

Synergistic interaction of non-covalent and coordination bond to fabricate recyclable bamboo-based material

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GRAPHICAL ABSTRACT



PUBLIC SUMMARY

- A non-adhesive bamboo-based material created using supramolecular design.
- The fabrication process incorporates non-covalent bonds and coordination bonds for structural integrity.
- The technique offers a sustainable solution to the recyclability of wood and bamboo-based structural materials.

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Biomass materials, such as wood and bamboo, possess properties such as renewability, biodegradability, and carbon storage, making them significantly advantageous in the circular economy. However, the processing of these materials often involves the use of synthetic resins, which is highly detrimental to their recyclability. Here, we report a strategy for preparing non-adhesive bamboo-based material (BBM) through the synergistic interaction of non-covalent bonds and coordination bonds, which exhibits a record-high bonding strength of 10.97 MPa, exceeding those of previously reported biomass adhesive materials. At displacement deformations below 1.4 Å, hydrogen bonds and van der Waals forces play a dominant role on bonding strength; conversely ionic coordination bonds maintain the interface's stability. The BBM also exhibits quasi-isotropic mechanical properties and excellent moldability. The life-cycle assessment reveals that producing BBM with a non-adhesive method leads to substantial reduction in carbon emissions compared with plywood, and carbon fiber composites. More importantly, the BBM is recyclable and reusable, with a retention in bonding strength of 84.1% after four cycles of recycling. This non-adhesive biomass material fabrication strategy fundamentally addresses the challenge of recycling woody solid waste, promoting the circular economy and global sustainable development.

INTRODUCTION

Global resource and environmental issues are becoming increasingly prominent, making the robust development of a circular economy an essential solution. Recycling materials can reduce dependence on natural resources, subsequently decreasing environmental pollution and solid waste generation.¹ The global annual production of wood-based materials is approximately 4.6 billion tons, with around 60% utilized for energy production and the remaining portion is converted into various woody solid waste.² This waste has brought problems of environmental pollution and resource waste, and thus improving the recycling rate of woody solid waste is key to advancing the circular economy. A primary limitation in the recycling of woody solid waste is the presence of adhesives in wooden products. Strong physical and chemical interactions exist between wood components and adhesives, complicating the recycling process and driving up costs. Thus, there is an urgent need for the development of non-adhesive processing technologies for wooden products.

Bamboo is a significant woody resource with a global forest area estimated at approximately 43.5 million hectares.³ Compared to timber, bamboo offers considerable advantages in a circular economy due to its faster growth rate, reaching maturity in just 3 to 5 years, and its stronger carbon sequestration capacity, which is nearly twice that of trees over the same land area.^{4,5} The natural structure and chemical composition of bamboo lend themselves well to non-adhesive processing, and several studies have explored this area. Bamboo's parenchyma cells, characterized by high cavity to wall ratios, are more easily deformed during hot pressing, which enables them to connect adjacent bamboo elements and form a dense interlocking structure.^{6,7} Additionally, bamboo contains natural adhesives, such as lignin, which can interact with aligned cellulose fibers through hydrogen bonding and physical entanglement under conditions of elevated temperature and humidity,

thereby achieving the self-bonding bamboo.⁸ Notably, the surface of bamboo contains a rich variety of oxygen-containing functional groups such as hydroxyl and carboxyl groups, which can be crosslinked through non-covalent bonds, such as hydrogen bonds and van der Waals forces.^{5,6} These non-covalent bonds exhibit low bond energy and dynamic reversibility, thereby facilitating the recycling and reuse of materials. However, the aforementioned non-adhesive strategies primarily enable adhesion between fiber units or parallel assembly of bamboo veneers, but they fall short in facilitating adhesion across more complex interfaces and cannot match the performance of bamboo-based composites produced with adhesives.^{9,10} Therefore, it is necessary to develop non-adhesive bonding methods that effectively address the requirements of complex interfaces in bamboo processing.

Here, we report a green bonding strategy that leverages the synergistic effects of non-covalent and coordination bonds, demonstrating notable advantages in recyclability compared to fiber composites and plywood (Figure 1A). This method involves a series of processes, including chemical pretreatment, ion cross-linking, assembly, and hot-pressing (Figure 1B). The non-adhesive bonding of bamboo veneers is facilitated by hydrogen bonds and van der Waals forces at the interface of the pretreated bamboo, as well as coordination bonds formed between carboxyl groups and metal ions. A key benefit of this method is the elimination of synthetic resins, which simplifies the recycling and reuse of bamboo-based materials (BBM).

The synergistic interaction of non-covalent bonds and coordination bonds enables BBM to achieve a bonding strength of 10.97 MPa, exceeding that of some traditional bio-based composites using synthetic resins, bio-based adhesives, and inorganic adhesives (Figure 1D & Table S2).^{8,11-19} Moreover, when compared to plywood, BBM demonstrates significant advantages in recovery rate, bonding strength, hardness, bending strength, low global warming potential, and energy consumption (Figure 1C & Table S1).^{11-16, 20-28} This green bonding approach provides a pathway for the production of recyclable wooden products, and promoting the exploration of sustainable materials and advancements in the circular economy.

MATERIALS AND METHODS

Materials

Moso bamboo (*Phyllostachys pubescens*) veneers with a thickness of 0.45–0.5 mm were provided by Yiyang Yiyi Bamboo and Wood Co., Ltd (Yiyang, China). Potassium hydroxide (KOH, 95%), sodium chlorite (NaClO₂, 80%), 2,2,6,6-tetramethylpiperidin-1-oxyl free radical (TEMPO, 98%), sodium hypochlorite (NaClO, active chlorine 13%–16%), anhydrous ethanol (CH₃CH₂OH, 99.5%), and calcium chloride dihydrate (CaCl₂·2H₂O, 99%) were all purchased from Shanghai Macklin Biochemical Co., Ltd. (Shanghai, China). Phosphate buffer (pH=6.86) was ordered from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China).

Pretreatment of natural bamboo veneers

First, natural bamboo veneers were immersed in 2 wt% KOH aqueous solution at 90°C for 2–4 hours. After dehemcellulose, samples were immersed in deionized water several times to remove the residual chemicals. Subsequently, the bamboo veneers are immersed in a phosphate buffer solution

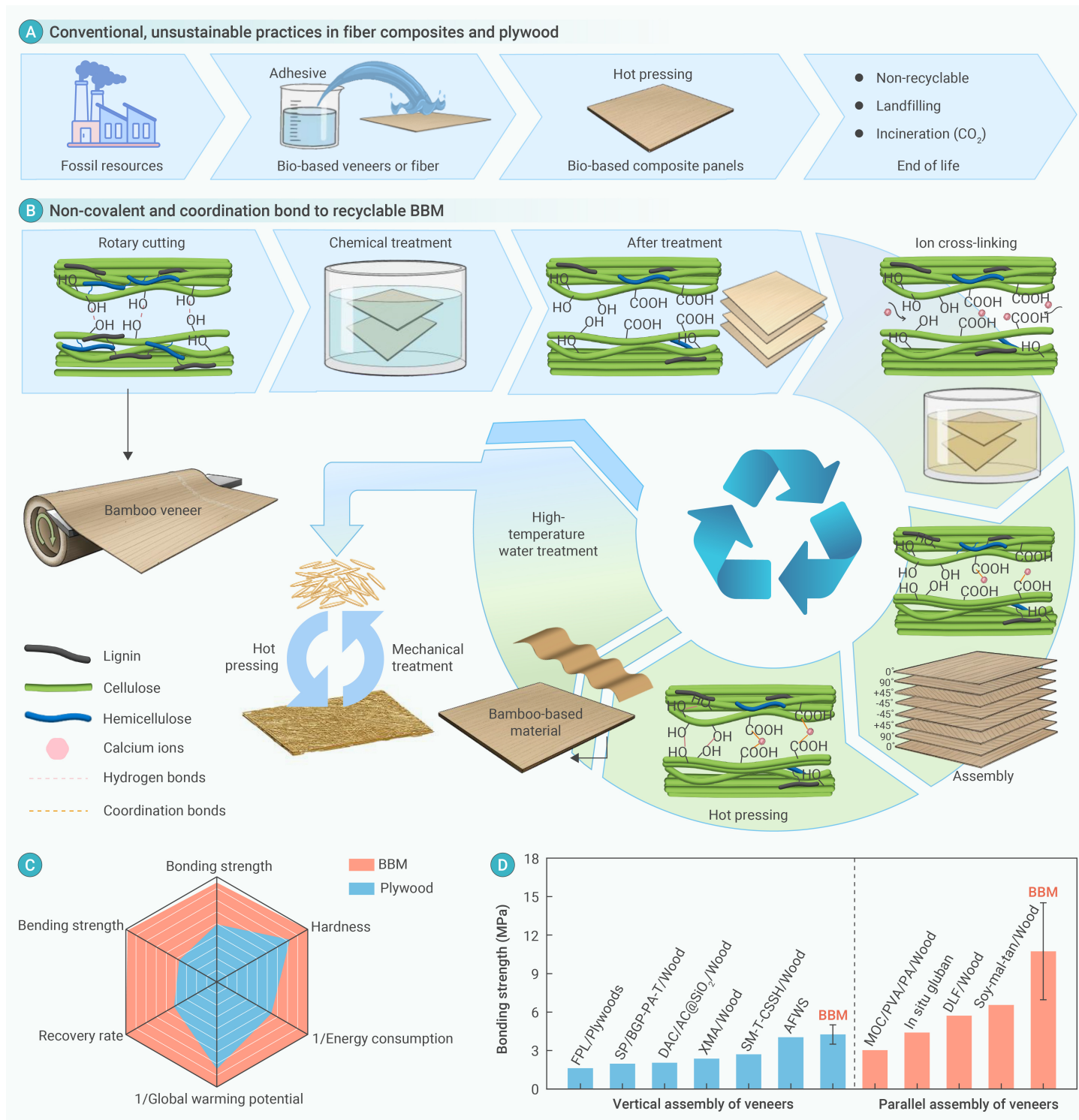


Figure 1. Synergistic interaction of non-covalent and coordination bond to fabricate recyclable bamboo-based material (A) Conventional, unsustainable practices in bio-based composites panels. (B) Recyclable schematic diagram about BBM synergistic through non-covalent and coordination bonds. (C) Radar plots comparing the performance of BBM and plywood. (D) Comparison of bonding strength between BBM and other traditional plywood.

(220 mL, pH 6.8) dissolving TEMPO (0.032 g), sodium chlorite (2.825 g) and sodium hypochlorite (0.64 mL) in a beaker. The reaction was then carried out at 60°C for 12–24 hours, followed by cleaning with ethanol and deionized water.

Preparation of ionically cross-linked bamboo veneers

The pre-treated bamboo veneers were immersed in calcium chloride dihydrate solutions of different concentrations (0.10 mmol/L, 0.15 mol/L, 2.0 mol/L, 2.5 mol/L) for 24 hours to obtain ionically cross-linked bamboo veneers.

Preparation of BBM

The ionically cross-linked bamboo veneers were assembled in 0°/90°/+45°/-45°/-45°/+45°/90°/0° (8 layers), and then pressed at temperatures of 140°C for 20 minutes under pressure of 8 MPa to produce BBM.

Bonding interface and chemical properties analysis

Scanning electron microscope (SEM) (GeminiSEM300, ZEISS, Germany) and energy-dispersive X-ray (EDX) analysis were utilized to characterize the microstructure and elemental distribution of the samples. The samples were cut into small pieces (5×5 mm) using a mini table saw, and the testing

surfaces were polished with a sliding microtome. The images of the sample morphology and elemental distribution were captured under a vacuum operation mode at 3.0 kV. Prior to SEM scanning, it is necessary to perform pre-sprayed with gold/palladium (Au/Pd) on the surface of the sample.

Fourier transform infrared spectroscopy (FTIR) was performed on a Thermo Scientific Nicolet iS20 spectrometer (Thermo-Fisher Scientific, US) to determine the functional groups of bamboo samples. The relative contents and chemical valency of elements were determined by an ESCALAB Xi⁺ type X-ray photoelectron spectroscopy (XPS) (Thermo-Fisher Scientific, US). The XPS spectra were collected at 0–1200 eV. Small-angle X-ray scattering instrument (SAXS) XEUS 2.0 (Xenocs SAS, Franch) was used to characterize fiber orientation and inter-fiber spacing. The X-ray wavelength λ was 1.54 Å and the sample-to-distance was 1188 mm for SAXS. The carboxyl content was determined by 855 type automatic potentiometric titrator (Metrohm, Switzerland), and the content of chemical composition in wood was measured using ultra performance liquid chromatography (UPLC).

Density functional theory (DFT) calculations

Periodic DFT calculations were performed using the Vienna Ab initio Simulation Package (VASP 5.4.4) with a plane-wave basis set. Electron-ion interactions were modeled using Bloch's projector augmented wave (PAW) method. A cutoff energy of 450 eV was utilized for all calculations, and a Monkhorst-Pack k-point grid of 5 × 5 × 5 was employed for optimizing the lattice slab structure of the cellulose model. Geometries were optimized by allowing all atoms to relax until energy convergence reached 1.0×10^{-5} eV/atom, with Hellman-Feynman forces converging to less than 0.02 eV/Å.

Molecular dynamics (MD) calculations

The GROMACS 2018.8 software was utilized for conducting all-atom MD simulations. The lattice parameters for the Pure I β crystalline cellulose model used in the simulations were $a = 24.853$ Å, $b = 24.620$ Å, and $c = 31.346$ Å. For the Ca-doped I β crystalline cellulose model, the lattice parameters were $a = 28.249$ Å, $b = 24.236$ Å, and $c = 31.470$ Å. The parameters for both undoped cellulose and Ca-doped cellulose were described using the universal force field (UFF). The systems were initially minimized with 5000 steps of steepest descent minimization, followed by heating to the target temperature over 50 ps and subjected to 100 ps simulations. The simulations were carried out in the isothermal-isobaric (NPT) ensemble using a Berendsen thermostat and barostat algorithm at 298 K and 1 atm, with a time-step of 0.5 fs. A 1.0 nm cut-off was employed for the direct electrostatic sum and van der Waals terms in the simulations.

Mechanical property test

Bonding strength, tensile and bending strength, and the modulus of tensile and rupture were tested using an E43.104 mechanics testing machine (MTS Co., Ltd.). Due to the thinness of the veneers, the dimensions of the specimens for the bonding strength test in this experiment were proportionally reduced according to the dimensions specified in the standards. The bonding strength of the 3-layer perpendicular layup and the 2-layer parallel layup specimens were respectively tested according to Chinese National Standards GB/T17657–2013 and GB/T33333–2016, with the corresponding bonding surface being 5 mm × 5 mm and 5 mm × 4 mm in sizes, respectively. For the measurement, a sample was clamped at both ends and stretched along the sample length direction until the two pieces separated at a constant test speed of 10 mm min⁻¹.

Tensile and bending properties tests were conducted in accordance with Chinese National Standards GB/T17657–2013. Tensile samples were approximately 100 mm by 5 mm in size at a constant test speed of 10 mm min⁻¹. Three-point bending samples were 100 mm by 20 mm with a span of 50 mm, and the tests were carried out at a constant speed of 50 mm min⁻¹. The hardness of the samples was tested using the SLX-D type Shore durometer (Shanghai Siwei Instrument manufacturing Co., LTD). Twelve points on the flat areas of the samples' surface were tested, and the results were averaged.

Life-cycle assessment (LCA)

Analysis follows ISO standard series 14040 and 14044. We evaluated the environmental impact of producing 1 m³ BBM and compared it with plywood and carbon fiber composites. The boundaries of this system include the acquisition, transportation, and production of BBM.

RESULTS

Enhanced self-bonding performance by bonding synergistic interaction

Figures 2A–B illustrate the structural evolution from natural bamboo to BBM. In natural bamboo, the parenchyma cells and fibers are remarkably well-aligned and the cellular structure remains largely intact (Figures S1A & C). After chemical treatment, the parenchyma cells and fibers became disorganized, and the debris of cell wall appeared on the surface of the specimen (Figure S1B). Subsequent to the hot-pressing process, the cell walls completely collapse, resulting in the disappearance of the porous structure of bamboo and the formation of a highly dense composite (Figure 2C). Additionally, the bonding interface between the two bamboo veneers is clearly observable (Figures 2D & S1D).

The bonding strength of BBM for vertical and parallel assemblies were 4.25 MPa and 10.97 MPa, respectively (Figure 3D). These values significantly exceed the bond strength of natural bamboo subjected to direct hot pressing. Compared with specimens prepared from oxidized bamboo veneers without added metal ions, the bonding strength for vertical and parallel assemblies of BBM increased by 2.6 times and 2.7 times, respectively (Figure 3D). In addition, BBM prepared through parallel assembly showed sufficient bonding strength to support individual weights of up to 75 kilograms without sustaining damage, demonstrating its stable shear resistance (Figure S2A). The bonding strength of BBM is significantly affected by variables such as chemical treatment time, metal ion type, and ion concentration. With two hours dedicated to hemicellulose removal followed by 18 hours of oxidation treatment, the sample exhibited optimal bonding performance (Figure S2B). Figure S3A compares the effects of Ca²⁺, Co²⁺, Fe³⁺, Cu⁺ and Zn²⁺ on the bonding strength of BBM. Considering the factors of recyclability, improved bonding strength, and low costs, calcium ions were selected for further experiments. The impact of calcium ion concentration on bonding performance revealed that an increase in calcium ion addition initially elevates bonding strength, which then declines beyond a certain concentration. The peak bonding effect was observed at a calcium ion concentration of 2 mmol/L (Figure S3B). This phenomenon can be attributed to the divalent nature of calcium ions, whereby one calcium ion can interact with two carboxyl groups. Additionally, the parameters of hot pressing, such as temperature, pressure, and duration, play a crucial role in the bonding strength. The optimal bonding performance was achieved at a pressure of 8 MPa, a temperature of 140°C, and a duration of 20 minutes (Figure S4). When the hot-pressing pressure exceeds 8 MPa, the bamboo veneer is prone to crushing, leading to a reduction in bonding strength.

The record-high bonding performance of BBM is mainly due to the synergistic effect of non-covalent interactions, specifically hydrogen bonds and van der Waals forces, in conjunction with coordination bonds. During the hot pressing process, hydroxyl crosslinking occurs along the cellulose molecular chains between bamboo veneers, facilitating the formation of hydrogen bonds. In addition, the reduction in distance between molecular chains also contributes to the generation of van der Waals forces.²⁹ These non-covalent interactions represent a significant source of bonding strength at the interface of the bamboo veneers. Furthermore, metal ions form coordination bonds with carboxyl groups present on the cellulose molecular chains, serving as another crucial factor that enhances interfacial bonding strength.

The changes in the content and functional groups of cellulose, hemicellulose, and lignin resulting from chemical treatment are fundamental to achieving effective bonding. Fourier transform infrared spectroscopy (FTIR) results show the changes in functional groups before and after chemical treatment. After dehemicellulose and TEMPO oxidation treatment, the characteristic peak at 1730 cm⁻¹ disappeared, confirming the successful removal of hemicellulose (Figures S5A–B). A new absorption peak of oxidized bamboo appeared at 1635 cm⁻¹, corresponding to the C=O stretching vibration of the carbonyl group. This observation indicates that some hydroxyl groups on the cellulose molecular chain were converted into carboxyl groups as a result of the oxidation treatment.

Carboxyl groups are crucial for the entry of metal ions and the formation of coordination bonds. The introduction of carboxyl groups enhances the negative charge on the cellulose molecular chain, leading to increased electrostatic repulsion among cellulose microfibril bundles.³⁰ This, in turn, enlarges the spacing between cellulose microfibrils, allowing for better penetration of metal ions. The changes in spacing between cellulose microfibrils was confirmed by small angle X-ray scattering (SAXS) analysis, which revealed that the distance increased from 70.4 nm to 86.5 nm following oxidized treat-

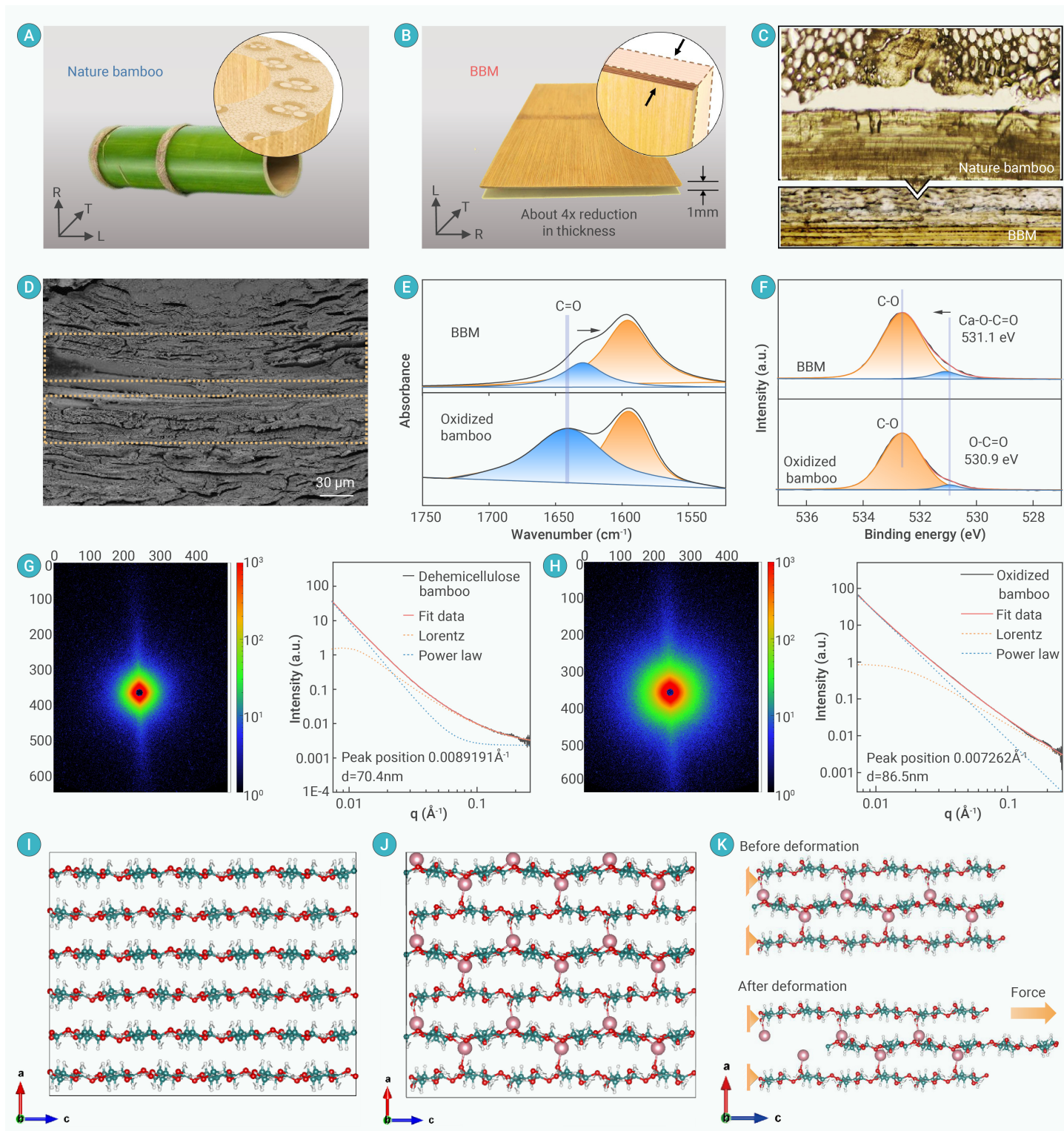


Figure 2. Enhanced self-bonding performance by bonding synergistic interaction (A) Image of natural bamboo and (B) BBM (illustration shows the cross-sectional variation). (C) Light microscope image of natural bamboo stacked vertically and BBM. (D) Scanning electron microscope (SEM) image of BBM, and the yellow line indicates the glued interface. (E) FTIR spectra of oxidized bamboo and BBM. (F) XPS partial peak fitting curve of oxygen element. Small angle X-ray scattering (SAXS) images and fiber spacing fitting curves of dehemocellulose bamboo (G) and oxidized bamboo (H). Molecular dynamics simulation models of pure cellulose (I) and cellulose doped with calcium ions (J). (K) schematic diagram of simulating cellulose chain-sliding within the cellulose unit cell.

ment (Figures 2G-H).

Figure 2F demonstrates that the binding energy of C=O increases from 530.9 eV to 531.1 eV, while the binding energy of -OH remains unchanged, confirming that calcium ions primarily crosslink with carboxyl groups. The C=O absorption peak of BBM at 1635 cm⁻¹ exhibits a significant leftward shift, confirming the formation of Ca-O-C=O coordination between calcium ions and carboxyl groups (Figures 2E & S5A).³⁰ Energy

dispersive X-ray spectroscopy (EDS) effectively reveals the uniform distribution of carbon elements, oxygen elements, and calcium ions within BBM (Figures S6A-C). Unlike the bonding observed between adhesives and wood, the Ca-O-C=O can achieve dynamic cross-linking through high-temperature water treatment. As shown in Figure S7, the binding energy of C=O decreases from 531.1 eV to 530.9 eV, suggesting that high-temperature water treatment can disrupt the Ca-O bonds, thereby enhancing the recycla-

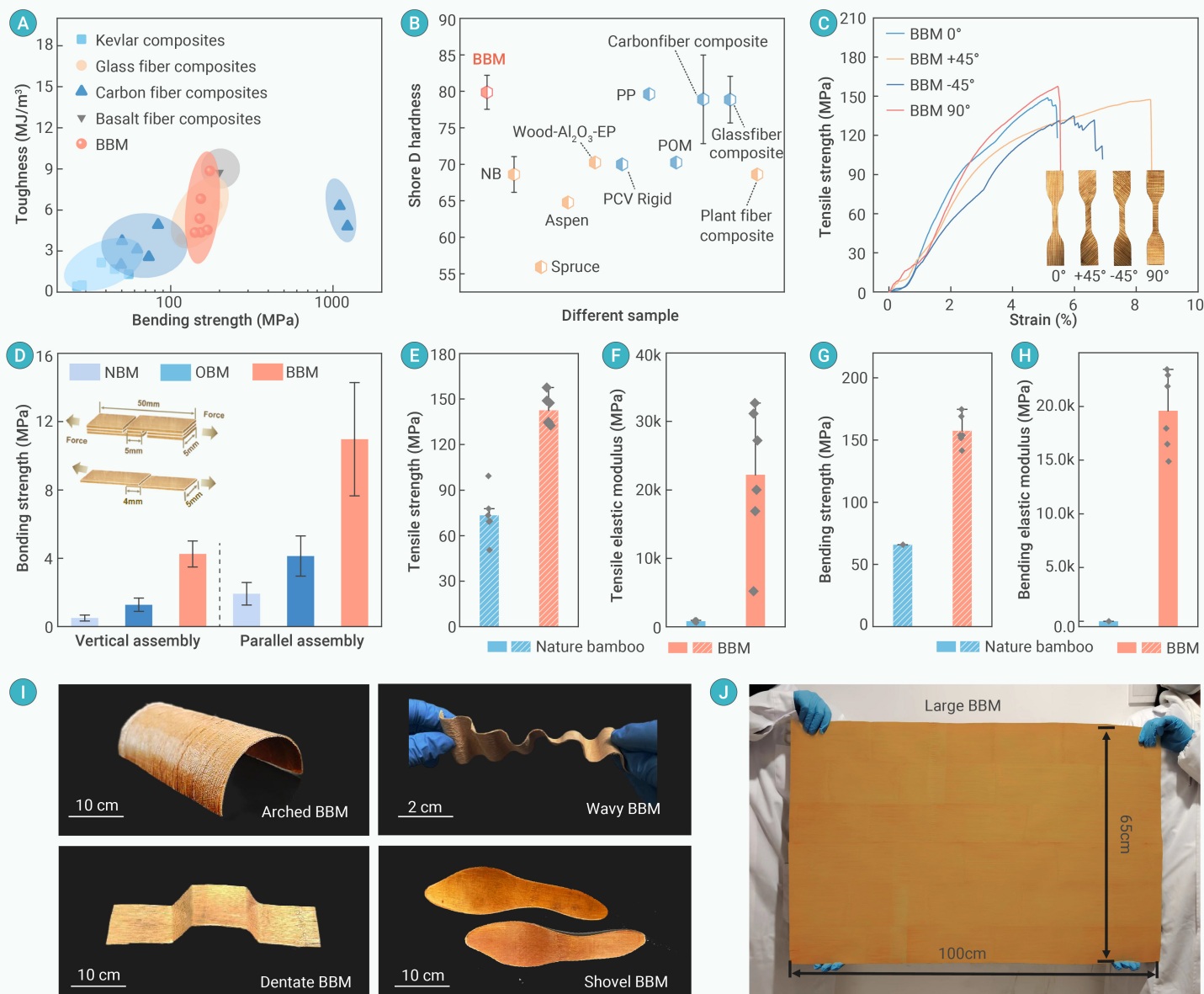


Figure 3. Mechanical properties and plastic workability of BBM (A) Toughness and bending strength of BBM. (B) Shore D hardness of BBM compared with wood, bamboo, rigid PVC and some fiber composites. (C) Typical stress-strain curves for BBM at different plane direction (0° , $+45^\circ$, -45° , 90°). (D) Bonding strength of natural bamboo veneers (NBM), oxidized bamboo veneers (OBM) and BBM. Comparison of the tensile strength (E) and tensile elastic modulus (F) of nature bamboo and BBM. Comparison of the bending strength (G) and bending elastic modulus (H) of nature bamboo and BBM. (I) Plastic workability of BBM. (J) Photograph of BBM with dimension of 1000 mm (length) $\times$ 650 mm (width) $\times$ 1 mm (thickness).

bility of BBM.

The simulation results of molecular dynamics simulations (MD) and density functional theory (DFT) further confirmed the main mechanism of BBM bonding performance enhancement, and clarified the contribution of non-covalent and coordination bonds to bonding strength. Figures 2I–J show the calculation models for pure cellulose system and calcium ion coordination, respectively. The MD results show that the absolute value of Lennard Jones potential energy decreases after doping with calcium ion (Figure S8A), indicating a weakening of van der Waals force interactions. This may be due to changes in molecular arrangement or the introduction of calcium ion, which alters the van der Waals forces between molecules. The absolute value of Coulomb potential energy also significantly decreased, indicating a weakening of charge interactions. The introduction of calcium ion may have altered the charge distribution or shielding effect in the system. The significant reduction in the number of hydrogen bonds indicates that the hydrogen bond interactions within the system weaken after doping with calcium ion (Figure S8B), possibly due to the introduction of calcium ion altering the arrangement of cellulose molecules. The DFT calculation model is shown in Figure 2K. After doping with calcium ion, Ca–O coordination bonds are gener-

ated, which provide additional binding and significantly improve the stability of cellulose crystals. The DFT calculation results show that there is little change in tensile strength before and after calcium ion doping at short distance displacement ($<1.4\text{\AA}$); When the distance displacement is greater than $1.4\text{\AA}$, calcium ion doping begins to take effect, and the required tensile force value significantly increases (Figures S8C–D). This finding suggests that the Ca–O coordination formed through calcium ion doping effectively maintains structural integrity when subjected to substantial displacement and deformation.

In summary, the doping of cellulose crystals with calcium ions resulted in substantial alterations to both their local and overall structure. While certain interactions, such as van der Waals forces and hydrogen bonds, are weakened, the overall stability of the structure is markedly improved due to the formation of robust Ca–O coordination bonds. This finding indicates that calcium ion doping enhances the stability and binding strength of cellulose crystals by introducing new and stronger chemical bonds.

Mechanical properties and machinability

We conducted a comprehensive analysis of the mechanical properties of

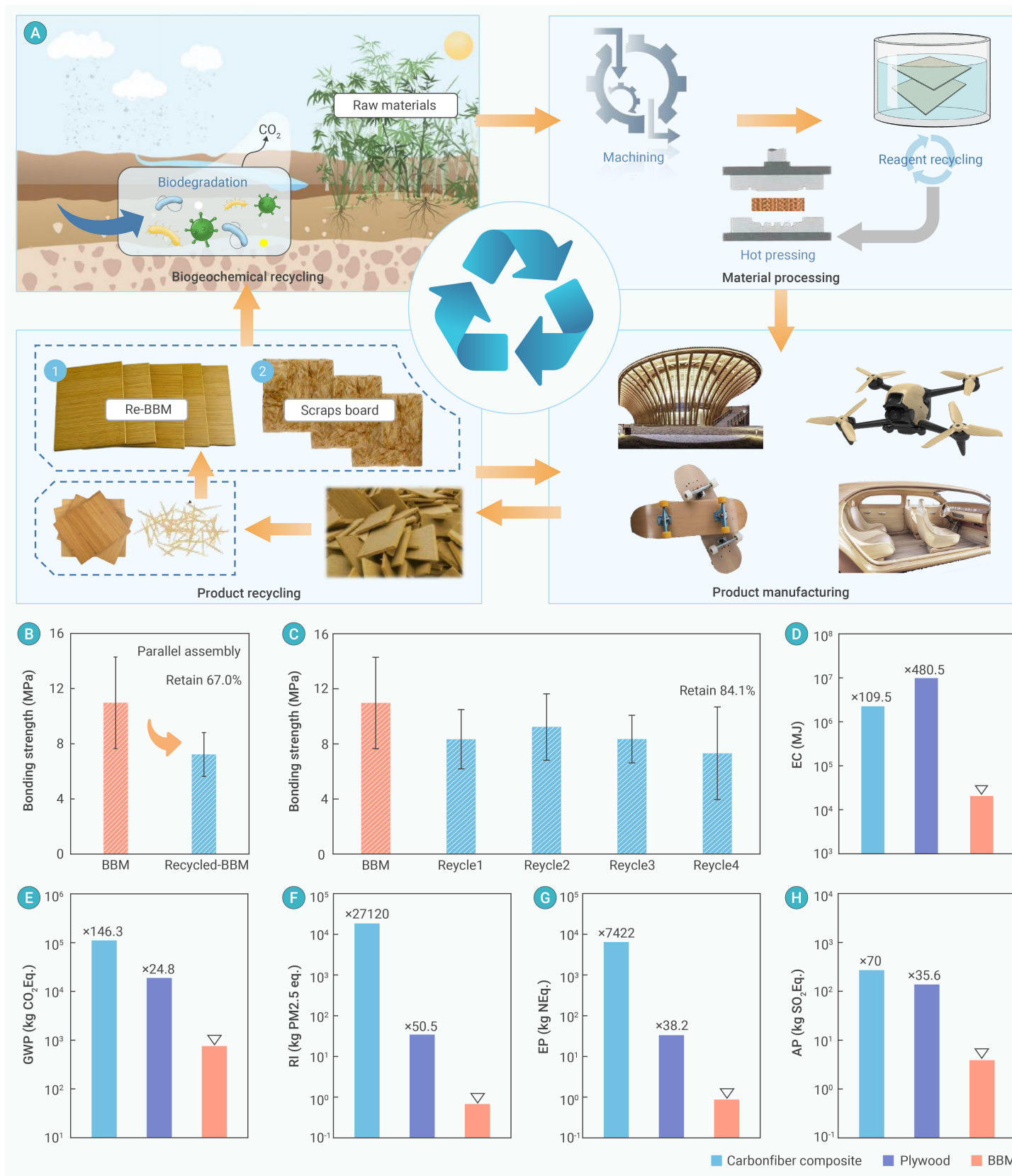


Figure 4. The environmental impacts and recyclability of BBM (A) Recycling of BBM. The environmental and energy impact of BBM, plywood, and carbon fiber composites regarding. (B) Bonding strength of recycled BBM and BBM. (C) The bonding strength of recycled BBM during four cycles after supplementing calcium ions. (D) Energy consumption; (E) global warming potential; (F) respiratory inorganics; (G) eutrophication potential and (H) acidification potential.

BBM, a plant fiber material known for its high toughness, which is comparable to that of carbon fiber and glass fiber composites (Figure 3A).³¹⁻³⁴ The BBM exhibits an average Shore hardness of 79.9, exceeding that of rigid PVC

and other plant fiber composites, and is close to the hardness of carbon fiber composites (Figure 3B).³⁵⁻³⁹ The tensile strength of BBM is 148.87 MPa, approximately 2.95 times greater than that of natural bamboo, while its

tensile elasticity modulus is enhanced by a factor of 26.34 compared to natural bamboo (Figures 3E-F). Additionally, results from three-point bending tests reveal that the bending strength and bending elasticity modulus of BBM are 157.4 MPa and 19,595.4 MPa, respectively, representing increases of 2.39 and 5.24 times compared to natural bamboo (Figures 3G-H). The enhancement of mechanical properties in BBM is primarily due to the removal of amorphous hemicellulose and some lignin after chemical treatment. This process leads to an increased relative content of crystalline cellulose in the treated bamboo (Figure S5B). Following hot-pressing treatment, the highly organized cellulose nanofibers form a dense structure, eliminating most pores in the bamboo cells, thereby effectively enhancing its load-bearing capacity under external forces.⁴⁰

The BBM is constructed using a quasi-isotropic assembly method, which imparts exceptional mechanical properties uniformly across all planar directions, ensuring consistent strength and stiffness. Figure 3C illustrates the quasi-isotropic tensile strength of BBM, which are equivalent across the 0-degree, 45-degree, and 90-degree axial directions. Furthermore, we investigated the effects of assembly angle count on the mechanical properties of the BBM. As demonstrated, specimens assembled at 45-degree and 90-degree showed lower tensile strength, bending strength, and bending elastic modulus compared to those of the quasi-isotropic assemblies (Figures S9A-B).

Figure 3I shows the excellent moldability of BBM, which can be fabricated into various complex three-dimensional structures, including arched, corrugated, dentate, and shovel shapes using auxiliary molds. We encapsulated a BBM corrugated structure between two bamboo veneers, creating a corrugated board. It can support a weight equivalent to that of a 1500-kilogram forklift, approximately 30,000 times the weight of the corrugated board itself (Figure S10A). Moreover, BBM can achieve repeated shaping and retain good dimensional stability (Figures S10B-C).

The BBM can be manufactured on a large-scale, a crucial factor for its application as an engineering material. The manufacturing process for large-scale BBM mirrors that of laboratory samples and consists of three primary steps: rotary cutting of bamboo veneer, chemical treatment and hot pressing. These methods are widely utilized in the wood processing and pulp industries, allowing for the customization of BBM in various dimensions to meet specific application requirements. Figure 3J presents a BBM specimen with dimensions of 1000 mm (length) × 650 mm (width) × 1 mm (thickness).

Furthermore, this synergistic strategy, which combines non-covalent and coordination bonds, is also applicable to wood plywood, as well as wood and bamboo fiberboards. As illustrated in Figure S11A, the bonding strength of poplar plywood and fir plywood are 8.3 MPa and 8.2 MPa, respectively. Figures S11B-D confirmed that this synergistic strategy can be applied to fiberboard. For bamboo and wood fiberboards, the bending strengths are 82.6 MPa and 38.8 MPa, the internal bonding strengths are 2.6 MPa and 1.4 MPa, respectively, while their water absorption thickness expansion rates are 33.9% and 28.2%.

Recycling and environmental impacts of BBM

The whole life cycle of BBM is a low-carbon and recycling process, encompassing the sourcing of raw materials, material processing, and product disposal after use (Figure 4A). Bamboo, a renewable and biodegradable natural resource, regenerates through photosynthesis and facilitates the cycling of water (H₂O), carbon dioxide (CO₂), and organic matter within ecosystems through self-metabolism. Following harvesting, natural bamboo can be converted into BBM through mechanical treatment, chemical treatment, and hot-pressing processes. This entire processing of BBM is characterized by low carbon emissions, and the chemical reagents used can be recovered and reused. Furthermore, once products made from BBM are discarded, they can still be recycled through mechanical processing and high-temperature water treatment, enabling secondary processing into non-adhesive bamboo composites. Even after multiple cycles of reuse, these materials remain biodegrade, posing no environmental burden. Therefore, the life cycle of BBM is both low-carbon and conducive recycling.

We conducted secondary processing of the recycled BBM and compared the bonding strength of BBM before and after recycling. Figure 4B shows the bonding strength of recycled BBM obtained through direct hot-pressing after

high-temperature water treatment, with a retention rate of only 67%. This reduction is attributed to the loss of metal ions during high-temperature water treatment, which reduces the number of coordination bonds present in recycled BBM. In contrast, when calcium ions were supplemented after the high-temperature water treatment, the bonding performance of the recycled BBM improved, achieving a retention rate of 84.1% after four recycling cycles (Figure 4C). This indicates that during the recycling process, coordination bonds were broken but were re-established upon the material's reintroduction to the ionic solution and subsequent hot pressing.

We conducted a gate-to-gate life-cycle assessment (LCA) to quantify the environmental impacts associated with the production of BBM. The results are compared to those of carbon fiber composites and plywood (Figures 4D-H & Table S3).^{23,41} This analysis focuses exclusively on the manufacturing process of BBM, excluding the extraction of raw materials and the disposal of the product. A comprehensive assessment of the chemicals and energy consumed during the preparation processes of BBM, which include dehemicellulose, oxidation, and hot-pressing treatments, enables to evaluate the environmental impact of industrial-scale BBM production (Figure 4A).

The environmental and energy impacts for the fabrication of 1m³ BBM were accessed. The results indicate that BBM manufacturing exhibits the lowest energy consumption, amounting to approximately 20,499.58 MJ, in contrast to carbon fiber composites, which require 2,244,000 MJ, and plywood, which requires 9,850,000 MJ (Figure 4D & Table S3). The major environmental analyses, including global warming potential (GWP), respiratory inorganics (RI), eutrophication potential (EP) and acidification potential (AP) for BBM, carbon fiber composites and plywood were also evaluated. BBM results in approximately 756.94 kgCO₂Eq. GWP, which is about 146.3 times lower than that of carbon fiber composites (110,772 kg CO₂eq.) and 24.8 times lower than that of plywood (18,800 kg CO₂eq.) (Figure 4E & Table S3).

Additionally, potassium hydroxide and sodium chlorite are commonly employed in the dehemicellulose and oxidation processes, which may present certain environmental risks. Among the various assessed environmental impact categories, BBM demonstrated a lower overall impact. Specifically, in comparison to carbon fiber composites and plywood, BBM exhibited reduced toxicity from respiratory inorganic emissions (Figure 4F). Furthermore, the values for EP and AP associated with BBM are negligible in comparison to those of carbon fiber composites and plywood (Figures 4G-H). In summary, the energy consumption and environmental impacts associated with the preparation of BBM are significantly lower than those involved in the production of carbon fiber composites and plywood. This green manufacturing method effectively minimizes environmental impact.

CONCLUSION

A non-adhesive fabrication strategy based on the synergistic effect of noncovalent and coordination bonds has been introduced to develop recyclable BBM from natural bamboo. This method involves a series of processes, including chemical pretreatment, ion cross-linking, assembly, and hot-pressing. The non-adhesive bonding of bamboo veneers is facilitated by hydrogen bonds and van der Waals forces at the interface of the pretreated bamboo, as well as coordination bonds formed between carboxyl groups and metal ions. The synergistic interaction of non-covalent bonds and coordination bonds enables BBM to achieve a bonding strength of 10.97 MPa, exceeding that of some traditional bio-based composites containing adhesives. The resulting BBM exhibits quasi-isotropic characteristics, maintaining consistent mechanical properties in all directions, with tensile and bending strengths measuring 148.87 MPa and 169.03 MPa, respectively. Additionally, its hardness and toughness, among other properties, have been significantly enhanced, far exceeding those of natural bamboo. More importantly, the BBM is biodegradable, recyclable, and reusable.

Once products made from BBM products are discarded, they can be recycled through mechanical processing or high-temperature water treatment. Even after four cycles of processing, the bonding strength remains at 84.1%. LCA results reveal that BBM has significantly lower environmental impacts compared to bio-based materials with adhesives, such as plywood and carbon fiber composites. The renewability of the raw material, low carbon footprint of the processing, and recyclability of the final product illustrate a closed-loop cycle. This presents a viable pathway for producing recyclable wooden products and promotes the exploration of sustainable materials, contributing to the development of a circular economy.

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AUTHOR CONTRIBUTIONS

Y.L. conceived the idea for this study and supervised the work. Z.F. designed the experiments. K.W., X.Y. carried out the experiments and performed material characterization. W.H. contributed to the DFT and MD calculations. W.L. contributed to the LCA analysis. Y.Z., W.Y. and W.L. provided useful suggestions. Z.F. and K.W. collaboratively analysed the data and wrote the manuscript. All authors contributed to the manuscript and approved the final version.

DECLARATION OF INTERESTS

The authors declare no competing interests.

DATA AND CODE AVAILABILITY

The authors declare that the main data supporting the findings of this study are available within the paper and its Supplementary Information. Extra data are available on reasonable request from the corresponding author.

SUPPLEMENTAL INFORMATION

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