#### **Review Article**

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# Wood-derived high-performance cellulose structural materials

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**Abstract:** The threats of nonrenewable energy consumption and environmental disruption caused by the extensive use of metals and polymers derived from petroleum have prompted the development of eco-friendly, high-performance, and long-lasting structural materials. After various treatments, cellulose materials exhibit exceptional properties such as high strength, fire resistance, hydrophobic properties, and thermal stability. Cellulose-based structural materials have excellent mechanical strength and the distinct advantages of being lightweight, inexpensive, and energy efficient. This review summarizes the recent progress in the preparation methods and properties of high-performance cellulose structural materials such as high-strength cellulose structural materials, thermal insulation cellulose structural materials, flame-retardant cellulose structural materials, hydrophobic cellulose structural materials, cellulose structural material with electrical properties, and other cellulose structural materials. The future of high-performance cellulosic structural materials and the prospective of their development are concluded.

**Keywords:** cellulose, structural material, composite, mechanical properties, strength

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#### 1 Introduction

Human existence and social development have been predicated based on materials. Structural materials are mechanical property-based materials that are used to construct stress components (1-3). The material for the structure often needs to meet certain physical or chemical requirements, such as thermal conductivity, corrosion resistance, and oxidation resistance, among others (2,4-6). Due to their superior mechanical properties, steel, ceramics, cement, and plastic have become the most widely used structural materials (7-10). However, these structural materials either have a negative impact on the environment and are nonrenewable (such as petroleum-based plastics, alloys, and steels) or are difficult to manufacture and, consequently, costly (such as composite) (11-13). Furthermore, their relatively high thermal conductivity prevents them from simultaneously meeting the requirements of modern structural materials for mechanical properties and thermal insulation. With the increasing of the global population, the production of traditional structural materials has increased dramatically, with high energy costs and high environmental impact, posing a formidable obstacle to the sustainable development of humanity (14). Consistently, there has been a significant interest in lightweight, sustainable, and environmentally friendly structural materials with exceptional properties that could be applied in a variety of fields (15).

The abundance of natural polymer materials on earth has the potential to provide alternative material answers to the formidable challenges of global pollution control (16). Cellulose, which consists of numerous glucose molecules linked by  $\beta$ -1,4-glycosidic bonds, is a linear polysaccharide with rigid molecular chains (17,18). It is the most common natural polymer substance. With approximately one trillion tons of production from plants worldwide, cellulose has become the largest reservoir of organic carbon (19–21). Because of its renewability, usability, nontoxicity, economy, biocompatibility, biodegradability, ecofriendliness, and thermal and chemical stability, cellulose has received particular attention (22,23). Because of its

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superior specific modulus and strength to most metals, composites and ceramics, cellulose has the potential to build novel structural materials as alternatives to traditional structural materials (24–26).

Thousands of years ago, long before it was discovered by science, cellulose has been utilized by humans as a structural material in the form of wood (27). However, the majority of natural woods have low strength, high flammability, poor durability, and high perishability, limiting their competitiveness in modern social fields such as high-rise buildings, harsh environments, and the automotive industry (28–30). In recent years, with the advancement of society and technology, scientists have processed cellulose materials using physical or chemical techniques to produce a variety of cellulose structural materials with superior performance (31-33). After undergoing various treatments, cellulose-based structural materials can acquire superior qualities such as high strength, low flammability, high durability, and high hydrophobicity, while retaining their original characteristics of low cost, environmental friendliness, and lightweight. The large-scale production and application of cellulose structural materials can significantly reduce the impact of conventional structural materials on energy and the environment, thereby addressing the challenge to sustainable development. Recent developments in cellulose and cellulose-based high-strength structural materials, flame-retardant structural materials, thermal insulation structural materials, hydrophobic structural materials, cellulose structural material with electrical properties, and other cellulose structural materials are reviewed in this work (Figure 1).

### 2 Fabrication of high-strength cellulose structural materials

Because structural materials are frequently fabricated into force-bearing components, the exceptional mechanical property is the foundation of structural materials. Cellulose materials can attain a high level of strength after being treated by different methods such as chemical modification, cold pressing, and hot pressing. Even after a series of treatments, cellulose material remains relatively light and has a low density.

### 2.1 Delignification-densification

Wood and bamboo, which are primarily composed of cellulose, hemicellulose, and lignin, are the two essential

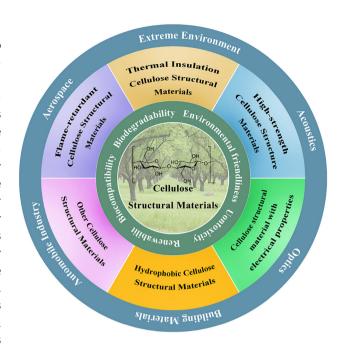


Figure 1: The advantages of cellulose structural materials as well as the preparative approaches and properties of high-strength cellulose structural materials.

cellulosic materials (34). In their cell wall, cellulose is the primary structural component responsible for maintaining cell shape, hemicellulose functions as a compatibilizer, and lignin acts as a cementitious component to harden (35). These cellulosic materials have increased porosity and decreased rigidity after lignin removal, which facilitates subsequent processing. The subsequent densification can improve the bulk density of the fibers, resulting in excellent mechanical properties (36,37).

Song et al. developed an easy and efficient method for constructing high performance cellulose structural material (38). Natural wood with porous structure was chemically treated in an aqueous solution of sodium hydroxide and sodium sulfite to remove lignin and hemicellulose, and then hot-pressed at 100°C to produce densified wood. Due to the complete collapse of the cavity and porous cell walls, the thickness of densified wood was reduced by approximately 80% and the density was increased by a factor of 3 compared to natural wood. Cellulose nanofibers in densified wood were densely packed, with a large interface area, resulting in the formation of a large number of hydrogen bonds between adjacent nanofibers. The densified wood had excellent mechanical properties that were superior to traditional wood and structural materials commonly used in engineering, such as steel, alloys, and plastics. Compared to untreated natural wood, the densified wood had 10.5 times higher tensile strength and 9 times higher fracture work (about 587 MPa and 3.9 MJ·m<sup>-3</sup>), respectively. Furthermore,

due to the inherent lightness of cellulose, its specific strength (451 MPa·cm<sup>-3</sup>·g<sup>-1</sup>) was even greater than that of lightweight titanium alloys (approximately 244 MPa·cm<sup>-3</sup>·g<sup>-1</sup>). Densified wood is a good substitute for structural metals and alloys due to its high toughness, high strength, and lightweight.

Burgert et al. (39) optimized the delignification and densification process, using  $\rm H_2O_2$  and  $\rm CH_3COOH$  to delignify natural wood and a combination of compression and transverse shearing to produce dense cellulose bulk materials. The obtained dense cellulose bulk material had excellent mechanical properties and could be prefabricated into a

variety of shapes. Wang et al. (40) prepared delignified oxidized hot-pressed wood by adding oxidation and moisture content adjustment steps between delignification and densification process. Oxidation and moisture content regulation contribute to the formation of new hydrogen bonds during hot pressing, which makes the wood much stronger.

He et al. produced a strong white wood with excellent performance by the delignification—densification method (Figure 2a) to develop high-performance structural materials for use in tall buildings (41). The preparation of

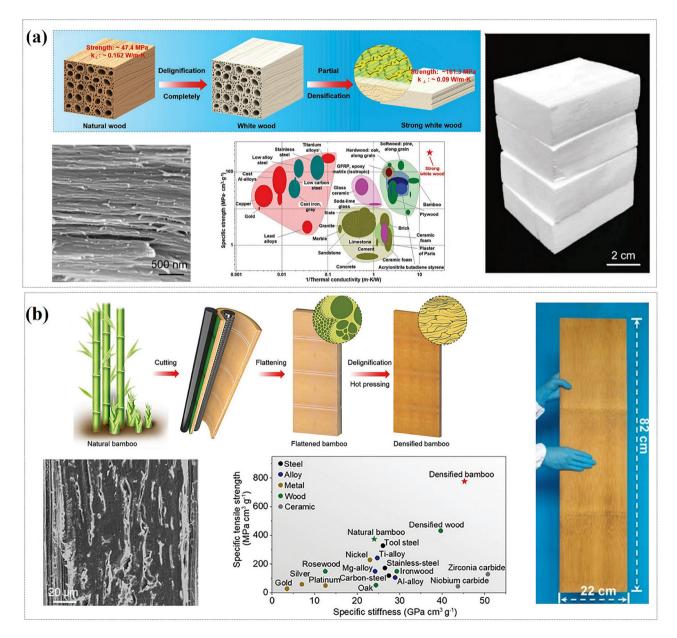


Figure 2: (a) Preparation process, photograph, SEM image, and the excellent performance of strong white wood. Reproduced with permission from the study by He et al. (41). Copyright © 2019; Wiley-VCH. (b) Preparation process, photograph, SEM image, and the excellent performance of densified bamboo. Reproduced with permission from the study by Li et al. (45). Copyright © 2020; Wiley-VCH.

**Fable 1:** Preparation and properties of delignification—densification cellulose structural materials

Raw materials	Raw materials Preparation methods	Delignification reagent	Delignification reagent Densification condition Tensile strength (MPa) Fracture work (MJ·m <sup>-3</sup> ) Ref.	Tensile strength (MPa)	Fracture work (MJ·m <sup>-3</sup> )	Ref.
Oak wood	Delignification, densification	NaOH/Na <sub>2</sub> SO <sub>3</sub>	100°C, 5 MPa, 24 h	584.3	5.3	(38)
Poplar wood				431.5	3.0	
Cedar wood				550.1	3.3	
Pine wood				536.9	3.0	
Spruce wood	Delignification, densification	$H_2O_2/HAc$	Universal testing machine	270	2.2	(36)
Poplar wood	Delignification, oxidation, densification	NaClO <sub>2</sub> /HAc/NaOH	70°C, 35 MPa, 48 h	328.8	8.3	(40)
Basswood	Delignification, densification	H <sub>2</sub> O <sub>2</sub>	60°C, halve the thickness	161.3	1.3	(41)
Basswood	Delignification, control water after drying, densification	NaClO <sub>2</sub> /HAc/NaOH	Room temperature, 10 ton	352	4.1	(42)
Bamboo stem	Delignification, densification	NaOH/Na <sub>2</sub> SO <sub>3</sub>	150°C, 5 MPa, 24 h	1,008	8.5	(45)

strong white wood was a straightforward and easily scalable technique. The strong white wood exhibited excellent mechanical properties and insulation properties, surpassing most structural materials used in high-rise buildings such as steel and cast iron.

Nature creates strong and resilient substances (such as bone and pearls) without the use of complex processes and molecules. Han et al. developed biomimetic structure materials with exceptional strength and toughness from cellulose nanofibers using a simple yet universal mechanism (42). Natural wood was transformed into a strong and resilient structural material via a series of procedure involving delignification, desiccation assembly, and hydrogen bonding formed by water molecules under pressure. The effect of various drying methods on the mechanical properties of fabricated bulk materials was studied. During drying and compression, water molecules were essential to assemble cellulose nanofibers into robust and resilient structural materials. The adjacent cellulose nanofibers formed hydrogen bonds with the water molecules, joining them together. Finally, this cellulose structure material exhibited concurrently increased tensile strength (352 MPa) and toughness (4.1 MJ·m<sup>-3</sup>) that were approximately 6 and 10 times greater than those of natural wood. This environmentally friendly and pollution-free process, which can produce nanocellulose bulk materials with superior mechanical properties, is anticipated to be used in the construction and aerospace industries.

China is the nation with the largest bamboo forest area and bamboo production in the world (43). The specific strength and specific rigidity of bamboo surpass those of steel. Bamboo has good machinability. The energy consumption per unit is less than that of concrete and steel (44). Li et al. (45) devised a straightforward and efficient top-down method that included flattening, delignification, and hot pressing for transforming natural bamboo into a strong and robust densified bamboo (Figure 2b). The densified bamboo had excellent mechanical properties due to the large number of hydrogen bonds and few structural defects caused by the long and neatly arranged cellulose nanofibers. The tensile strength of densified bamboo has reached 1 GPa, outperforming a variety of wood materials, steels, and alloys, as well as the previously mentioned strong densified wood (38). And bamboo can grow up to 100 cm a day, which is approximately 1.3 times that of the fastest growing trees.

The preparation conditions and mechanical properties of cellulose materials prepared by delignification—densification method are summarized in Table 1. In this way, cellulose structural materials with excellent mechanical properties can be prepared. In addition, the raw material, delignification method, and densification method all affect the mechanical properties of the materials.

### 2.2 Pressing

To improve fiber-matrix adhesion in cellulose fiber-based structural materials, their processing method frequently employs chemical solvents and environmentally hazardous substances, thereby increasing their carbon footprint (46,47). Cellulose possesses a strong self-binding ability and an enhanced hydrogen bond network in microstructure and nanostructure. Based on these properties, Arévalo and Peijs (48) prepared binder free panels from microfibrillated flax fibers. These fully biobased materials did not require any pre-treatment, solvents, or binders, making them eco-friendly. In theory, the binder free panels could be recycled into new all-cellulose materials via a simple repulping process. After optimizing the treatment conditions, binder-free panels with superior flexural modulus and strength (approximately 17 GPa and 120 MPa) and low water absorption were produced, outperforming the majority of conventional panel board materials. This work presents a novel eco-friendly alternative to conventional building materials.

Through micro/nano-scale structural design, natural wood particles were converted into regenerated isotropic wood (RGI-wood) with excellent properties (49). Sawdust was transformed into a bulk structure material via alkaline solution treatment, oxidation, calcium ion crosslinking, and hot pressing (Figure 3a). After nanocrystallization of the surface, the specific surface area of cellulose nanofibers on the material surface increased and the hydrogen bonds were denser. The RGI-wood obtained exhibited excellent physical properties, mechanical properties, and stability, making it a promising material in the construction industry.

Yousefi et al. produced cellulose nanofiber boards (CNF-board) with a 3 mm thickness and no adhesives or additives (50). The extremely absorbent CNF gel was placed in a cold press, gradually dehydrated, and cast to form a CNF mat. CNF-mat was then dehydrated to produce CNF-board. During the vacuum drying process, the CNF-board self-densified and contracted in-plane and out-of-plane. In contrast to conventional cellulose fiber board, the CNF-board obtained after five cycles of wetting and drying showed an average high water activation size recovery rate of 96%. The CNF-board exhibited a tensile strength of 85 MPa and a flexural strength of 162 MPa, which were greater than those of cellulose fiber board and some other conventional polymers, wood-based composites, and structural steel. CNF-board is applicable to future generations of eco-friendly advanced materials due to its superior properties.

Prosvirnikov et al. produced binder-less composite compressed boards from activated cellulose material obtained by steam-blasting (51). This treatment altered the chemical composition of the high-moisture wood raw material and increased the obtained wood particles' specific surface area. The obtained pressed board materials had a tensile strength of 45 MPa. Hajihassani et al. (52) utilized a hygrothermomechanical technique to densify wood. The hygrothermal treatment enhanced the properties of dense wood, particularly its dimensional stability.

The cellulose structural materials are expected to have both high strength and good toughness. However, the inherent conflict between these two properties makes it difficult to simultaneously realize both (53). From cellulose nanofibers, Guan et al. (54) have devised a simple and stable strategy for preparing high-performance bulk structural material (Figure 3b). Simple hot pressing was used to transform the multilayered pretreated bacterial cellulose nanofiber hydrogels into high-performance CNF plate (CNFP). The specific strength of CNFP (~198 MPa/(Mg·m<sup>-3</sup>)) was four times that of steel, conventional plastic, and aluminum alloy. In addition, CNFP had a higher specific impact toughness (~67 kJ·m<sup>-2</sup>/(Mg·m<sup>-3</sup>)) and half the density (1.35 g·cm<sup>-3</sup>) of aluminum alloy. In addition, CNFP retained its strength despite being subjected to 10 rapid thermal shocks between 120°C and -196°C (Figure 3c and d). These results demonstrate the exceptional thermal dimensional stability of CNFP, which makes it a promising candidate for use as a structural material under extreme temperatures and alternating heating and cooling. Moreover, they assembled Ca<sup>2+</sup> -crosslinked cellulose nanofiber hydrogels into polymeric structural materials with excellent mechanical properties and thermal stability by means of component pressing with directional deformation (55). These biodegradable, eco-friendly structural materials are ideal alternatives to non-renewable petrochemical plastics.

#### 2.3 Composites

Due to variations in composition and structure, the properties of each substance are distinct. The addition of functional components can enable cellulose materials to obtain superior properties such as excellent mechanical property, high light transmission, and high flame retardancy, among others.

By removing lignin and filling with a specific polymer (epoxy resin), Zhu et al. (56) produced highly transparent anisotropic wood composites. To investigate the structure–process–property relationship of wood, two kinds of transparent wood were prepared according to the orientation of original channels relative to the wood

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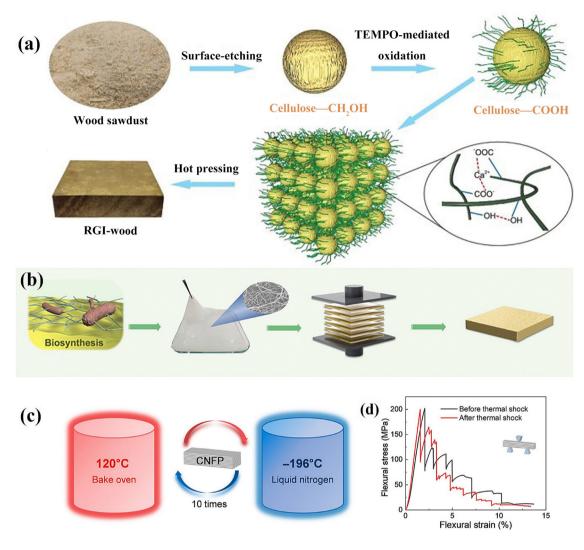


Figure 3: (a) Schematic showing the production process of regenerated isotropic wood. Reproduced with permission from the study by Guan et al. (49). Copyright © 2020; Oxford University Press. (b) Schematic showing the production process of cellulose nanofiber plate (CNFP). (c) Schematic of ten rapid cycles of CNFP at extreme temperatures (120°C and –196°C). (d) Flexural stress–strain curves of CNFP before and after ten rapid cycles. Reproduced with permission from the study by Guan et al. (54). Copyright © 2020; American Association for the Advancement of Science.

plane (Figure 4a and b). In the transparent wood obtained, the natural cellulose structure was greatly retained, the colored lignin was eliminated, and the pores were filled with epoxy resin (Figure 4c), resulting in a high transmission of light and an excellent mechanical property. In addition, their performance varied based on differences in microstructure. L-wood had stronger optical anisotropy and greater mechanical strength than R-wood (about two times) (Figure 4d). Ding et al. (57) obtained transparent wood by a similar method and coated the wood with perfluorodecyltriethoxysilane (FAS) to obtain hydrophobic transparent wood. Wang et al. (58) manufactured transparent compressed wood (TCW) by delignifying, densifying,

and impregnating it with UV-curable resin. The obtained TCW possessed excellent mechanical properties, high surface hardness, excellent thermal stability, and high light transmittance, making it suitable for use in photovoltaics, construction, and aviation.

Xing et al. (59) prepared epoxy resin/bamboo fiber composite structural materials using a straightforward two-step process that included lignin removal and epoxy resin infiltration. The adhesion between epoxy resin and bamboo fiber was strong, making the composite to have a high tensile strength (162.12 MPa), high toughness (67.14 kJ·m<sup>-2</sup>), and an excellent stability in humid environments. In a separate study, hydrogen peroxide/acetic acid vapor was used to

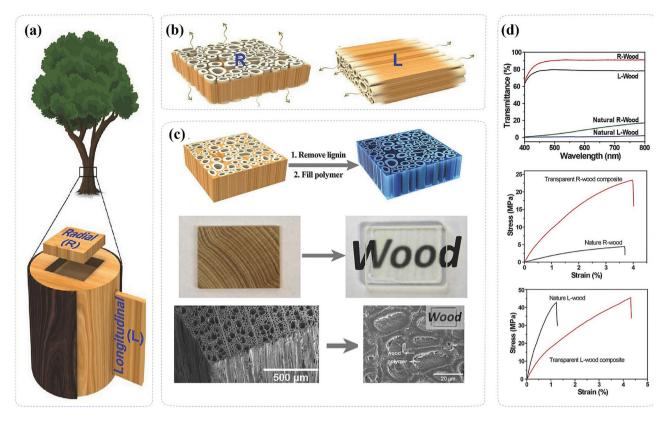


Figure 4: (a) Schematic illustration of two types of wood blocks. (b) The lumina orientation is different in the two types of wood blocks. (c) Schematic drawings, photographs, and SEM images of natural wood and transparent wood. (d) Total transmittance and stress-strain curves for nature wood and transparent wood. Reproduced with permission from the study by Zhu et al. (56). Copyright © 2016; Wiley-VCH.

eliminate lignin from wood (60). After subsequent epoxy resin casting and hot pressing, high-performance wood-based composites (CDW/Ep) were produced. It was anticipated that the CDW/Ep's superior mechanical properties (tensile modulus and strength of 10.0 GPa and 316.7 MPa, respectively) and high dimensional stability would promote the development of eco-friendly structural materials.

Tian et al. prepared wood-high density polyethylene composites (61) and rubber-filled wood fiber composites (62) using recycled tire rubber as filler. The utilization of recycled tire rubber as raw material is beneficial to the reuse of resources. The prepared composites can be reasonably utilized under certain conditions and provide strong support for sustainable development.

Inspired by nature, Guan et al. (63) devised a simple and effective method for producing all-natural structural materials. Ca<sup>2+</sup> was used to crosslink CNFs with TiO<sup>2</sup>– mica to produce hydrogel. Through directional deformation assembly, the obtained hydrogel was converted into a sustainable structural material resembling nacre (Figure 5a). With a well strength and toughness of 281 MPa and 11.5 MPa·m<sup>1/2</sup>, respectively, and excellent thermal stability, this structural material is expected to replace plastics as a

renewable structural material. Through delignification, oxidation, epoxy coating, and cross-lamination, Sun et al. (64) created eco-friendly structural materials with an ultrahigh cellulose content of 96.1 wt% (Figure 5b–d), also inspired by nacre. The material obtained had a high isotropic strength and abrasion resistance of 137 MPa and 1.79 MJ·m<sup>-3</sup>, respectively (Figure 5e).

Many other cellulose structural materials have been prepared in composite form to meet functional requirements. Han et al. (65) prepared transparent fibers from plant fibers by delignification and epoxy impregnation. The obtained transparent fiber had high tensile strength, high Young's modulus, and good light conductivity, which is expected to be used in optical fiber. Chen et al. (66) prepared a composite hydrogel actuator by polymerizing poly(N-isopropylacrylamide) into anisotropic wood. With both good mechanical properties and fast actuation speeds, this brake is a good candidate for smart applications. Han et al. (67) prepared structural layers of wood-based winding pipes with nonwoven reinforced poplar veneer strips, phenolic, and epoxy resins. The effect of water content on tensile strength was evaluated. The prepared structural layers have excellent water resistance and provide data reference and 8 — Wenze Yan et al. DE GRUYTER

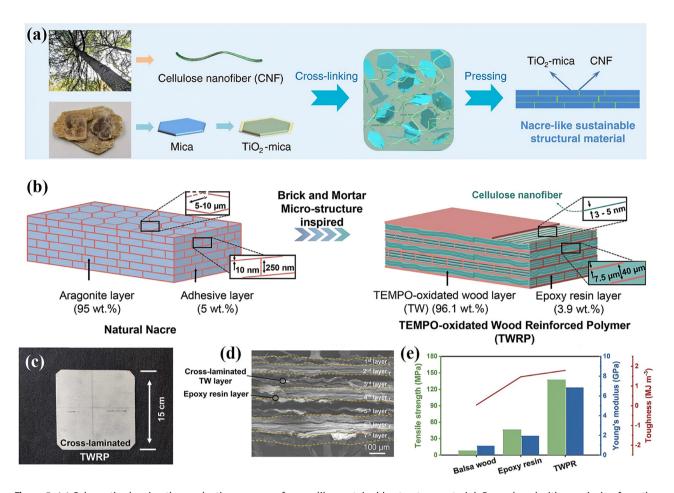


Figure 5: (a) Schematic showing the production process of nacre-like sustainable structure material. Reproduced with permission from the study by Guan et al. (63). Copyright © 2020; Springer Nature. (b) Schematic showing the structure of nature nacre and TEMPO-oxidized wood reinforced polymer (TWRP). (c) Photograph and (d) SEM image of TWRP. (e) Excellent mechanical strength of TWRP. Reproduced with permission from the study by Sun et al. (64). Copyright © 2022; Elsevier.

theoretical basis for optimizing the production process of such materials.

The preparation and properties of the cellulose composite structural materials are summarized in Table 2. The addition of new components brings functionalities such as light transmission and hydrophobicity to the composites. Densification has a key influence on the strength of the composites.

### 3 Thermal insulation cellulose structural materials

Residential and commercial buildings are major consumers of energy. Considerable energy is lost through walls, floors, and windows during heating and cooling. Therefore, thermal insulation performance is an essential component of all structural building materials (68).

Leng and Pan (69) fabricated a spray-dried cellulose nanofibrils modified polyurethane foam (PUF). The addition of CNFs increased the precursor's viscosity and inhibited cell coalescence. During the foaming process, due to the inherent high strength of CNFs, the force of resistance against cell expansion increased, the size of newly formed cells decreased, and the likelihood of producing defective cells decreased. This increased the resistance to heat transfer, resulting in improved thermal insulation with a decrease in thermal conductivity of about 40%. Therefore, it has significant potential as a structural insulating material in building.

Li et al. (70) created cellulose nanofiber-based composite foam for the production of structural insulated panels (SIPs). Epoxy-resin-coated CNF walls and epoxy-resin-bonded CNF microstructure joints increased the mechanical strength, according to the findings. However, the presence of excess epoxy residue increased the density of the foam and resulted in modest improvements in

Table 2: Preparation and properties of cellulose composite structural materials

Raw materials	Preparation methods	Strength (MPa)	Advantages	Ref.
Basswood/epoxy resin	Delignification, infiltration	45.4	Transparent	(56)
Balsa wood/epoxy resin/FAS	Delignification, infiltration, coating	17.7	Transparent, hydrophobicity	(57)
Balsa wood/UV resin	Delignification, infiltration, densification	113.7	Transparent	(58)
Bamboo slices/epoxy resin	Delignification, infiltration	162.1	Ductility	(59)
Poplar wood/epoxy resin	Delignification, infiltration, densification	316.7	Dimensional stability	(60)
CNF/TiO <sub>2</sub> -mica	Crosslinking gelation/densification	281	Heat stability	(63)
Balsa wood/epoxy resin	Delignification, TEMPO-oxidation, densification, coating	137	Heat stability	(64)

compressive modulus and strength. The epoxy resin/CNFs composite foam had low density, excellent mechanical properties, and thermal insulation property, and hence could be utilized as an insulated structural core for SIPs.

Inspired by nature, Wang et al. (71) prepared a structural aerogel with excellent thermal insulation and mechanical properties using cellulose nanofibers and zirconium phosphate/graphene as raw materials. The biomimetic structural aerogel produced by space restriction strategy and unidirectional freeze-casing technique possessed low thermal conductivity (18 mW·m<sup>-1</sup>·K<sup>-1</sup>), high specific Young's modulus (104 kN·m·kg<sup>-1</sup>), and a high density. Using natural wood and silica as raw materials, Yan et al. (72) created a composite aerogel with excellent insulation properties by freeze-drying technology and the sol-gel technique. Composite aerogels with excellent thermal insulation properties (0.032 W·m<sup>-1</sup>·K<sup>-1</sup>) and freeze resistance in liquid nitrogen (-196°C) were produced. Similarly, Han et al. (73) prepared an anisotropic, lightweight, superhydrophobic, and thermally insulating rattan aerogel by a three-step process of delignification, freeze drying and silanization. The investigated aerogel may be a promising material for high-performance structural heat insulation.

Glass, an essential building material, has a high thermal conductivity, resulting in significant energy losses (74). As an energy-saving structural material, transparent wood is a suitable alternative to traditional glass because of its superior mechanical properties, light transmittance, and thermal insulation. Wang et al. (75) transformed natural wood into large-size transparent wood with relatively high light transmittance (68%) and haze (82%) by delignification and polymer (PMMA) permeation. Since the large-size transparent wood with any thickness of any size can be facilely made and wood processing wastes can be used as raw material, this method for producing transparent fiber wood has a high preparation efficiency and high resource utilization.

Jia et al. (76) produced clear wood materials with superior thermal insulation and optical properties by delignification and infiltration of epoxy resin. The delignification process created a porous microstructure, allowing the epoxy resins to enter easily, resulting in the dense structure of the clear wood. The removal of lignin greatly reduced the scattering behavior of light in the wood, which, in conjunction with the extremely dense structure, results in superior optical properties. In addition, the thermal conductivity of transparent wood, about 0.35 W·m<sup>-1</sup>·K<sup>-1</sup>, is only one third of that of regular glass, which greatly reduces the heat loss. Mi et al. (77) optimized the optical, thermal, and mechanical properties of the transparent wood by extending the bleaching process with sodium hypochlorite (NaClO<sub>2</sub>) and immersing the porous cellulose scaffold in polyvinyl alcohol instead of epoxy. Zhang et al. (78) designed two types of wood-plastic composite structural wood walls and investigated their thermal insulation properties. Both composites had good thermal insulation properties and can be applied to insulated structures in cold regions.

### 4 Flame-retardant cellulose structural materials

As early as thousands of years ago, wood has been the most primitive cellulose structural material used by our ancestors to make houses, bridges, containers, and other structures (79–82). Compared to the steel and concrete manufacturing processes, the use of wood not only reduces environmental pollution due to its green characteristics, but also contributes positively to the carbon footprint (83,84). Due to its high fire risk, wood presents challenges to the modern construction industry despite its many positive qualities.

Gan et al. (85) utilized delignification and densification to significantly enhance the fire-resistance of wood materials. The densified wood obtained had a highly dense laminate structure and low air permeability. In case of a fire, the surface of the densified wood can form a dense carbon layer that insulates oxygen and heat to prevent the fire from spreading (Figure 6a). Densified wood demonstrated a very high compressive strength of 101 MPa, but its ignition time ( $t_{\rm ig}$ ) was 2.08 times that of natural wood (Figure 6b and c). The maximum rate of heat release was decreased by 34.6%, and the compressive strength of the material after 90 s of flame exposure was increased by more than 82 times. This mechanically strong and fire-retardant cellulose structure material has great application potential in the field of safe and environmental protection construction.

Incorporating functionalized components into cellulose materials is an effective method for enhancing their flameretardant properties. Gan et al. (86) further improved the flame retardant of densified wood by introducing boron nitride coating on the surface and obtained the BN-densified wood with anisotropic thermal conductivity (Figure 6d). The incoming heat of BN-densified wood will pass along the surface and enter the wood interior with difficulty. Meanwhile, the flame-retardant coating effectively reduces the transport of oxygen and volatiles, preventing the material from burning violently. In comparison to uncoated densified wood, the  $t_{ig}$  of BN-densified wood was increased by two times, and the maximum heat release rate (HRR) was decreased by 25%, indicating a great improvement in flame retardancy (Figure 6e). In addition, it exhibited excellent mechanical properties due to the neat and dense arrangement of internal cellulose fibers. In conclusion, the BN-densified wood is a safe, renewable, and flame-retardant cellulose structure material with great application prospects.

Chen et al. (87) fabricated a compact flame-resistant wood by permeating delignified wood with bentonite nanosheets and densification. Delignification increased the porosity of wood cell wall, allowing bentonite nanosheets to permeate into the microchannels of delignification wood to produce a surface with high compatibility and flame retardancy. Additionally, the densely packed laminate structure produced by densification, hydrogen bonds, and Al–O–C bonds gave the wood laminates an exceptional mechanical tensile strength. Similarly, Wang et al. (88) manufactured highly flame-resistant wood via delignification, phytic acid impregnation, and subsequent hot pressing.

By delignification and introducing a PAM-based hydrophobic association hydrogel (HAH), Mai et al. (89) fabricated a wood composite with excellent mechanical properties and fire resistance. The composite exhibited excellent flame retardancy and mildew resistance due to the presence of HAH.

Guo et al. (90) used an efficient and stable method to transform bamboo into a flame-retardant boric acid (BA)-bamboo/epoxy composite structural material. Unlike Xing et al. (59), they treated the delignified bamboo surface with BA prior to epoxy resin infiltration, which promoted the carbonization of bamboo/epoxy composite and significantly improved the flame retardancy. BA-bamboo/epoxy composites had a 26.5% higher limiting oxygen index and a 63% lower peak HRR than delignified bamboo/epoxy composites.

Using polydopamine as binder, Zhang et al. (91) prepared CNCs/MXene coating with flame retardant and fire warning performance from Ti<sub>3</sub>C<sub>2</sub>TX MXene and cellulose nanocrystals (CNCs), and further prepared smart wood by layer-by-layer assembly technique. In the event of a fire, the CNCs/Mxene coating could insulate oxygen and smoke, resulting in smart wood with excellent flame and smoke suppression. In addition, when a fire occurred, the resistance of the CNCs/MXene coating dropped dramatically due to oxidation, which allowed the smart wood to accurately sound the fire alarm.

Chen et al. (92) successfully impregnated PI into delignified wood and prepared a flame-retardant transparent wood/polyimide (FRTW/PI) composite. After removing the lignin's color, the polyimide impregnation increased the optical transparency and decreased light scattering (Figure 6f). The obtained FRTW/PI showed an excellent mechanical strength of 169 MPa (Figure 6g). In addition, FRTW/PI composites exhibited excellent self-extinguishing properties within 2 s, demonstrating effective flame-retardant properties (Figure 6h). Flame-retardant wood composite material could be ideal for colored glass in future buildings.

Thomas et al. (93) studied the potential of biomaterials including starch, chitosan, fish gelatin, and rice bran as wood fire retardant coatings, which can help to obtain cellulose structural materials with good flame-retardant properties. The substrates and the required amount of the additive were added to warm water ( $\approx 60^{\circ}$ C) and subsequently dispersed to create a fireproof coating. Their research demonstrated that the formula containing fish gelatin had the highest fire resistance and could be used to coat cellulose structural material with a fire-resistant coating.

Cellulose nanofiber aerogel is a promising thermal insulation material (94), as it possesses not only high mechanical and thermal insulation properties but also high flammability (95). Wang et al. (96) prepared CNF aerogels with superior thermal stability using freeze-drying and post-crosslinking techniques. As co-additives, *N*-methyloldimethylphosphonopropionamide and 1,2,3,4-butanetetracarboxylic acid could impart excellent elasticity, flame retardancy, and flexibility to CNF aerogels. Huang et al. (97) created composite

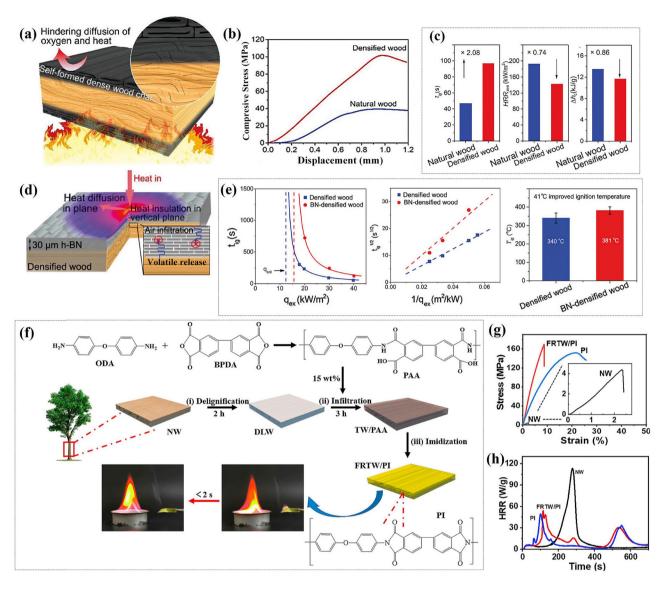


Figure 6: (a) Schematic illustration of flame-retardant principle of densified wood. (b) Mechanical properties and (c) flame-retardant performance of densified wood. Reproduced with permission from the study by Gan et al. (85). Copyright © 2019; Wiley-VCH. (d) Schematic illustration of heat conduction in BN-densified wood. (e) Flame-retardant performance of BN-densified wood. Reproduced with permission from the study by Gan et al. (86). Copyright © 2020; Wiley-VCH. (f) Schematic illustration of the preparation of the FRTW/PI composite. (g) Mechanical properties and (h) flame-retardant performance of FRTW/PI composite. Reproduced with permission from the study by Chen et al. (92). Copyright © 2020; Elsevier.

aerogels with excellent flame retardancy by esterifying and freeze-drying CNF aerogels with a novel modifier (DOPO-IA).

## 5 Hydrophobic cellulose structural materials

Frequently, the application environment of structural materials is intricate and harsh. It is frequently employed

in moist, wet, and even corrosive environments. Consequently, it is vital to enhance the waterproofing and corrosion resistance of structural materials.

Aldalbahi et al. (98) developed a photoluminescent composite by combining rare earth-doped nanoparticles of strontium aluminate with methyl methacrylate. The obtained photoluminescent composite was combined with delignification wood to produce multifunctional, UV-shielding, superhydrophobic, and flame-retardant wood. Han et al. (99) created lignocellulosic carbon material with strong hydrophobicity and heat stability by carbonizing and freeze-drying delignified

wood. The carbonization process increased the porosity and electrical conductivity of the material, allowing this carbon material to be used in a variety of applications. Han et al. (100) used the vacuum dipping technique to create superhydrophobic wood with high mechanical resistance from natural wood, silica nanoparticles, and vinyltriethoxysilane (VTES). The random distribution of  $SiO_2$  nanospheres coated with oligomers of different sizes formed a superhydrophobic surface and improved the wear resistance of the material. Sun et al. (101) created a novel all-wood material with high mechanical strength and excellent water resistance by delignification, TEMPO oxidation, lignin deposition, and hot pressing (Figure 7a). This simple and effective modification method was anticipated to solve the two major issues of superhydrophobic

wood in practical applications viz. poor durability and complex production process (102).

Through the *in situ* growth of nano-SiO<sub>2</sub> and hotpressing, Li et al. (103) created a superhydrophobic, high-strength, and lightweight wood-based structural material (SH-wood) inspired by nature (Figure 7b). The microscale roughness produced by the sandpaper was combined with the nano-SiO<sub>2</sub> particles to provide the SH-wood a micro/nano structure (Figure 7c and d). This micro/nano structure conferred superhydrophobicity to SH-wood with static and dynamic contact angles of 159.4° and 3°, respectively. SH-superhydrophobic wood's properties allowed it to maintain exceptional stability and durability in harsh environments. Furthermore, SH-wood had a high tensile

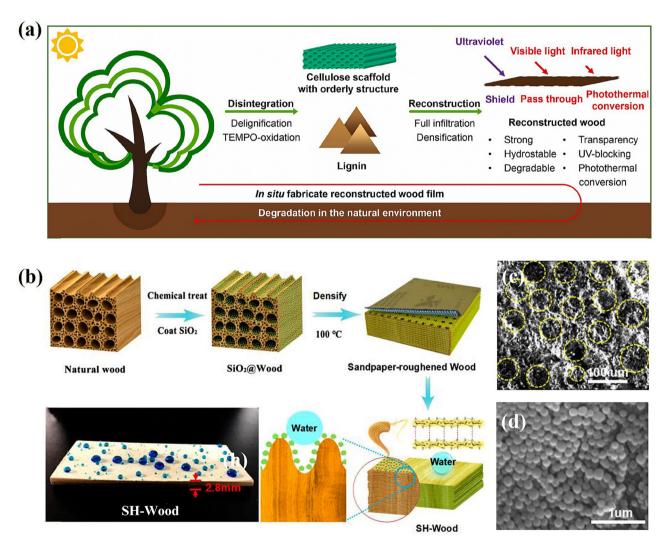


Figure 7: (a) Schematic illustration of the fabrication of reconstructed wood and its characteristics. Reproduced with permission from the study by Sun et al. (101). Copyright © 2022; Elsevier. (b) Photograph and the preparation process of SH-wood. (c and d) SEM images of SH-wood at different magnifications, in which (c) shows the micron roughness and (d) shows the nano roughness of SH-wood. Reproduced with permission from the study by Li et al. (103). Copyright © 2020; Wiley-VCH.

**Table 3:** Preparation and properties of wood-derived hydrophobic cellulose structural materials

Raw materials	Preparation methods	Water contact angles (°)	Advantages	Ref.
Basswood/MMA/APP/LSA	Delignification, infiltration	163.8	Flame-retardant/ultraviolet protective/luminescent transparent	(86)
Poplar	Delignification, freeze drying, pyrolysis	135	Flame retardant/electrical conduction	(66)
Populus/SiO <sub>2</sub> /VTES	Hydrothermal vacuum dipping	151	Mechanically stable/durable	(100)
Basswood/SiO <sub>2</sub>	Delignification, infiltration, hot press	159.4	Acid resistance/alkaline resistance	(103)
Fir wood/Ni nanoparticles	Infiltration, calcination	152.1	Electromagnetic interference shielding	(104)
Rice straw	Freeze drying, covalent crosslinking	151	Oil and organic solvent adsorption	(105)
Bacterial cellulose/SiO <sub>2</sub>	Freeze drying, infiltration	142	Oil adsorption/robustness	(106)

strength, approximately seven times stronger than untreated natural wood. This structural material with high strength, superhydrophobicity, and high environmental resistance produced by a low-cost, scalable synthesis method had the potential to replace steel in construction, automobiles, and aircraft.

The preparation and properties of hydrophobic cellulose structural materials are summarized in Table 3. Various materials and methods can be employed to give the materials superhydrophobicity. Furthermore, the incorporation of new components often enhances the material's properties, thus expanding its potential applications.

## 6 Cellulose structural material with electrical properties

In recent years, with the widespread use of electronic devices, materials with excellent electrical properties such as good stability, reliable electrical conductivity, and high capacitance have received increasing attention. Due to the unique anisotropic honeycomb three-dimensional (3D) pore structure of wood, it can be used as a framework to prepare a variety of cellulose structural materials with excellent electrical properties.

Wang et al. (107) converted natural basswood into a thick carbon electrode that can be used in supercapacitors by delignification and carbonization. The obtained thick carbon electrodes had a high specific surface area, which contributed to excellent electrochemical performance (specific/area capacitance ~3,040 mF·cm<sup>-2</sup>/118 F·g<sup>-1</sup> at 1 mA·cm<sup>-2</sup>, cycle stability ~82.2% after 10,000 cycles at 50 mA·cm<sup>-2</sup>, and capacitive contribution of 76.64% at 5 mV·s<sup>-1</sup>). Using a similar method, another woodderived thick carbon electrode was prepared (108). Further, they prepared thicker carbon electrodes with better performance by adding enzyme catalysis (109) or phytic acid treatment (110) step between delignification and carbonization process.

Using carbonized wood as a framework, Yang et al. (111) prepared electrode materials for supercapacitors by surface immobilization of layered porous  $\rm NiCo_2O_4$  nanosheets. The 3D porous structure derived from wood can provide favorable conditions for charge transfer. The obtained electrode material had excellent electrochemical properties: it exhibited a specific capacitance of 1,730  $\rm F\cdot g^{-1}$  at 1  $\rm A\cdot g^{-1}$  in 1 mol·L<sup>-1</sup> of KOH, and the capacitance was maintained at 80% after cycling. Meanwhile, the supercapacitor assembled with the obtained electrode material had a high energy density. Similarly, Wang et al. (112) prepared porous monolithic

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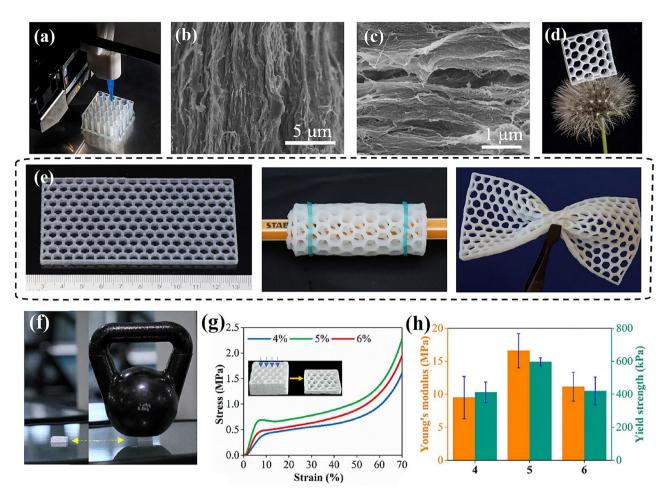


Figure 8: (a) Photograph of 3D printing of all-cellulose honeycomb structure. (b) Longitudinal and (c) transverse SEM images of freeze-dried cellulose honeycomb. (d) The freeze-dried cellulose honeycomb standing on top of dandelion, showing that it is lightweight. (e) The freeze-dried honeycomb structure can be restored after crimping and twisting. (f) Photograph of a piece of 3D printed cellulose honeycomb structure (5 wt%) supporting a 6.8 kg kettlebell. (g) Compressive stress—strain curves, (h) Young's modulus and yield strength of free-dried 3D printed honeycomb structure, showing the outstanding mechanical performance. Reproduced with permission from the study by Jiang et al. (122). Copyright © 2021; Elsevier.

electrodes with excellent performance by sequentially decorating redox-active silver nanoparticles and NiCo<sub>2</sub>O<sub>4</sub> nanosheets on carbonized wood. These electrode materials have great prospects for application in energy storage devices.

The widespread use of electronic devices has made the electromagnetic radiation a very serious problem. Lightweight, renewable, and economical electromagnetic shielding materials are getting increasing attention. Ma et al. (113) prepared 3D porous carbon skeleton/magnetic composites (Co/C@WC) by zeolitic imidazolate framework-67 (ZIF-67) growing *in situ* on wood and carbonizing. The obtained composites had excellent mechanical properties, electrical conductivity (3,247 S·m<sup>-1</sup>), and electromagnetic shielding ability (about 43.2 dB at 8.2–12.4 GHz). In addition, they prepared ultrathin wood films with excellent electromagnetic shielding properties by compression,

carbonization and *in situ* growth of ZIF-8 (114). These studies contribute to the wide application of renewable materials in electromagnetic shielding field.

### 7 Other cellulose structural materials

### 7.1 Radiation cooling cellulose structural materials

A significant portion of the total energy consumed annually is utilized for cooling buildings. Passive radiative cooling provides a zero-energy method for enhancing the energy efficiency of buildings, which has attracted considerable interest among researchers. Li et al. (115) created a structural material with sub-ambient cooling effects during the day by delignifying it completely and then mechanically pressing it. In the infrared spectrum, the cooling wood emitted more energy than it received from the sun. By simulating the potential impact of cooling wood, the researchers determined that it conserved 20–60% of energy, which was most evident in hot environments. In addition, cooling wood possessed exceptional mechanical properties, which allowed it to be used in high-rise buildings. This mass-producible cooling-wood is an ideal alternative to traditional building materials and is expected to contribute to global environmental pollution control and carbon neutrality.

Li et al. (116) assembled delignified cellulose fibers and  ${\rm SiO_2}$  microspheres into 3D blocks, which were then hot-pressed to form a lignocellulosic bulk. The cooling lignocellulosic bulk obtained possessed excellent mechanical properties, flame retardant properties, and outdoor antibacterial properties, and was capable of achieving 24 h cooling with a temperature reduction of 6°C during the day and 8°C during the night. This high-performance, expandable, cooling wood structure material has excellent application potential in the field of sustainable building.

### 7.2 Cellulose-based sound absorbing structure material

By silica-sol reinforcement, Dong and Wang (117) created an ultra-lightweight cellulose-based foam material (ULW-CFM) with great sound insulation. The type and surface tension of the foaming agent had an important influence on the foamability and stability of the suspension in foam forming technology (118,119). During the foam formation process, the material had a light weight, a large volume, and a high porosity because of the isolation of the foam. The porous structure created a partially permeable structure that effectively reduced the reflection of sound waves. Silica-sol modification effectively improved the mechanical and thermal properties of the material. In addition, the modified ULW-CFM demonstrated excellent sound absorption with a sound absorption coefficient of 0.65 above 5,000 Hz, which was superior to the PUF currently available in the market. The ULW-CFM had the benefits of low cost, biodegradability, exceptional mechanical properties, and excellent sound absorption property, and was expected to become a new generation of structural sound absorption material.

Jia et al. (120) prepared perforated fiberboard with side hole structure by using layered processing process.

Using medium density fiberboard as the substrate, the perforated structure and perforation method were designed to achieve the purpose of broadening the sound absorption band and improving the acoustic performance of wood perforated board. This study provides a reference for the structural design of perforated sound-absorbing panels, solves the technical problems of processing complex hole patterns, and is of great significance for the industrial application of multi-band sound-absorbing wood perforated panels.

### 7.3 3D printing cellulose structure materials

3D printing is an emerging computer-aided design and manufacturing technology. Recently, 3D printing has received a great deal of attention in comparison to conventional manufacturing because it can produce complex and/or specific 3D objects (121). By utilizing 3D printing technology, Jiang et al. (122) were able to create a low-density and high-strength cellulose honeycomb structure material (Figure 8a-c). The 3D cellulose honeycomb structure was so light that it could balance on a dandelion (Figure 8d). Due to the presence of strong hydrogen bonds, 3D cellulose honeycomb structures exhibited a high degree of wet elasticity that allowed them to withstand significant deformation and return to their original shape (Figure 8e). Because of the molecular arrangement of the cellulose chains, the 3D-printed honeycomb structure material retained its stability under 15,800 times its own weight after being freeze-dried (Figure 8f). This was superior to the majority of 3D-printed structures and cellulose foams. Moreover, after being filled with cellulose nanofiber aerogel, it demonstrated excellent thermal insulation properties. Biomedical, environmental, and structural engineering applications for this 3D-printed, cellulose-based material with high wet and dry tensile strength (Figure 8g and h) are deemed to have vast potential.

### 7.4 3D-molded wood

Wood has poor formability and is difficult to machine into complex shapes compared to metals and plastics. Using cell wall engineering, Xiao et al. (123) described a processing strategy for transforming hardwood slabs into multifunctional 3D structures. After a process of lignin removal, water evaporation, and rapid water impact, the wood formed a unique, wrinkled cell wall structure, allowing it to be shaped and dried to obtain the desired shape (Figure 9a and b). In addition, the 3D-molded wood maintained the inherent anisotropic wood structure and enhanced the interaction between wood fibers, thereby enhancing the

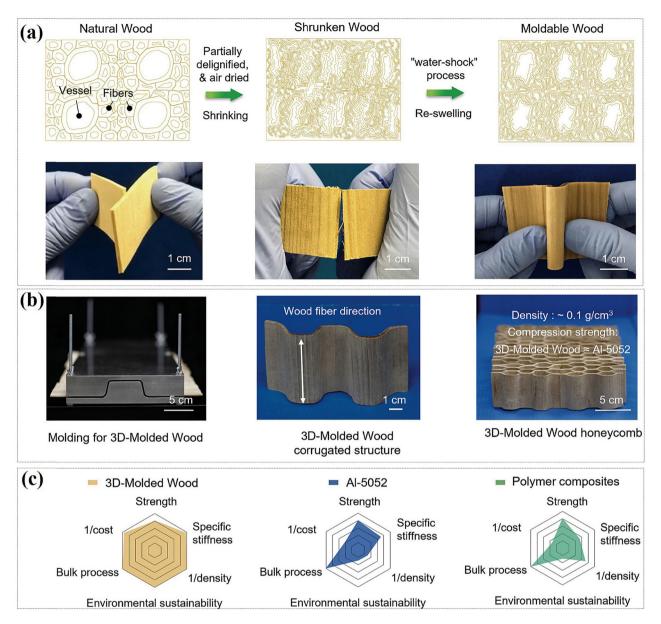


Figure 9: (a) Schematic illustration and photographs of the wood cell wall structures during the fabrication of the moldable wood, highlighting the critical role of the water-shock process, which forms wrinkled cell walls. (b) Photographs of molding for 3D-molded wood, 3D-molded wood corrugated structure, and 3D-molded wood honeycomb. (c) Comparison of 3D-molded wood, Al-5052, and polymer composites by radar map. Reproduced with permission from the study by Xiao et al. (123). Copyright © 2021; American Association for the Advancement of Science.

mechanical strength. This cell wall engineering process conferred structural versatility to wood that was previously restricted to plastics and metals (Figure 9c), giving it the potential to be used in structural engineering.

### 8 Conclusion and prospective

In response to the growing demand for structural materials in contemporary society, cellulose structural materials are

evolving toward functionalization and intelligence. In this work, we discuss the preparation and properties of novel cellulose structural materials with a variety of properties, including high strength, fire resistance, thermal insulation, and hydrophobicity. Incorporating modern science and technology, the new cellulose structural materials retain the inherent environmental friendliness, lightness, and reproducibility of cellulose materials, while being endowed with superior properties such as flame retardant, thermal insulation, hydrophobicity, etc. These new cellulose structural materials are rapidly evolving as possible

replacements for structural materials of the previous generation, such as metals, ceramics, and plastics. However, it should be noted that the true stability and sustainability of cellulose structural materials, from the mass production of raw materials to commercial applications, remain a challenge that requires further research. The feasibility of cellulose structural materials in practical application environments needs to be verified. Moreover, most of the research on cellulose structural materials has been focused on a few specific species like natural wood. The possibility of preparing structural materials from more sources of cellulose needs to be explored. The solution to these problems requires the joint efforts and cooperation between the research community and commercial organizations. We hope that the research and application of cellulose structural materials can contribute to sustainable development and the achievement of carbon neutrality.

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