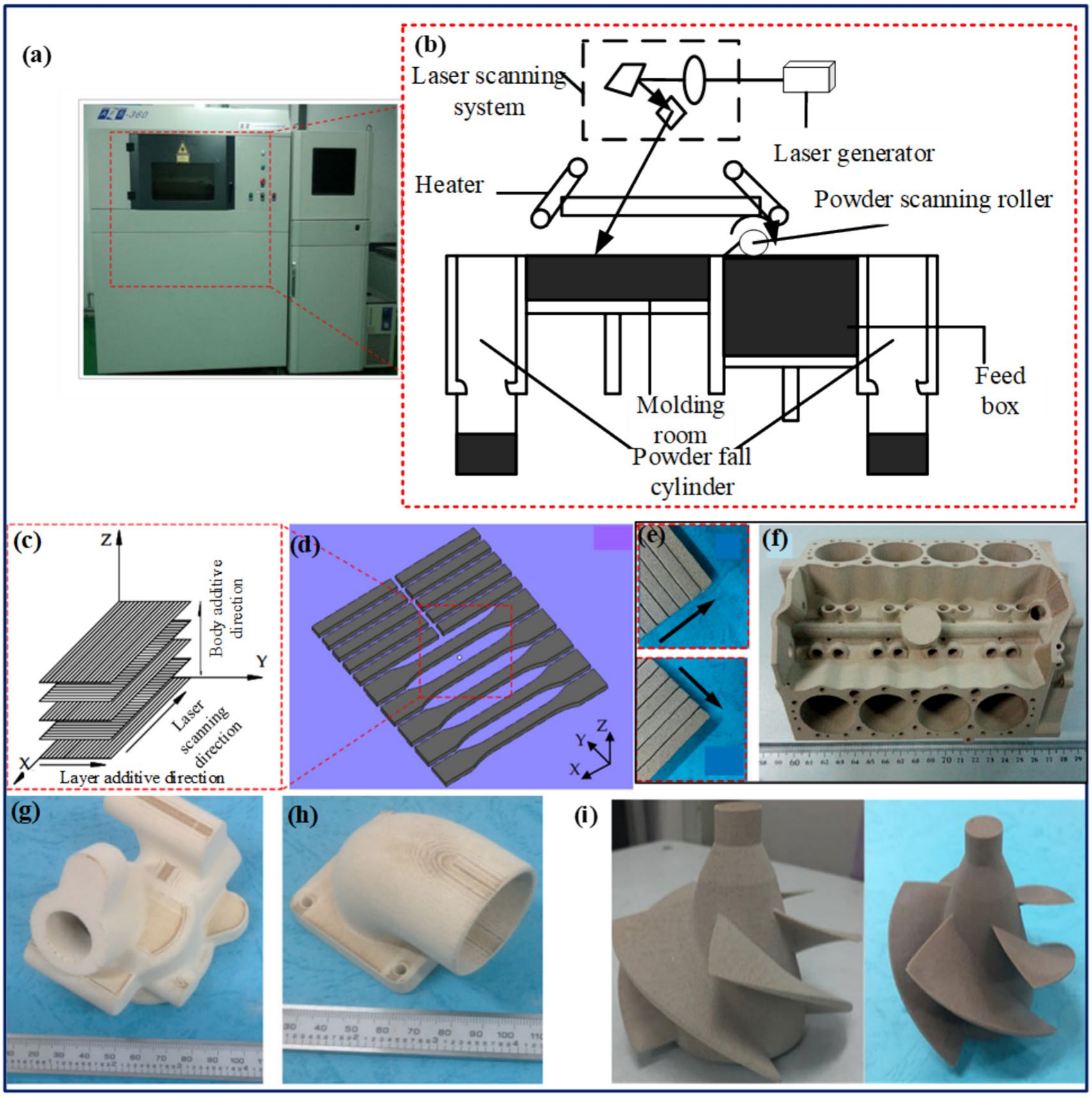

Fig. 8 **a–d** Schematic process and build orientation of SLS method [110]; **e, f** walnut shell composites parts for automotive industry fabricated by SLS [111]; **g, h** green parts manufactured by SLS from bamboo-copolyester composite [112]; and **i** complex shape fabricated from sisal-polyether (sulfone) composite through SLS [113]

For example, an earlier study fabricated a walnut shell composite through SLS. The study examined the impact of various manufacturing parameters on the mechanical properties of the printed parts, including preheating temperature, scanning speed, layer thickness, laser power, and scan spacing. The results revealed that these parameters significantly influenced the mechanical properties of the printed objects. The optimized parameters for achieving superior properties were determined to be a laser power of 12 W, scanning speed of 2000 mm/s, layer thickness of 0.15 mm, scan spacing of 0.2 mm, and preheating temperature of 80 °C [107].

In PBF, particle size also has an influential impact on the mechanical properties, similar to VP as discussed above. For example, this study fabricated NFRCs via SLS from polyether sulfone and sisal fiber, comparing the mechanical properties, such as tensile and bending strength, with different particle sizes of sisal fiber (80 to 160 μm). The highest tensile strength of 3.24 MPa and bending strength of 7.12 MPa were obtained when the particle size was 120 μm [113].

Furthermore, the mechanical performance of natural fiber composites can be significantly influenced by factors such as fiber ratio, particle size, treatment, and the addition of nanoparticles. These parameters are described in Table 3, which also highlights some promising applications of natural fiber-based composites fabricated using the SLS method.

Table 3 Material composition, percentage of natural fiber, proposed applications, and considered factors for SLS fabricated natural fiber-reinforced composite

Material composition, 3DP method	Natural fiber content (%)	Considered parameters	Main findings	Applications	Ref
Polyether sulfone + wood powder (pine) + carbon nanotube	14.3%	Effects of microwave treatment	Bending strength improved	Complex structure	[105]
Polyether sulfone + wood powder (pine) + aluminum powder (Al)	14.3%	Effect of Al powder	Bending strength improved by 128%	Manufacturing industries	[114]
Polyether sulfone + wood powder (Eucalyptus)	20%	Size and distribution of wood fiber	Performance of the parts enhanced	Industrial applications	[115]
Co-Polyester + walnut shell	40%	Power of laser, scanning speed, scan spacing and the temperature for preheating	Manufacturing parameters optimized	Automotive, aerospace, construction industries	[107]
Polyether sulfone + sisal	10,15,20,25%	Sisal fiber ratio and particle size	Mechanical properties enhanced	Complex shape (fan blade)	[113]
Polyether sulfone + sisal	20%	Temperature used for preheating, power of laser and its speed	Mechanical strength significantly increased	Complex geometries	[116]
Polyether sulfone + sisal	20%	Preheating temperature, scanning speed, and laser power	Quality of the parts improved	Aerospace, automobile, casting parts, building and construction materials	[117]
Copolyamide + copolyester + walnut shell	40%	Preheating temperature, amount of powder	Mechanical properties increased	Automobile, aerospace, medical materials	[109]
Copolyester + Wal nutshell	40%	Particle size	Surface roughness, mechanical properties provide in a satisfactory range	Rapid prototyping, industrial parts	[118]
Polyether sulfone + wood + carbon nanotube	14%	Carbon nanotube content	Thermal conductivity improved	Complex parts	[119]
Copolyester + wood	90% (powders of wood and powders of hot-melt adhesive)	Laser intensity	Tensile and bending strength significantly improved	Rapid prototyping	[120]
Copolyester + bamboo	20, 25, 30%	Mixing ratio	Mechanical properties increased	Sustainability purpose	[112]

5 Applications of 3D/4D printed composite products through integrating lignocellulosic materials

Nowadays, industries are increasingly focused on manufacturing 3D and 4D printed composite parts or products using materials based on lignocellulosic wood and natural fibers. This trend is likely for their inherent characteristics, like biodegradability, low carbon footprint, ease of collection and handling, low manufacturing cost, and their potential to combat climate change challenges [121, 122]. However, the adoption of advanced manufacturing methods such as 3D and 4D printing, combined with the use of polymers like PLA, ABS, and PEEK (polyetheretherketone), along with natural fibers, fillers, or powders, can enhance not only mechanical strength but also provide substantial mechanical performance. This includes benefits such as durability and prolonged service life, making these materials superior to those produced through traditional or conventional manufacturing methods [16, 123, 124]. Apart from this, sustainability, recyclability, eco-friendliness, and improved thermal and physical properties are some other core benefits of 3D/4D printed parts [68, 123]. Furthermore, it has unique features like enabling the design of complex geometries, and fabricated components mostly provide better surfaces with decent mechanical properties [125].

5.1 Wood-polymer composite 3D/4D products

AM is a process renowned for fabricating components in 3D or 4D. The key distinction between 4 and 3D printing lies in dimensionality. While 3D printing creates items in three dimensions, 4D printing adds an additional dimension, allowing components to change over time in response to various stimuli such as temperature, light, pressure, water, and other environmental conditions. This capability of 4D printing enables the creation of dynamic and adaptive structures with enhanced functionality [126]. Incorporating wood lignocellulose into polymeric materials can significantly enhance the mechanical properties of 3D/4D printed components. However, bioderived polymers are mainly employed in AM, especially in FDM technology.

The most probable reasons for adopting bioderived polymers in combination with wood are their availability, comparatively low manufacturing temperatures requirements, and superior physical and economic sustainability compared to other petroleum-based materials [122]. In Fig. 9, some wood polymeric products are highlighted. Different biodegradable polymers are added with wood fillers to fabricate these products. For example, in Fig. 9a, 5 wt% wood is added with PLA to fabricate this barrel through FDM 3D printing technology [49]. Similar to this, some

Fig. 9 **a** FDM 3D printed wood-PLA barrel [49]; **b** 3D printed wood product [133]; **c** FDM 3D printed wood-PLA-gypsum composite [127]; **d** extrusion based wood composite [134]; **e** a truncated cone of wood 3D printed, **f** liquid deposition modeled wood composite 3D printed tower [127]; **g** direct cryo writing 3D printed wood composite nut and screw [128]; **h** 3D printed signal material from wood [135]; **i** FDM printed gyroid sandwich structure of wood-PLA composite [129]; **j** 3D printed bamboo-PLA composite for furniture products [130]; **k, l** FDM 3D printed mobile shell application of wood-polyurethane composite [131]; **m** large scale 3D printed poplar-PLA composite for building products [132]; **n** 4D printed flax-PLA composite [136]; **o** 4D printed shape-changing wood-PLA composite [121]; **p** 4D printed flax-poly butylene succinate shape changing composite [43]; **q** 4D printed wood composite shape changing phenomenon [137]

wood polymer products are shown in Fig. 9b–d, h, which are printed via FDM. However, a different 3D printing technique, namely liquid deposition modeling, is adopted to fabricate this tower, depicted in Fig. 9f. The 3D part is produced by utilizing beech wood powders with particle sizes of 250 µm and 400 µm, along with methylcellulose, which serves as both a binding agent and a lubricant [127]. In Fig. 9g, the nut and screw are illustrated, which is printed through the direct cryo writing 3D printing method from wood [128]. However, in Fig. 9i, green composite products with the gyroid core, which are fabricated through FDM 3D printing technology and PLA/wood composite, are presented [129]. This study also highlighted that wood-based polymer composite which can significantly reduce the printed parts weight and help produce a lightweight structure. Figure 9j shows some furniture manufactured by FDM 3D printing technology from wood-PLA [130]. However, the study discussed that wood-PLA furniture can provide excellent scratching and wearing resistance and could be a sustainable alternative to plastic-based household objects. In Fig. 9k, l, the mobile phone shell is printed via 3D FDM technology from polyurethane/wood composites [131]. Moreover, in Fig. 9m, a building material is presented, which is manufactured in a large-scale 3D printing machine from poplar/PLA composite. This study concluded that adopting fibrillated fibers based polymeric composite derived particularly from biomass can be a suitable alternative to traditional carbon fiber composite [132].

In Fig. 9n–q, some 4D printed products are shown, which are particularly fabricated from natural fibers. For instance, Fig. 9n illustrates a 4D printed flax fiber biocomposite reinforced with PLA, which can change shape in response to varying humidity levels [136]. Similarly, another study also demonstrated a biomaterial composite combination of wood-PLA, enabling it to change its original shape under different climate conditions. However, for this shape-changing 4D printed structure, mainly shape-changing depends on the proportion of the wood content in the composite and the proportion of the materials to the two-layer actuator [121]. In Fig. 9p, q, the various shape-changing of 4D printed biocomposites is shown [43, 132].

5.2 3D/4D printed nanocomposites

Combining nanomaterials with particular materials or composites can substantially improve and tailor the mechanical properties of any final product. Also, different nanomaterials can facilitate the desired function of the materials or composites. Thus, many industries are adding nanomaterials to enhance the composite performance. For instance, this study fabricated an ABS nanocomposite with 5% modified montmorillonite, resulting in a 43% improvement compared to those made with single-constituent ABS in FDM 3D-printed parts [138]. Similar to this, another study also fabricated PLA/graphene nanocomposite and investigated the electrical, thermal, and electromagnetic characteristics of the nanocomposite. The results reflected that this nanocomposite could be a suitable option in tissue engineering, bioelectronics, where sensing applications are particularly focused [139]. However, the addition of cellulose with nanocomposite materials also can improve the biological performance of the nanocomposite. This study highlighted the satisfactory non-toxic performance of cellulose-based polyurethane-graphene nanocomposites printed via the FDM 3D printing method [140].

In the FDM 3D printing method, there is limited discussion regarding the direct incorporation of nanomaterials with natural fiber or woody lignocellulose. However, in this study, PLA, PHB (poly(3-hydroxybutyrate)), and 1% plum seed shell composites were designed and found to exhibit superior thermal and physical performance compared to samples made using compression molding in FDM 3D printed parts. This highlights the potential of incorporating nanomaterials in combination with natural fibers for enhanced properties in FDM-printed components. [141]. Indeed, the utilization of FDM 4D printing for fabricating nanocomposite or lignocellulose nanocomposite materials is a relatively limited area of study discussed in recent research [142]. Recently, a 4D printed wood nanocomposite was fabricated using FDM technology. The study found that a wood-PLA composite reinforced with nanosilica exhibited better thermal, mechanical, and shape memory performance compared to a wood-PLA composite reinforced with nanoalumina [59]. These findings highlight the potential of incorporating nanomaterials in wood-based composites using FDM 4D printing for improved material properties and functionality [59]. Another study also obtained similar performance in carbon nanotube-based nanocomposite [143]. Figure 10a, b illustrates a schematic process of the cellulose-based nanocomposite fabrication process. Moreover, in Fig. 10c–e, mechanical properties and some of the applications of this nanocomposite are provided.

Several studies have fabricated 3D/4D printed nanocomposites to enhance the desired mechanical properties through the VP method [143, 145–151]. This study fabricated nanocellulose-based composite via VP and obtained that small fraction of nanocellulose can remarkably enhance the flexural and tensile properties. The study also highlighted some potential challenges, such as undesired voids, and found the perfect combination or percentage with other materials for fabricating nanocomposite through the VP method [147]. However, 3D/4D printed cellulose-based nanocomposites made of VP method have recently been in high demand, particularly in biomedical applications such as biosensing, due to their superior electrical and antibacterial performance and mechanical strength [152]. Table 1 lists various

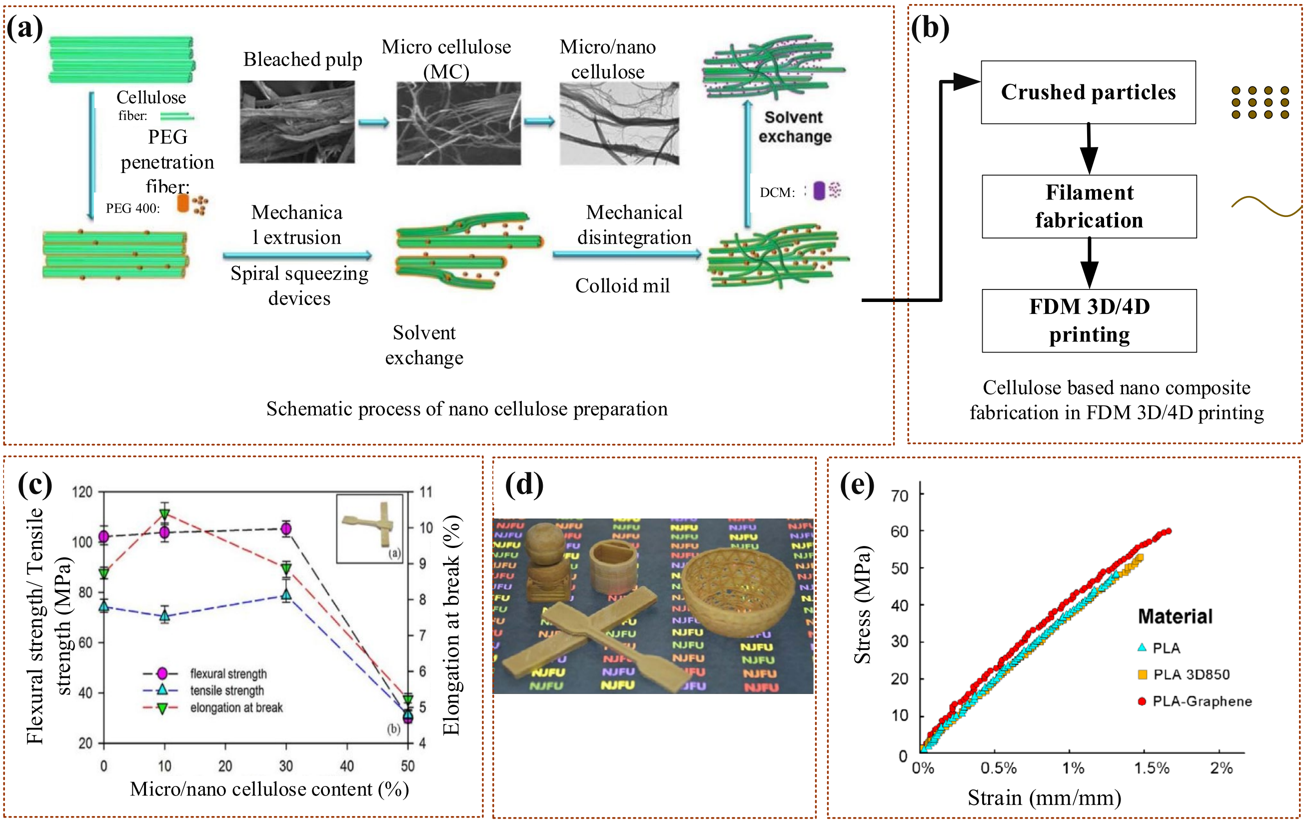

Fig. 10 **a, b** Schematic process of nanocellulose-based composite preparation, filament fabrication, and 3D printing; **c** mechanical characteristics of 3D printed softwood pulp cellulose-based composite; **d** different objects of FDM printed nanocellulose-based composite [144]; **e** stress–strain curve of PLA/graphene nanocomposite [142]

applications of 3D/4D parts printed using FDM, VP, and powder bed fusion techniques, which incorporate different fibers, nanomaterials, and nanocellulose. Figure 11a presents a schematic process of nanocomposite fabrication for any nanoparticles through vat polymerization. In addition, in Fig. 11b–e, some mechanical properties with SEM images (scanning electron micrograph) of cellulose-based nanocomposite at different nanocellulose concentrations with some complex shapes manufactured through VP are presented.

In PBF, incorporating nanomaterials with cellulose fiber, limited studies are conducted. The most probable reason would be that PBF is mostly used in metallic materials, where high strength and better load-bearing materials are also required, for example, in bone tissue applications [155]. However, adding nanomaterials with metallic and thermoplastic materials enormous studies can be found in recent research works [156, 157]. A recent study demonstrates the fabrication of a nanometallic composite consisting of TiN and $AlSi_{10}Mg$ using SLM technology. The study findings indicate that this nanocomposite exhibits excellent mechanical properties, including high tensile strength measured at 492 ± 5 MPa, significant ductility of 7.5% ± 0.29, and microhardness of 157 ± 4.9 HV [156]. Similar to this, another study fabricated nanocomposite of polyamide and multi wall carbon nanotube and graphene nanoparticle via SLS and obtained better thermal, mechanical, electrical, and morphological properties [157]. However, in any nanocomposite, the nanoparticle percentage plays a crucial role in the desired properties [156, 157]. Table 4 highlights some AM manufacturing processes, such as FDM, VP, PBF, DED, and BJ based 3D/4D printed nanocomposites, with their applications found in the literature.

6 Mechanical, physical, and thermal properties of 3D/4D printed products developed through integrating lignocellulosic materials

A thorough comprehension of the performance attributes of these composites (3D/4D) is crucial for their successful integration into various applications. Moreover, the mechanical and physical characteristics of 3D/4D printed products are also extremely significant. Factors such as strength, stiffness, toughness, thermal properties, and dimensional stability are among the key considerations

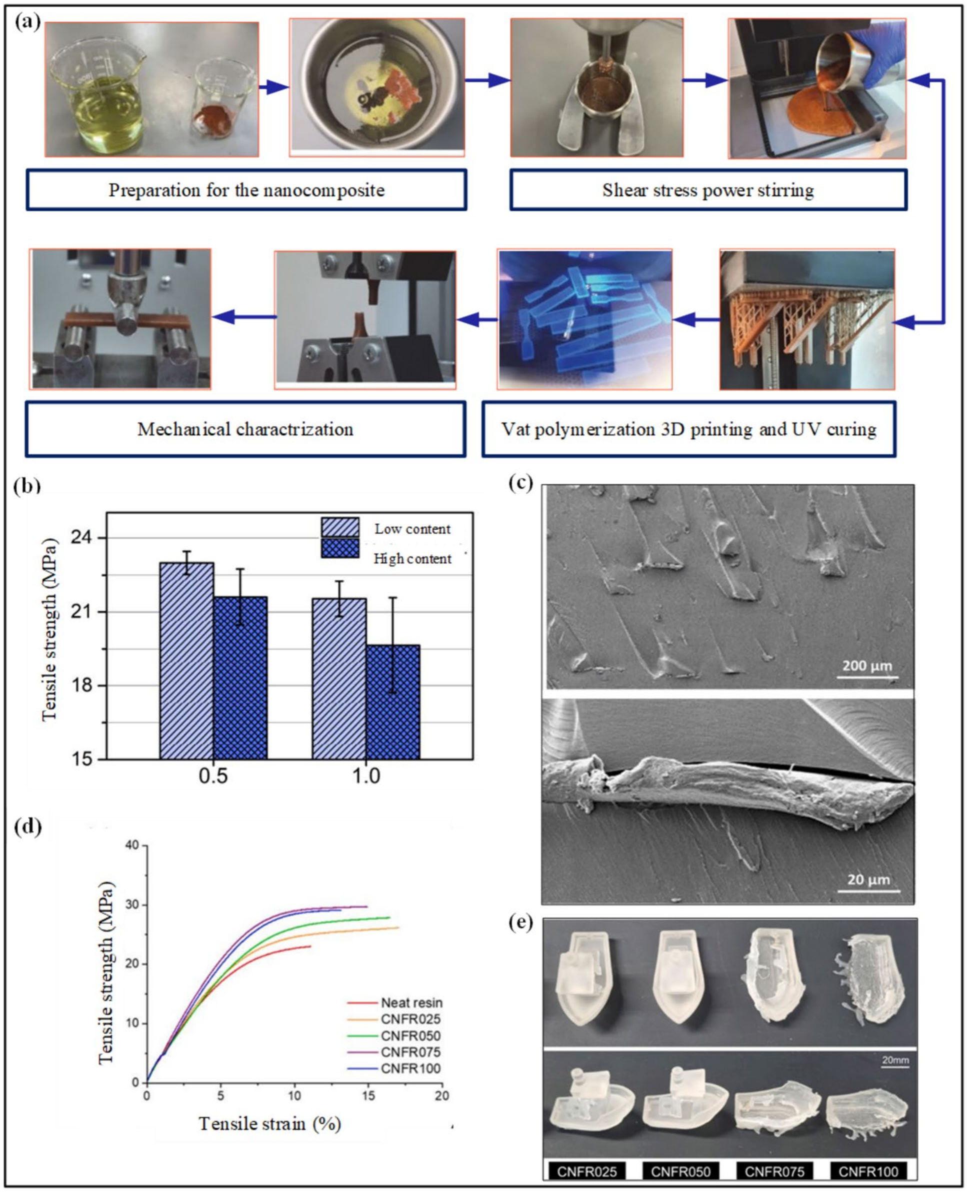

Fig. 11 **a** Schematic process of 3D printed vat polymerization nanocomposite fabrication [153]; **b** tensile strength of cellulose-based nanocomposite at different concentrations, **c** SEM images of cellulose nanocomposite at different magnifications [154]; **d** stress–strain curve of nanocellulose composite fabricated in VP technique; **e** different complex shaped fabricated at different nanocellulose concentration in vat polymerization 3D printing technique [147]

during the design and manufacturing process of 3D/4D printed composites.

6.1 Mechanical characteristics

The mechanical characteristics of composites produced by 3D/4D printing using lignocellulosic materials are subject to several aspects, such as the composition and quantity of lignocellulosic fibers, the alignment of fibers, and the settings used during the printing operation. The mechanical characteristics of these composites may be assessed using important parameters like tensile, bending, and compressive strength. The use of lignocellulosic fibers, such as those derived from wood, shown improved mechanical properties of composites via the process of matrix reinforcement. In Fig. 12a–f, tensile properties, water absorption (%), layer height, weight %, and thermal conductivity of wood lignocellulose composite are highlighted, respectively. The arrangement and dispersion of fibers inside the printed structure have a substantial impact on overall mechanical characteristics, namely, the strength and stiffness. Interfacial adhesion of the matrix and fibers material affects composite load transmission and failure behavior. Zarna et al. [167] did a study to inspect the tensile properties of biocomposites made from granulated or milled thermomechanical pulp fibers and the PLA. The addition of PHA and PBAT in the biocomposite formulations was examined to review their impact on the melt flow index and tensile characteristics [167]. The findings revealed that biocomposites with 20 wt% PBAT demonstrated superior strength and toughness compared to those

Table 4 Different AM technology to fabricate 3D/4D printed nanocomposite, their composition, considering factors, tensile strength, and potential applications

AM technology	Materials composition	Natural fiber content (%)	Considering factors	Tensile strength (MPa)	Findings/improvements	Applications	Ref
3 DP (FDM)	ABS, 5% montmorillonite	N/A	Comparison in FDM and injection molding	64.43	FDM printed nanocomposite provided better mechanical and thermal properties	N/A	[138]
3 DP (FDM)	PLA, 10% graphene	N/A	Dimensional accuracy and mechanical properties	40.2	Mechanical, electrical and electromagnetic properties were significantly improved	Bioelectronics	[139]
3 DP (FDM)	ABS, 7% multiwall carbon nanotube	N/A	Effect of raster angle	58	Tensile and electrical conductivity dramatically increased	Electrical conductor	[140]
3 DP (FDM)	TPU, PLA, 5% graphene oxide	N/A	Percentage of graphene oxide	60	Biocompatibility, mechanical and thermal properties significantly improved	Tissue engineering	[158]
3 DP (FDM)	PLA, PHB, nanocellulose	1	Comparison in fabrication methods	89	Interfacial adhesion, thermal stability enhanced	Engineering applications	[141]
3 DP (FDM)	The poly(3-hydroxybutyrate-co-3-hydroxyhexanoate) and nanocellulose	5–20	Effect of reinforcing agent	–	Thermomechanical properties enhanced	Scaffold	[159]
3 DP (FDM)	PLA, PVC (polyvinyl chloride), wood, Fe_3O_4 (20%)	5	Impact of processing parameters	24.21	Tensile properties improved	Civil engineering applications	[160]
4 DP (FDM)	Bisphenol epoxy, benzoxazine, 4% carbon nanotube	N/A	Effect of different printing directions	60	Mechanical strength, thermal stability, and shape memory performances improved	Automotive, biomedical, electronics and aerospace	[143]
4 DP (FDM)	Wood, PLA, silica, alumina	96–100	Effect of nanoparticles	–	The mechanical performance of wood nanocomposite improved compared to wood PLA composite	Automotive, biomedical, electronics and aerospace	[56]
3 DP (VP)	Nanocellulose, resin	0.5–1	Effect of nanocellulose content	30	Mechanical properties improved	Complex shape	[147]
3 DP (VP)	Nanocellulose, resin	0.1–0.5	Biobased acrylic weathering performance	–	Mechanical performance improved	Outdoor applications	[149]
3 DP (VP)	Nanocellulose, resin	0.25–1	Effect of nanocellulose percentage	35	Mechanical strength improved	Multiapplication	[154]
3 DP (VP)	Nanocellulose, copper, resin	1	Synergetic effect of filler loading	–	Mechanical properties and antibacterial performance significantly improved	Biomedical applications	[150]
3 DP (VP)	Nanocopper, resin (0.5–1%)	N/A	Filler percentage and printing angle effect	81.1	Antibacterial performance improved	Biomedical applications	[153]
4 DP (VP)	Liquid crystal, resin	N/A	Different shape and their effect investigated	–	Satisfactory tensile, electrical properties	Meta-material sensor	[148]
3 DP (PBF)	Polyamide, 0.5% carbon nanotube, 3.0% graphene	N/A	Reinforcement percentage	20–40	Mechanical properties improved	Advanced applications	[157]

Table 4 (continued)

AM technology	Materials composition	Natural fiber content (%)	Considering factors	Tensile strength (MPa)	Findings/improvements	Applications	Ref
3 DP (PBF)	TiN/AlSi10Mg, 0–6% TiN	N/A	Effects of TiN percentage	492 ± 5	Tensile strength, ductility and hardness improved	Multidisciplinary applications	[156]
3 DP (PBF)	Ti6Al4V/CaP	N/A	Particles size distribution	–	Particle size distribution, flow behavior improved	–	[161]
3 DP (PBF)	316 stainless steel, 3% TiC	N/A	Size and percentage of nanoparticles	832	Mechanical strength improved	Engineering applications	[162]
3 DP (DED)	Aluminum, 0.5% carbon nanotube	N/A	Nanoparticles content	89	Microhardness improved	Advance applications	[163]
3 DP (DED)	Austenitic stainless steel	N/A	Different microstructures	804	Strength and ductility enhanced	Military structural parts, jet engine part and biomedical devices	[164]
3 DP (BJ)	Palladium and graphene oxide	N/A	Different time and cyclic number	–	Electronic conduction capability improved	Electrochemical supercapacitors	[165]
3 DP (BJ)	Graphene, polyvinyl alcohol powder	N/A	Surfactant concentration	–	Mechanical and electrical properties improved	Energy storage applications	[166]

*3 DP-3D printing and *4 DP-4D printing

with the PHA [167]. Moreover, filaments were produced for 3D and 4D printing, and it was observed that 3D printed specimens using milled TMP (thermomechanical pulp) fibers exhibited a 33% spike in tensile strength compared to specimens incorporating TMP fiber granulate due to better fiber alignment and lower agglomeration [167]. Agaliotis et al. [168] studied the tensile performance of 3D-printed PLA-based composites reinforced with fiber from nature. They reported that highest tensile properties were achieved when using a 0° raster angle and a flour content of 1% by weight [168].

The addition of 3 wt% maleic anhydride to composites containing 1 wt% flour resulted in a slight increase in maximal stress [168]. The maximal stress for these composites increased from 51.2 ± 4.4 to 63.9 ± 3.7 MPa. The research also found that most of the samples had a straight-line elastomeric behavior up to about 40 MPa, then a nonlinear behavior until they reached the highest stress [168]. After reaching the peak stress, a decrease in stress was observed as strain increased until the failure happens [168].

6.2 Physical properties

The physical attributes of composites derived from lignocellulosic materials, which are produced by 3D/4D printing techniques, comprise a diverse array of features. These traits include density, dimensional stability, and moisture absorption. Lignocellulosic composites often demonstrate reduced density in comparison to conventional materials, rendering them well-suited for lightweight applications. The preservation of dimensional stability is a crucial factor to address due to the inherent tendency of lignocellulosic fibers to absorb moisture, resulting in alterations in dimensions and diminished mechanical properties. The inherent color, texture, and aesthetic characteristics of lignocellulosic materials further have a role to determine the physical parameters of the printed composites. Moreover, wood fibers exhibit hygroscopic properties and undergo swelling when subjected to immersion in water. The aforementioned phenomenon may be used to fabricate structures with the ability to change form in the context of 3D printing. Therefore, it is plausible to think about the use of wood fiber reinforced filaments in the context of 4D printing. Ahmad et al. [171] investigated the physical properties of thermoplastic materials reinforced with the oil palm (*Elaeis guineensis*) fibers using FDM. According to their findings, an increased fiber content improved the moisture resistance of the composite samples [171]. Specifically, in a thickness swelling test, a composite with 7 wt% oil palm fibers demonstrated superior dimensional stability compared to samples with 3 wt% and 5 wt% fiber content [171]. Thus, adding these fibers to an ABS matrix improved the mechanical and physical properties of thermoplastic polymeric goods reinforced with oil

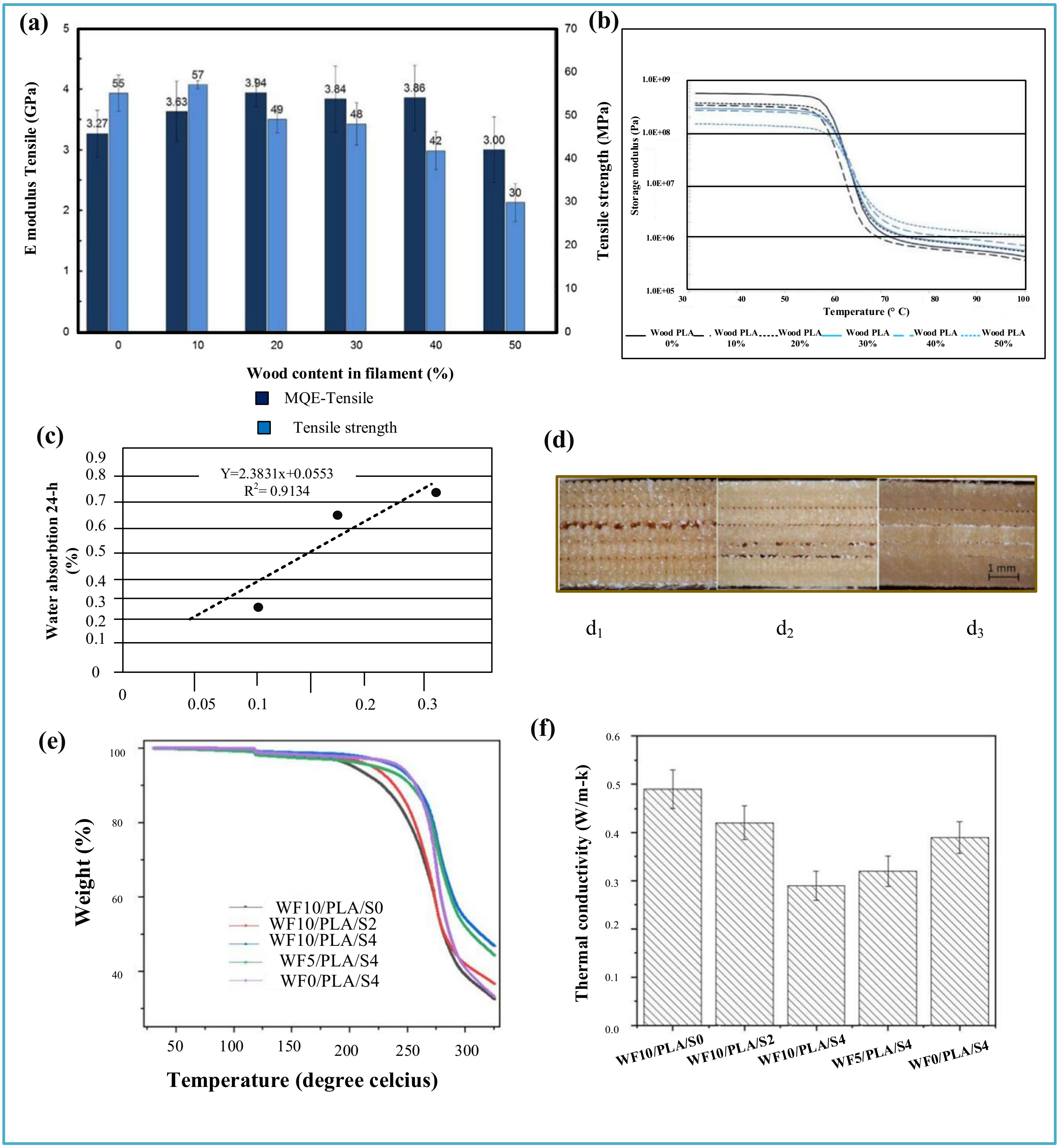

Fig. 12 **a** Tensile properties of 3D printed composite filaments from wood particle/PLA [169]; **b** influence of temperature on the storage modulus of 3D printed composite filaments from wood particle/PLA [169]; **c** regression investigation of 3D printed wood/PLA samples water absorption versus printing layer thickness [170]; **d** 3D printed wood/PLA specimens microscopic cross section [170]; **e, f** thermal stability and conductivity as a result of WF and the nano-SiO_2 [51]

palm fibers [171]. Considering the well-known susceptibility of lignocellulosic fiber reinforced composites are prone to absorb water, therefore a comprehensive investigation is essential to understand the water absorption characteristics of oil palm fiber reinforced composites [171]. Fiber pores allow water to pass across the interface. Therefore, increased loading of the fiber content in ABS matrix has the potential to enhance its moisture absorption capacity. In a separate study by Julia et al. [172], it was seen that lignocellulosic fiber reinforced composites quickly starts to absorb water, even after the fibers had been dried out before the printing operation. The printing process involved storing the filament in a drying chamber containing silica bags, which led to the discovery of a solution.

6.3 Thermal properties

The thermal properties of composites materials are often superior to traditional materials [173, 174]. However, the thermal characteristics of composites formed from lignocellulosic materials and produced by 3D/4D printing techniques are more satisfactory than conventionally manufactured composites. It is important to comprehend the behavior of these composites throughout diverse temperature settings in order to ensure their effective utilization in a multitude of domains. Therefore, an exploration of thermal conductivity, heat resistance, and coefficient of thermal expansion qualities are encompassed here further. Thermal conductivity is another crucial property that governs the heat conduction capabilities of 3D/4D printed composites derived from lignocellulosic materials. The thermal conductivity of composites may be affected by the presence of lignocellulosic fibers, which exhibit a lower thermal conductivity in comparison to the matrix material. The heat transfer channels and total thermal conductivity of a composite structure may be impacted by the dispersion and orientation of its fibers. The thermal conductivity of lignocellulosic composites may be modified by the manipulation of many factors, including the fiber content, fiber alignment, and the choice of matrix materials with distinct thermal conductivities. The coefficient of thermal expansion measures how much a substance expands or contracts with temperature. The coefficient of thermal expansion of 3D/4D printed lignocellulosic composites depends on various aspects, including the kind and quantity of fibers and the matrix material. Lignocellulosic fibers often exhibit lower coefficients of thermal expansion (CTE) in comparison to the matrix material. The use of these fibers in composites may effectively mitigate the total coefficient of thermal expansion of the composite. The anisotropic behavior of coefficient of thermal expansion is influenced by the arrangement and dispersion of fibers inside the composites. The consideration of CTE matching between the composites and other components or materials is of paramount importance in situations where dimensional stability has significant significance.

Heat resistance pertains to the capacity of 3D/4D printed composites derived from lignocellulosic materials to endure high temperatures without experiencing substantial deterioration or compromising their mechanical characteristics. Lignocellulosic fibers, especially those exhibiting elevated thermal stability, play an important role in enhancing the heat resistance performances of composite materials. The thermal stability of lignocellulosic fibers may be affected by several variables, including the kind of fiber, the amount of fiber, and the circumstances under which processing takes place. A comprehensive comprehension of the degradation processes and temperature thresholds of lignocellulosic composites is of utmost importance in the process of material selection and optimization of printing settings for particular high-temperature applications. Thermal analysis techniques are crucial for characterizing the thermal properties of 3D/4D printed composites made from lignocellulosic materials. One commonly used method is differential scanning calorimetry (DSC), which investigates parameters like melting and crystallization temperatures. Thermogravimetric analysis (TGA) is employed to assess thermal stability and decomposition temperature. These methods reveal composite thermal behavior, facilitating material selection and optimization. TGA/DTG analysis is particularly useful for identifying changes in mass caused by breakdown, oxidation, and release of volatile substances. While wood/natural fibers are susceptible to thermal degradation, incorporating nanoparticles can enhance thermal stability. This phenomenon has also been observed in wood/nanosilica/PLA 4D composites [56]. However, incorporating nanosilica or nanoalumina into the 4D composite system can significantly enhanced this thermal stability[56].

7 Application of sustainable 3D/4D printed products developed from lignocellulosic cellulosic materials: direct or indirect use

As mentioned above in Tables 1, 2, 3, and 4, 3D/4D printing techniques are remarkably adopted by current industries to fabricate multidisciplinary products or parts for direct or indirect use. However, recently, although 3D/4D printing has been widely adopted in multiple industries, in structural, aeronautical, biomedical, and electrical industries, the adoption of 3D/4D printing technique is comparatively more than any other domains due to the requirements of advanced features, current industrial demands, and revolutions. From an engineering and materials perspective, recent challenges include designing and manufacturing 3D/4D printed parts under optimal conditions, such as minimizing time, cost, and environmental impact. In addition to these critical requirements, achieving a better surface finish is essential for many industrial applications. The other requirements would differ based on the purpose of the applications. For example, in structural and aeronautical applications, the better load-bearing capacity of the material is a higher motivation than any other aspect [175, 176]. However, the biocompatibility of materials and enhanced cell adhesion are the most significant requirements in the biomedical domain [177].

Moreover, in electrical applications, electrical conductivity is the first and foremost requirement in any material. These crucial conditions undoubtedly can only be met by 3D/4D printing technology compared to traditional manufacturing processes [178]. This section will highlight the most recent and advanced applications of 3D/4D printing techniques manufactured from wood lignocellulose and natural fiber-reinforced composites in structural, aeronautical, biomedical, and electrical applications.

7.1 Structural application

In structural applications, wood lignocellulose or natural fiber-reinforced composites have emerged significantly in the recent times. For example, a recent study designed a flax fiber-reinforced biocomposite via FDM 3D printing, providing substantial mechanical performance and enabling its use in structural applications [179]. Similarly, another study designed a honeycomb cellular structure from wood and PLA filament through FDM technology and found suitable mechanical performance for structural applications. Moreover, this study also compared three combinations (PLA-PLA, wood-PLA, and wood-wood) and found that the wood-wood cellular structure provided less weight than others. In contrast, the PLA-wood combination is moderately better regarding mechanical strength (tensile strength—45 MPa) [180]. Another current study designed PLA and olive wood composite and obtained suitable mechanical performance. This study concluded that future research needs to be conducted to employ this biocomposite in structural applications since the flexural properties (flexural modulus-1.4 to 2.1 GPa) and hardness (30–40) properties are relatively low after adding the wood [31]. In Fig. 13a–d, some structural applications of 3D printed wood lignocellulose and natural fiber-reinforced composites are presented. Although there is a substantial research improvement in 3D printed wood lignocellulose-based structural applications [31, 179, 180], there are still some limitations. For instance, most studies used the thermoplastic matrix to fabricate the structures. In general, most thermoplastics are inherently brittle.

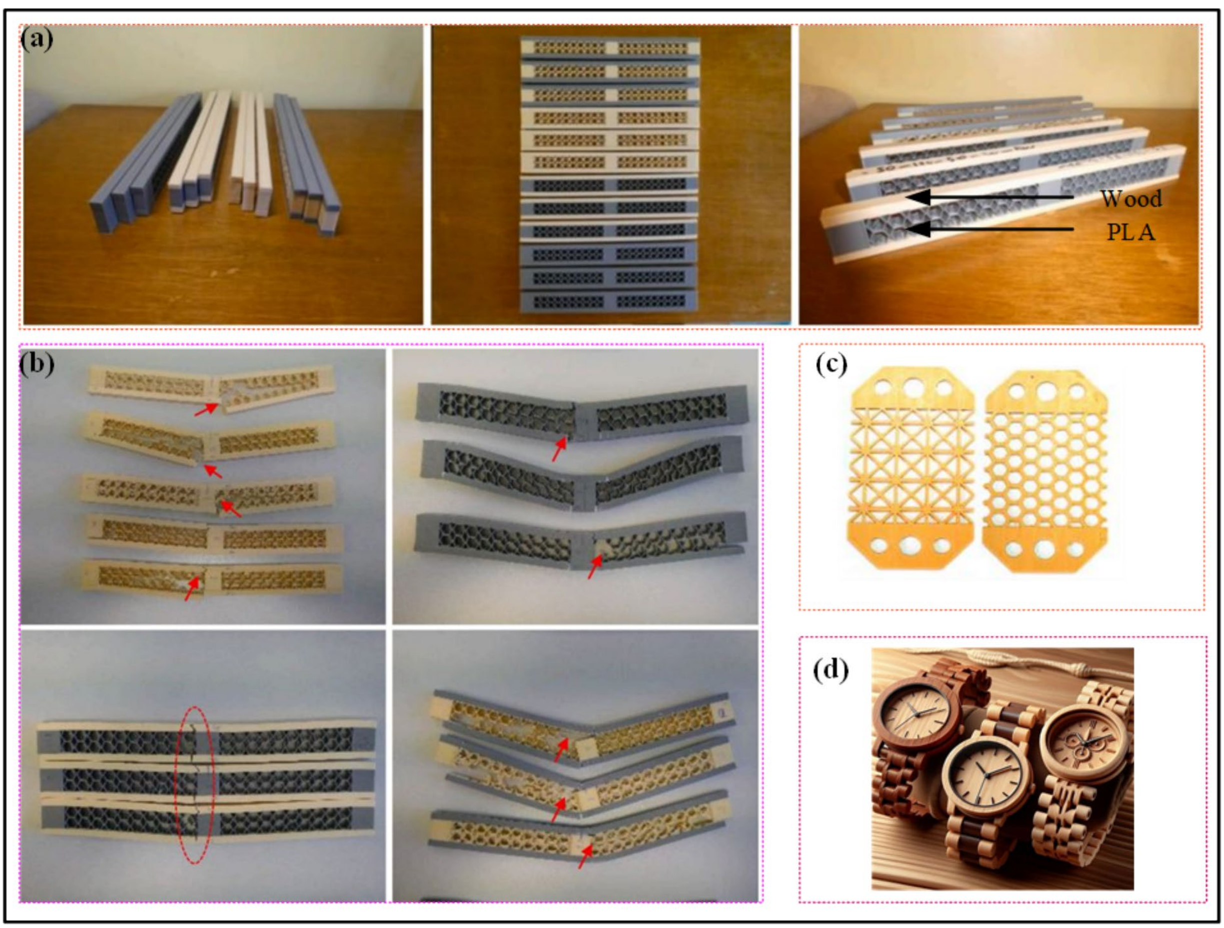

Fig. 13 **a** FDM printed timber-PLA composite honeycomb cellular structure for structural applications; **b** failure mechanisms in cellular structure in different combinations of timber and PLA composite [180]; **c** cellular structure 3D printed wood composite from wood-PLA [182]; **d** 3D printed wood watches for potential fashion items [183]

Therefore, they are unsuitable where large structural applications are required [181]. Apart from this boundary, extensive research is needed in durability, fatigue, and reproducibility for structural applications of the wood lignocellulose biocomposite [86].

7.2 Biomedical applications

Natural fiber or wood lignocellulose-based 3D/4D printed composite or nanocomposite also caught significant attention in the medical or biomedical industry, particularly in the pharmaceutical, blood vessels, bone scaffold, dental, wound healing, drug release mechanism, and so on [184–186]. This is likely because the natural fibers mostly have polymeric characteristics of natural origin and a favorable biocompatible nature [184]. However, from the engineering point of view, in cellulose fiber, the structure of plant walls, mainly supports in biomedical applications. Along with these, cellulosic 3D printed composites have vast applications in drug delivery mechanisms since they are able to bind a considerable amount of drugs on the surfaces and facilitate high payloads and optimum dosing power.

In Fig. 14a, the cellulose nanofibers preparation process with an example of polyurethane with cellulose nanofiber is presented. In the overall process, chemical treatments and ultrasonication play a crucial role in converting cellulose into nanocellulose. The chemical process specifically removed the impurities from the fiber surface, while ultrasonication helped to breakdown the particles into nanoscale. In recent times, nanocellulose and nanofibrillated cellulose based 3D printing composite materials remarkably using in biomedical applications [187]. For example, this study designed a scaffold and compared the toxicity level of 3D printed nanofibrillated cellulose composite at the different concentration levels of nanofibrillated cellulose and found that cellulose-based 3D printing composite does not have any toxicity and also has suitable mechanical properties for bone-scaffold [186], illustrated in Fig. 14m. However, Fig. 14a presents an example of waterborne polyurethane and cellulose nanofiber. However, this combination depicted in Fig. 14h, i scaffolds also provided satisfactory cell viability (%) at different days. Moreover, the study found that the presence of carboxyl groups in nanocellulose significantly influenced the composites viscosity, resulting in better overall mechanical and biological performance. Hence, it would be a suitable application in tissue engineering scaffolds and medical devices [185]. Similarly, another study has highlighted a nanocellulose-based composite and proposed its applications in valve implantation, as depicted in Fig. 14b, c [188]. However, in Fig. 14d, a 3D bioprinted auricular cartilage regeneration model is designed, and it demonstrates considerable cell viability, as depicted in Fig. 14l. In this study, nanofibrillated cellulose and alginate were used to prepare the bioink solution [189]. In Fig. 14e, 3D-printed membranes, which are fabricated from bacterial cellulose, are presented. This membrane is enabled to facilitate wound healing, as depicted in Fig. 14f, g. Moreover, the study proposed its applications in medicine and odontology, particularly in venous ulcers in the kidneys, and prospective applications in diabetic ulcers [190]. However, the direct incorporation of wood cellulose in biomedical applications has also been recently studied. For example, a study designed a neck collar (orthosis) using FDM 3D printing fabricated from PLA and agricultural wood waste and discovered its lightweight properties for potential biomedical applications [191].

7.3 Aeronautical applications

Fuel consumption is the main motivation that leads the aerospace industry to reduce the weight of structures and components and produce lightweight structures since fuel prices have increased drastically in recent times. In addition, the lightweight structure enables more efficient fuel utilization, allowing it to cover longer distances with improved fuel efficiency. For example, this study highlighted that after utilizing polymer-reinforced composite in Boeing 787, the fuel efficiency increased by around 20% [192]. Moreover, to address fuel consumption, the aviation industry's recent goal is to reduce carbon emissions and manufacturing costs. This challenge can be effectively addressed through the use of natural fiber-reinforced 3D/4D composites. Additionally, the inherent satisfactory strength, stiffness, corrosion resistance, superior fatigue performance, and the ability to fabricate complex shapes are some of the benefits of using natural fiber-reinforced 3D/4D products in aeronautical applications [193].

However, there are some other limitations in natural fiber-reinforced composites, such as low flame retardancy and not satisfactory mechanical strength compared to other composites like polymer-reinforced composites and metal matrix composites. Considering these two limitations, these studies suggested that biocomposite could only be suitable in aircraft interior structures like cabins, seats, floors, and decks where there is a low possibility of fire risks [192, 194]. In recent times, limited studies have addressed the use of 3D/4D painted natural fiber or wood composites in aeronautical applications. In a current research, a hybrid composite honeycomb core was developed through 3D printing using natural fibers [195]. The core material consisted of lignin-acrylonitrile butadiene styrene (ABS), while the skin was made from industrial hemp combined with aluminized glass fiber epoxy. This composite structure was specifically designed for applications in morphing wing and unmanned aerial vehicle systems [195]. Nevertheless, some research has broadly emphasized that biopolymers such as PEEK,

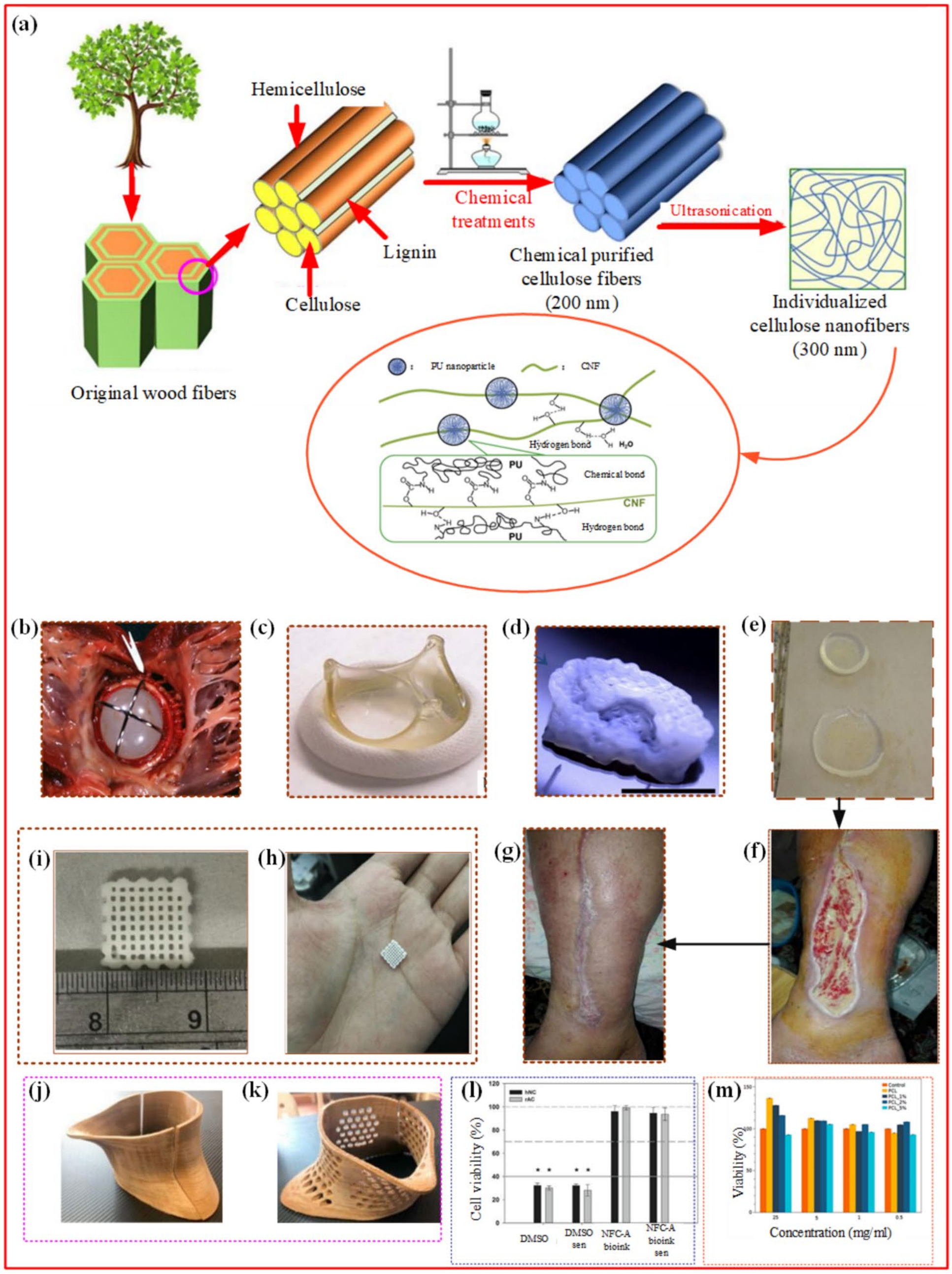

Fig. 14 **a** Cellulose to nanocellulose breakdown process and an example of nanocellulose chemical reaction with polyurethane [188]; **b-c** heart valve from nanocellulose polyurethane composite [188]; **d** 3D bioink printed nanofibrillated cellulose and alginate composite for auricular cartilage application [189]; **e** 3D printed bacterial cellulose for membrane application; **f, g** improvement after 3D printed membrane implantation [190]; **h, i** 3D printed nanocellulose fiber and polyurethane composite for scaffold applications [185]; **j, k** 3D printed neck orthosis from hemp filament [191]; **l, m** cell viability of cellulose composite at different concentration level [186, 189]

ABS, and PLA can find suitable applications in the aeronautical field when combined with other polymers [196–198]. For example, this study found that the 3D printed PEEK and continuous carbon fiber reinforced composite exhibited satisfactory shear and flexural strength over 35 MPa and 480 MPa, respectively, which could be an alternative to metal material in aviation and aerospace applications. In Fig. 15a–c, conventionally fabricated natural fiber-reinforced composite for commercial interior parts and acoustic panels is illustrated. In addition, in Fig. 15d–i, 3D printed composite part applications in aeronautical, drone, and space are presented. Figure 15j, l cashew nutshell extracted 3D printed ABS composite honeycomb core for aeronautical applications are depicted.

7.4 Electronics

3D/4D printed natural fiber or wood-based lignocellulosic composite materials also hold potential applications in the electronics industry due to their notable electrical conductivity [200]. However, various studies have developed different approaches and combinations of materials for 3D/4D printed parts, which can facilitate tailoring the overall electrical conductivity of these sustainable composites. For example, this

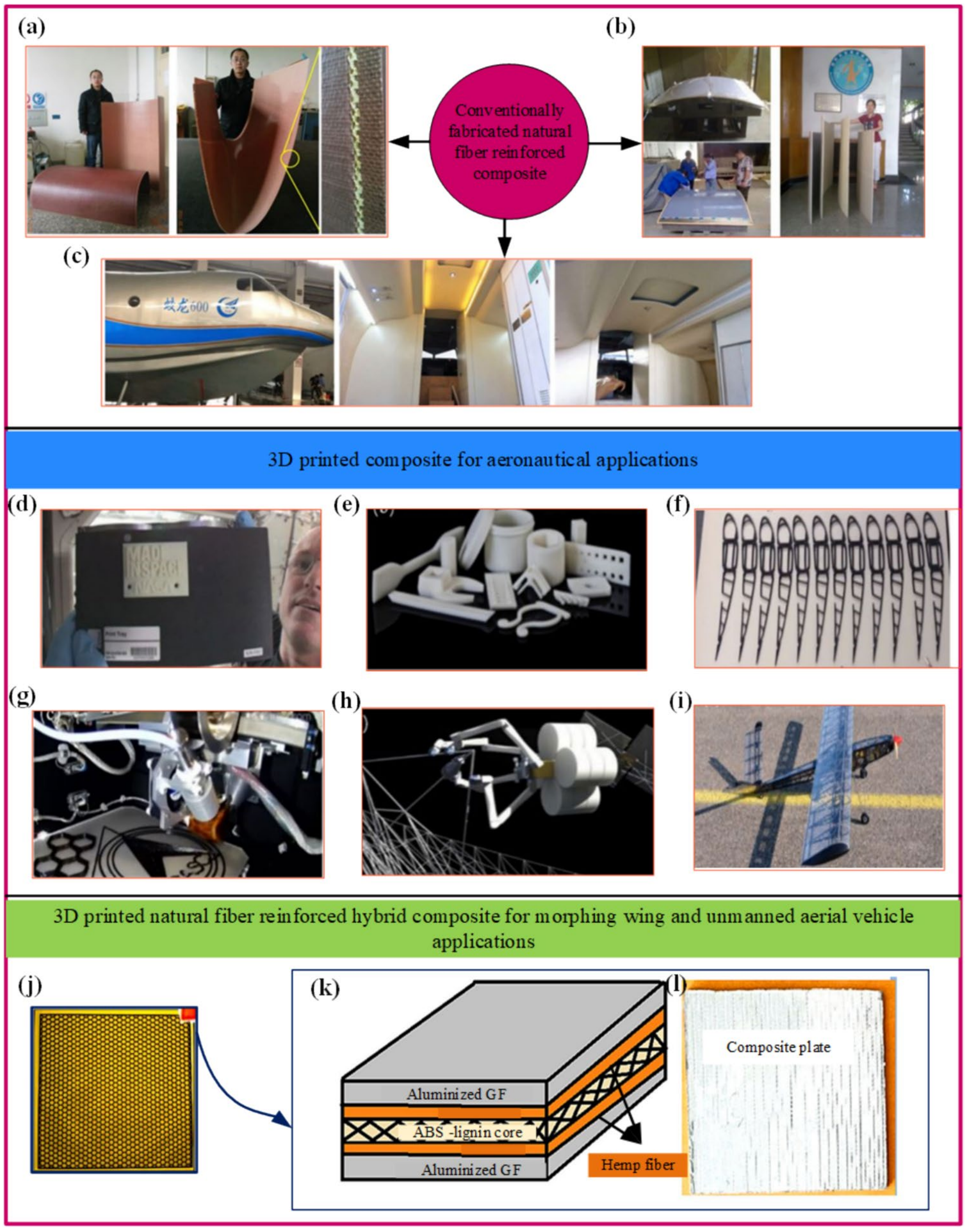

Fig. 15 Conventionally fabricated **a** plant fiber reinforced composite for commercial aircraft interior part; **b** acoustic panel; **c** seaplane interior made from plant reinforced composite [199]; **d, e** 3D printed parts in space applications; **f, g** 3D printed carbon fiber PLA composites for aircraft structures; **h** spiderfab for fabricating large structures; **i** 3D printed carbon fiber reinforced composite for drone structures [196]; and **j, l** cashew nutshell extracted 3D printed honeycomb core for morphing wing and unmanned aerial vehicles [195]

study fabricated 3D polypyrrole-nanocellulose composite electrodes, which exhibited outstanding mechanical properties, making them suitable for energy storage applications [201]. Similarly, this study fabricated wood nanocellulose polypyrrole composite, obtained significant electromechanical properties, and proposed suitable applications in ion exchange and energy storage [202]. In Fig. 16a–e, the applications of this nanocomposite are illustrated. Along with natural fibers, adding carbon fiber to the 3D printed parts can substantially improve the electrical conductivity of the final components. This study fabricated a bio composite by SLS 3D printing machine combining pinewood powder with a resin called phenolic, performed a carbonization process, and found the biocomposite provides enhanced electromechanical properties [203]. In Fig. 16c, the electrical conductivity of this composite is presented. Apart from the carbonization process, there are alternative methods like acid washing that can enhance the electrical conductivity of materials. For instance, a study investigated the electrical conductivity of carbon fiber-reinforced polypyrrole-cladophora nanocellulose composite electrodes that underwent acid washing compared to water washing. The results of the study showed that the electrodes subjected to acid washing exhibited improved conductivity [204]. This study fabricated a unique hybrid composite using poplar wood, PLA, and graphite through FDM technology [205]. It compared the electrical resistivity, which is inversely related to electrical conductivity; a decrease in resistivity corresponds to an

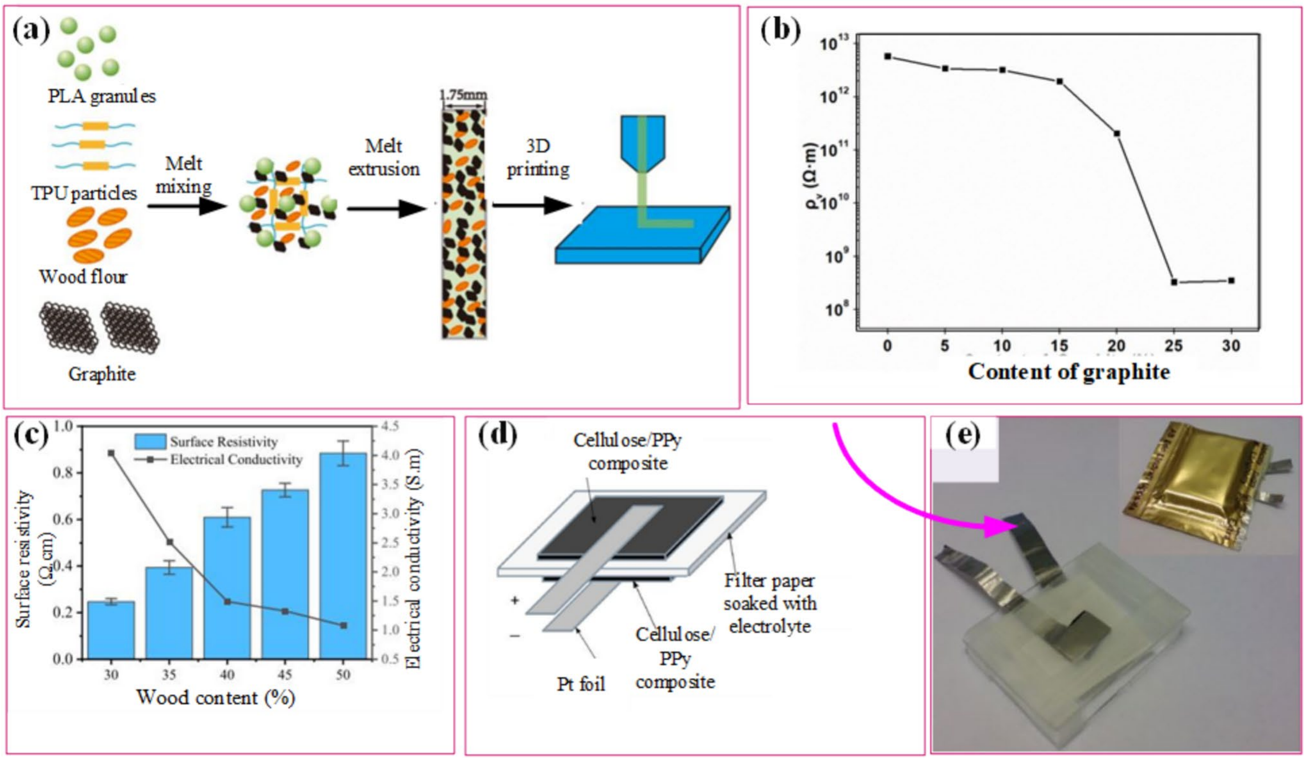

Fig. 16 **a** Fabrication process schematic for a PLA-TPU-wood-graphite composite with enhanced electrical conductivity [205]; **b** influence of graphite content on the electrical resistivity of the composite [205]; **c** impact of wood content on the electrical conductivity of a wood carbonized composite produced by SLS [203]; **d, e** application of nanocellulose composite paper in battery technology [206]

increase in conductivity. The study observed a reduction in electrical resistivity. Furthermore, it concluded that the proportions of graphite and wood content significantly influence the electrical conductivity. In Fig. 16a, b, the fabrication process of this exceptional composite and electrical resistivity is demonstrated.

8 Challenges, opportunities, and economical importance of 3D/4D printed composites developed through integrating lignocellulosic materials

The current advancements in the manufacturing industry indicate that the present time is dominated by the use of 3D/4D printing technology. Currently, there is a predominant emphasis on synthetic polymers and fibers, which are closely linked to significant environmental consequences [207, 208]. There is a growing need for printed products with a greater focus on sustainability. However, there exist several obstacles that must be addressed in order to successfully integrate natural printable materials into the process of 3D/4D printing [59, 209]. The prototype of automotive and aerospace components is becoming more costly and time-consuming for their intricate designs and/or organic shapes. Therefore, the use of 3D/4D printing methods entails the increasing substitution of metal components with polymer composites in order to mitigate the weight in automotive and aerospace sectors. In recent investigations, it has been seen that the use of polymer-oriented composites in engine components has resulted in enhanced fuel economy [210]. The use of 3D/4D printing is a significant method for producing polymer composites with functional gradients, achieved by the incorporation of particle reinforcement, fiber reinforcement, and nanostructures. Composite materials have the ability to be tailored according to specific requirements by manipulating the volume fractions. Consequently, the mechanical qualities may be tuned to suit the precise demands of the given application. One notable benefit associated with 3D/4D printing composites is their thermal stability [211, 212]. Some recent challenges of 4D printing and associated products are shown in Fig. 17.

Polymer composites designed for automotive or aerospace applications offer a diverse selection of materials capable of withstanding high demands such as temperature resistance, strength, and strength-to-weight ratio. However, for the betterment of future generations and to promote environmentally sustainable practices, it is crucial for researchers in the field of 3D and 4D printing to prioritize the use of natural

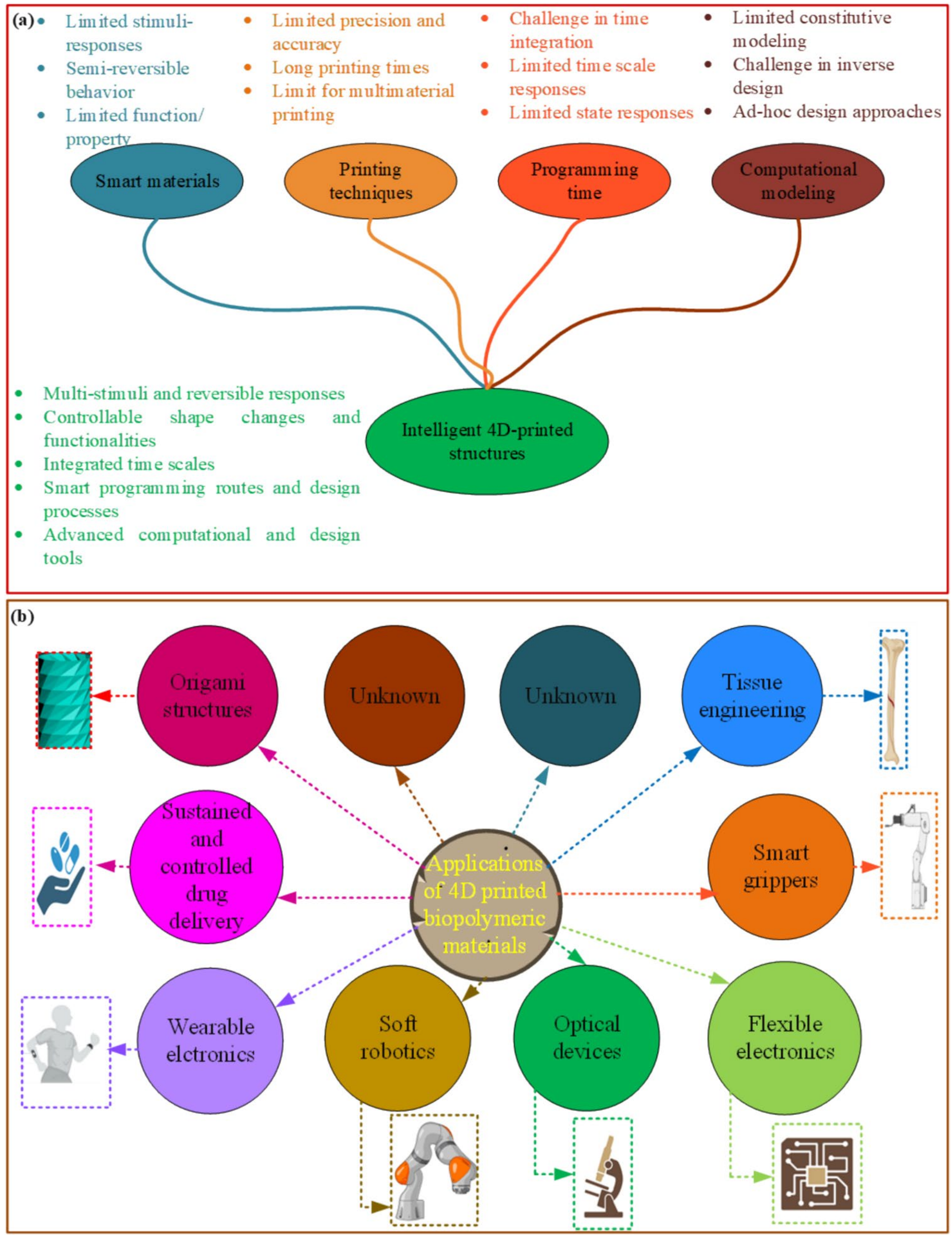

Fig. 17 **a** Recent challenges of 4D printing; **b** future domains of 4D printed natural fiber-based composites or wood lignocellulose composites [213]

materials. There is also a need to channel research efforts towards the development of green composite materials specifically designed for 3D and 4D printing technologies. Currently, the number of research programs dedicated to this area is limited, highlighting the importance of expanding research efforts in this direction. The presence of natural fillers in green polymer composites presents a promising avenue for future academic study, with the primary objective of mitigating the reliance on non-renewable petroleum-based resources. Based on the aforementioned criteria, current and prospective research endeavors focus on the identification of the most appropriate biodegradable matrix and the optimization of processing parameters. Here the associated challenges, opportunities, economic importance, and significance of 3D/4D printed composites made of lignocellulosic biomass have been addressed further:

8.1 Challenges

The incorporation of lignocellulosic materials into 3D/4D printed composites poses numerous challenges. Lignocellulosic fibers often exhibit heterogeneous geometries and intricate structures, hence posing difficulties in attaining uniform dispersion and alignment within the composite material [211]. The aforementioned phenomenon has the potential to impact the mechanical characteristics and overall efficacy of the printed objects. In Fig. 17a, b, different challenges and applications of 4D-printed wood lignocellulose or natural fiber-based composites are depicted. However, the unknown domain needs to be attempted for the advancement of future materials. Moreover, it is notable that lignocellulosic materials have a tendency to absorb moisture, a characteristic that might results in the dimensional instability and a decrease in mechanical strength [121, 214]. Moreover, the availability of recycled and biobased PLA, PET, ABS, PP, HDPE, LDPE, and PS has been identified for the purpose of fabricating 3D FDM filaments. PLA, ABS, and their associated composites are extensively used within the business sector because to their affordability and broad accessibility. On the other hand, materials that are recognized for their limited suitability for 3D printing, such as PET, LDPE, HDPE, PS, PP, recycled materials, and their composites, exhibit significant potential when appropriate processing treatments (pre-, in-, and post-process) are incorporated. Several treatments can be employed to address the issue. These treatments include low-temperature plasma treatment, coating, surface heating prior to layer formation, utilization of compatibilizers, plasticizers, and fillers, as well as altering the surface through coating. In addition, it is crucial to conduct comprehensive characterization and have a deep knowledge of the characteristics of lignocellulosic sources, such as different wood species or agricultural leftovers. This is essential in order to achieve consistent and predictable outputs in the process of printing.

The 3D/4D printing is the most favorable option for fabricating natural fiber or any composite materials than traditional manufacturing processes [61, 215–217]. Despite having enormous positive effects, AM can have some potential challenges for fabricating natural fiber composite materials. For example, in FDM 3D printing machines, nozzle clogging is a regular troublesome during producing the natural fiber-reinforced composites. Aside from this, another obstacle is to remove or reduce the polar group (-OH) from the fiber surface since most of the natural fiber may have wax and moisture. As a result, this polar group can reduce the fiber and matrix bonding which may impact the performances of composites. Therefore, a possible solution would be to remove the unwanted substances from the fiber surface through chemical reactions, which would facilitate better fiber and matrix bonding in the FDM-printed composite parts or materials [218]. In addition, in most 3D/4D printing methods, manufacturing parameters have a crucial contribution or influence on mechanical performances. An imperfect combination may provoke some voids, resulting in poor mechanical performance [219]. In this case, using some data analysis methods or optimizing the manufacturing parameters can improve the mechanical properties [71]. Compared to the FDM manufacturing method VP is more convenient since it is a nozzle-free technique, and the resolution is relatively better in the fabricated parts [220]. However, in this method commonly faces three general challenges, namely, dimensional accuracy, shrinkage, and poor mechanical properties, which can be observed in the fabricated parts. These challenges mainly occur due to imperfect printing parameters, slicing software, support parameters, and a lack of post-processing [221]. Besides these significant difficulties in the VP method, uneven laser or projector sources and their directions, as well as improper viscosity in the resins, may significantly affect the mechanical properties. These aspects need further discussion in order to overcome these issues and obtain better-printed parts without compromising any mechanical properties [82]. This applies similarly to fabricated composites or vegetable fiber-reinforced composite products, which require focused attention in future studies [222]. In the previous discussion, it was emphasized that the SLS method is particularly well-suited and has been increasingly utilized for fabricating natural fiber-reinforced composite materials compared to other methods such as SLM, and other similar techniques. However, this method can have potential challenges, like mixing the powder in a perfect ratio and achieving better interfacial interactions between the fiber and matrix [223]. Additionally, the aging of powder at printing temperatures and storage time is another difficulty that may negatively affect the printing quality and mechanical properties [223]. Moreover, surface modification and reducing the porosity in the composite parts also need attention to study for improving the mechanical or thermal properties.

8.2 Opportunities

Despite the inherent difficulties, the incorporation of lignocellulosic materials into 3D/4D printed composites has substantial prospects. The use of lignocellulosic fibers into composites has been shown to significantly improve their mechanical characteristics, resulting in increased strength, stiffness, and durability [2]. The inherent renewability of lignocellulosic materials is in accordance with the growing need for sustainable manufacturing practices, hence creating opportunities for environmentally friendly and biobased solutions. Furthermore, lignocellulosic fiber-based 3D and 4D composites possess distinctive visual characteristics, including wood-like textures and natural color variations. These aesthetic qualities make them highly sought-after in contexts where visual appeal holds significance. Applications that value the natural and organic appearance of woody appearances, such as interior design, furniture manufacturing, and decorative elements, can benefit from the use of lignocellulosic 3D and 4D products. The incorporation of these materials adds a touch of authenticity and natural beauty, enhancing the overall visual appeal of the end products. The capacity to accurately manipulate the alignment and arrangement of lignocellulosic fibers inside the composite framework enables the creation of unique functionality and customizable material characteristics [224, 225]. Moreover, in the future, Micro-CLIP technology is also expected to play a significant role in 3D and 4D printing with natural fibers or wood, offering promising and multifaceted applications. As research progresses, Micro-CLIPs can be developed to work seamlessly with a broader range of natural fibers and wood composites, enhancing the structural integrity and performance of printed objects, thereby making them suitable for various applications. Future advancements may also focus on creating customizable Micro-CLIPs tailored to specific designs and applications. This flexibility will empower designers to innovate new forms and functions, leading to more complex and efficient products [226]. Additionally, the integration of Micro-CLIPs with smart materials could enable dynamic responses to environmental changes, such as moisture or temperature. In 4D printing, this capability would facilitate the creation of structures that adapt over time, enhancing functionality and user experience. The versatility of Micro-CLIPs could see their applications extend beyond 3D and 4D printing to industries such as automotive, aerospace, and construction. Their ability to securely fasten components could revolutionize assembly processes in these fields.

8.3 Economic importance

The incorporation of lignocellulosic elements into 3D/4D printed composites has substantial economic significance. Lignocellulosic materials, such as wood and agricultural leftovers, and natural fibers are widely available and often regarded as surplus or secondary outputs of many businesses. Through the use of easily accessible resources, there is possibility for a substantial decrease in the cost of raw materials. This, in turn, positions lignocellulosic composites as a viable and economically advantageous choice when compared to conventional materials. Additionally, the use of lignocellulosic composites in the context of 3D/4D printing has the potential to facilitate the fabrication of components that are characterized by their low weight. This attribute contributes to the reduction of material consumption, transportation expenses, and energy demands throughout the whole of the manufacturing procedure. This phenomenon may lead to enhanced operational effectiveness and decreased production costs. Moreover, the size of the worldwide 4D printing market was reported to reach USD 65.4 million by the year 2019, which is going to significantly increase by 2025 for wood and other lignocellulosic material-based 4D composite products [227]. According to the report from the MarketsandMarkets database, the total market size of 3D and 4D printed products is expected to grow from USD 399.7 billion in 2024 to USD 866.5 billion in 2029, at a Compound annual growth rate (CAGR) of 16.7% [228]. This significant market expansion highlights the growing demand and opportunities for lignocellulosic wood and natural fiber-oriented 3D and 4D printing technologies.

9 Conclusion and future perspectives

Lignocellulosic materials offer several advantages, such as their renewable nature and abundant availability of natural resources, which reduce reliance on fossil fuel-derived polymers and enhance the sustainability of manufacturing processes. Incorporating lignocellulosic materials into 3D/4D printing results in a decreased carbon footprint, as it requires less energy consumption and generates fewer greenhouse gas emissions. This not only supports environmental sustainability but also promotes the adoption of a circular economy. The research identifies noteworthy vegetable fibers and woods that are suitable for 3D/4D printing, enabling producers to select the most appropriate materials based on their unique characteristics. The study highlights further the sustainable potential of lignocellulosic wood and natural fiber-based materials in 3D/4D printing, offering a favorable alternative to conventional polymer composites and encouraging a more eco-conscious approach to production. The study also entails the characteristics, manufacturing processes, mechanical properties, and potential applications of these environmentally-friendly 3D/4D printing materials.

Compared to synthetic fibers, lignocellulosic materials have a lower impact on environments. Incorporating lignocellulosic materials into 3D/4D printing can significantly reduce the environmental impact associated with production, aligning with the principles of a circular economy. Various approaches, such as binder jetting, powder-based printing, and filament extrusion, show promise, but further investigation is needed to improve these processes and achieve consistent and reliable outcomes. The mechanical and physical characteristics of 3D/4D printed composites made from lignocellulosic wood and vegetable fibers have gained significant attention. These materials demonstrate comparable or even superior tensile strength, flexural properties, impact resistance, and dimensional stability compared to conventional polymer composites. However, further research is necessary to enhance material compositions and processing methods for consistent and predictable results.

The future prospects for 3D/4D printed composites from lignocellulosic resources are also highly promising. Ongoing research and development efforts focus on addressing issues related to variability and moisture susceptibility of lignocellulosic fibers. This involves optimizing processing methods, applying surface treatments, and developing material formulations to improve overall performance and reliability in additive manufacturing. Advancements in additive manufacturing technologies, such as multi-material printing and hybrid printing processes, offer opportunities to combine lignocellulosic materials with other functional materials, expanding their potential applications across various sectors. The integration of 4D printing, enabling self-transformation and shape-changing properties over time, also presents intriguing possibilities for lignocellulosic composites, allowing for flexible and adjustable structures and broadening their utility. Overall, lignocellulosic wood and natural fiber-based materials hold significant promise in the field of 3D/4D printing, providing sustainable and environmentally friendly alternatives to conventional polymer composites. Further research and development are necessary to improve material compositions, enhance processing methods, and explore novel applications. By leveraging the properties of these materials, we can actively contribute to a more environmentally friendly future in the industrial sector and promote the ideals of a circular economy. The following characteristics of sustainable 3D/4D printing products require significant attention to foster the growth of this promising industry and contribute to the development of an environmentally friendly world.

- Different processing methods, such as vat polymerization, powder-based printing, and filament extrusion, are currently under investigation for the 3D/4D printing of lignocellulosic materials. The primary objective is to understand the advantages and disadvantages of each method in order to optimize material and application requirements.
- The incorporation of binders, additives, and strengthening agents has the potential to enhance the printability and mechanical properties of 3D/4D printed objects made from lignocellulosic materials. This improvement includes enhancing interlayer adhesion, dimensional stability, and overall functionality of the printed components.
- Studies on the mechanical and physical properties of composites made from lignocellulosic wood and vegetable fibers have demonstrated their ability to achieve tensile strength, flexural properties, impact resistance, and dimensional stability that are comparable to or even superior to those of polymer composites. However, further research is needed to refine material compositions and processing methods in order to achieve more consistent and reliable results.
- The future prospects for utilizing 3D/4D printed composites made from lignocellulosic resources are promising. Ongoing research and development efforts aim to address challenges related to the variability and moisture susceptibility of lignocellulosic fibers by improving processing techniques, applying surface treatments, and developing optimized material compositions.
- Advancements in additive manufacturing technologies, such as multi-material printing and hybrid printing processes, offer opportunities to combine lignocellulosic materials with other functional materials, thereby expanding their range of potential applications.
- The integration of 4D printing, which enables self-transformation and shape-changing properties over time, presents exciting possibilities for lignocellulosic composites, allowing for the creation of dynamic and flexible structures.
- Vegetable fiber and lignocellulosic wood products have potential uses in various sectors related to 3D/4D printed items. Their lightweight composition and strong mechanical performance make them well-suited for constructing complex geometries and reducing material waste.
- Given the increasing emphasis on sustainability and environmental consciousness, the use of lignocellulosic materials in 3D and 4D printing can effectively contribute to achieving these goals.
- To fully harness the capabilities of 3D/4D printing using lignocellulosic wood and natural fibers, continued research and development activities aimed at enhancing material compositions, refining printing techniques, and expanding their range of applications are imperative. The adoption of sustainable practices in 3D/4D printing can have a significant impact on the industrial sector's efforts to protect the environment and advance a circular economy by minimizing ecological harm.

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