

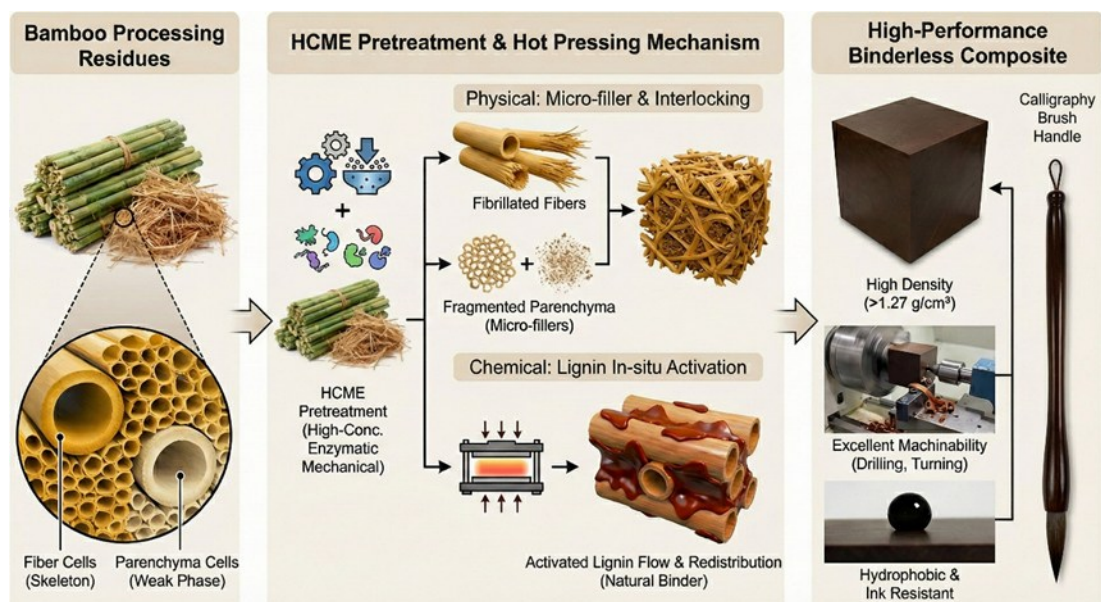
High-density Binderless Bamboo Brush Handles via High-consistency Mechano-enzymatic Pretreatment: Micro-filler Effect and Machinability

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GRAPHICAL ABSTRACT



High-density Binderless Bamboo Brush Handles via High-consistency Mechano-enzymatic Pretreatment: Micro-filler Effect and Machinability

Zheng Xue ^a and Chen Hang ^{b,*}

The resurgence of traditional culture has driven market demand for high-quality writing brushes. Natural bamboo frequently suffers from hygroscopic cracking, whereas polymer substitutes face the dual challenges of aesthetic deficiency and formaldehyde emission. While flat-molded binderless technology offers environmental advantages, it remains inadequate for addressing the high density and durability requirements of cylindrical artifacts. This study uses high-consistency mechano-enzymatic (HCME) pretreatment and cylindrical molding. It transforms bamboo processing residues into high-density binderless brush handles. Under HCME treatment, fibers fibrillate and parenchyma cells fragment. These changes induce microstructural reorganization. This reorganization constitutes a critical micro-filler effect. Porosity decreased to 3.27%, enabling a high density of 1.27 g/cm³. A thickness swelling (TS) below 5.1% effectively mitigated hygroscopic defects, while the material exhibited robust ink resistance. Attributable to micro-brittleness, chips broke cleanly during lathe turning; this eliminated fiber tearing and yielded a mirror-like surface finish. This formaldehyde-free approach achieved mechanical performance comparable to the tactile sensation of precious hardwoods but provided a potential pathway for extending binderless technology to the manufacturing of high-value cultural artifacts.

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Keywords: Bamboo processing residues; HCME pretreatment; Binderless technology; Chinese brush handles; High density; Hydrophobicity; Machinability

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INTRODUCTION

Chinese brushes serve as carriers of intangible cultural heritage, possessing significance that transcends their function as simple writing tools. The resurgence of the “Guochao” (national trend) has driven market demand for stationery that combines traditional aesthetics with modern durability (Qian *et al.* 2024). However, traditional brush manufacturing is constrained by the inherent thin walls and density variation of bamboo culms. This material is susceptible to humidity, which induces hygroscopic cracking, resulting in poor consistency and difficulties in long-term preservation (Li *et al.* 2022). Early attempts at substitution involved wood-plastic composites (WPC) or phenolic resin-impregnated bamboo. While these solutions addressed dimensional stability, the artificial plastic texture remains inferior to natural tactile qualities (Ziaei Tabari *et al.* 2017; Xu *et al.* 2023). Furthermore, the potential risk of formaldehyde emission (Lu *et al.* 2012) contradicts the eco-friendly philosophy intrinsic to calligraphy (Lao and Chang 2023).

Binderless technology offers a sustainable alternative. Biomass components, specifically lignin and hemicellulose, undergo *in-situ* activation and reorganization under hot-pressing without exogenous adhesives. The application of such eco-friendly flat fiberboards in furniture validates their potential to replace traditional resin-based composites (Cheng *et al.* 2024).

Binderless composites have advanced significantly in flat building materials. However, translating them into cylindrical brush handles remains challenging. Brush handles require exceptional density, tactile quality, and water resistance. Existing research on binderless bamboo materials has been largely confined to two-dimensional flat board preparation (Pintiaux *et al.* 2015), while three-dimensional special-shaped profile molding has remained as an unresolved challenge. During cylindrical molding, the combined effects of radial pressure gradients and heat transfer hysteresis hinder effective densification in the geometric core layer. Consequently, a void remains in the market for high-value cultural artifacts that are both genuinely green and high-performing. The density of conventional binderless bamboo boards rarely exceeds 1.0 g/cm³. In contrast, *Pterocarpus santalinus* provides a unique tactile sensation due to its high density of 1.1 to 1.2 g/cm³, a quality unattainable by low-density materials (Arunkumar and Joshi 2014). Current densification theories rely heavily on the physical entanglement of long fibers (Shi *et al.* 2023). Mechanisms that achieve high density *via* microstructural filling must be explored. Such mechanisms are needed to ensure a low center of gravity. A low center of gravity is necessary for writing stability. Furthermore, writing brush applications inherently involve water and colloid-rich ink. Although enzymatic pretreatment enhances bonding, the hydrophilicity of the material persists (Sun *et al.* 2021; Wang *et al.* 2022), creating a high risk of hygroscopic swelling. Conventional low-consistency enzymatic hydrolysis often induces excessive cellulose degradation. The resulting material brittleness leads to chipping during lathe turning and fine carving, which compromises precision machining requirements (Ding *et al.* 2023).

This study realizes the high-value utilization of bamboo processing residues through a high-consistency mechano-enzymatic (HCME) strategy. Traditional low-consistency enzymatic hydrolysis typically limits solid content to below 10%. This causes substantial loss of natural binders (*e.g.*, lignin) into the waste liquid. In contrast, HCME technology not only preserves these key components but also significantly reduces water consumption and subsequent drying energy. Mechanical shear force and cellulase are applied synergistically at 35% solid content. Bamboo fibers undergo *in-situ* fine defibrillation. Meanwhile, lignin surface activation is triggered. High temperature and pressure within custom cylindrical molds induce material self-bonding, ultimately forming high-performance composite brush handles.

This study's objectives were the following: application of binderless technology to Chinese traditional brush handle manufacturing; elucidation of the micro-filler effect; and demonstration of the hydrophobic and machining mechanisms. This study develops a prototype for a cultural artifact that is completely formaldehyde-free, retains a natural bamboo scent, and exhibits excellent performance. The study reveals how parenchyma cell fragments generated during HCME fill the voids in the fiber skeleton. This mechanism constitutes the basis for overcoming density bottlenecks, and thereby addressing the theoretical challenge of achieving a texture comparable to precious hardwoods in binderless materials.

LITERATURE REVIEW

Evolution of Bamboo-based Composites

As an alternative to traditional wood, fast-growing bamboo offers significant potential (Adier *et al.* 2023). The technical iteration of bamboo-based composite preparation has evolved from “full-gluing” to “binderless” processes. Although scrimber and laminated bamboo lumber exhibit excellent strength, their heavy reliance on phenolic or urea-formaldehyde resins during manufacturing poses health risks associated with formaldehyde emission (Lao and Chang 2023).

Binderless technology has emerged as a green solution. Steam explosion, heat treatment, or mechanical activation induces biomass to achieve self-bonding (Chen *et al.* 2023). While mechanical reinforcement of two-dimensional flat materials dominates current research (Cheng *et al.* 2024), converting these into 3D special-shaped profiles, such as cylinders, presents severe challenges regarding material fluidity and densification uniformity. Research on molding processes and applications for fine crafts remains scarce.

Self-bonding Mechanism and Microstructural Reorganization

High densification and uniform molding of 3D profiles depend on dual breakthroughs at the microscopic level: the multi-scale interlocking network formed by long fiber skeletons and micron-scale fillers (Wang *et al.* 2022), and the *in-situ* activation and redistribution of lignin during hot-pressing (Cheng *et al.* 2024; Pintiaux *et al.* 2015).

Studies indicate that for binderless biomass materials to exceed the density of high-grade hardwoods ($> 1.1 \text{ g/cm}^3$), internal microscopic voids must be effectively eliminated. Constructing a multi-scale interlocking network constitutes one of the most effective pathways to achieve ultra-high density comparable to high-grade hardwoods ($> 1.1 \text{ g/cm}^3$). (Wang *et al.* 2022). Traditional mechanical separation removes parenchyma cells, which impairs packing efficiency. Moderately fragmented parenchyma cells can act as micro-fillers, embedding within the gaps of the long fiber skeleton to achieve three-dimensional interlocking (Lao and Chang 2023). However, systematic research on the precise regulation of fragmentation degree and its contribution to extreme densification remains absent.

The core mechanism endowing binderless materials with water resistance lies in the glass transition and redistribution of lignin (Cheng *et al.* 2024; Pintiaux *et al.* 2015). Thermoplastic lignin softens during hot-pressing and migrates to the surface layer, where a natural hydrophobic binding layer forms. However, the tight internal bonding forces within natural bamboo constitute significant resistance to migration. Biological enzyme pretreatment selectively severs the hydrogen bond network; the exposed active sites of lignin accelerate activation and flow (Cheng *et al.* 2024; Wang *et al.* 2022; Chen *et al.* 2023). Existing literature rarely addresses the directional migration mechanism of lignin to the material surface under the synergistic action of high-intensity mechanical force and enzymatic hydrolysis, nor its application in complex profiles.

Figure 1 depicts the proposed mechanistic pathway transforming bamboo processing residues into high-performance brush handles. Fibers and parenchyma cells in the raw material undergo structural reorganization and chemical modification *via* the HCME process. Fiber fibrillation coupled with cell fragmentation and lignin exposure ultimately leads to three key performance indicators: high density, hydrophobicity, and machinability.

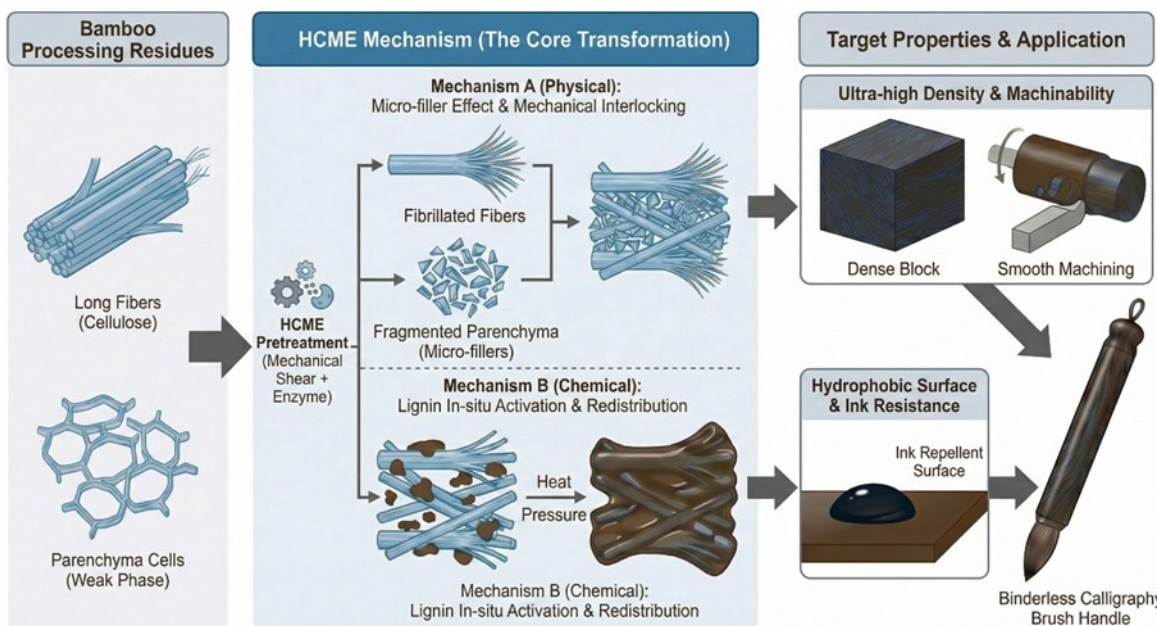


Fig. 1. Conceptual framework diagram

EXPERIMENTAL

Preparation and Screening of Raw Materials

Bamboo processing residues from 5-year-old Moso bamboo harvested in Zhejiang Province, China, formed the material basis of this study, with sawdust and shavings constituting the primary raw materials. Established standardized procedures regulated the collection and pretreatment steps (Cheng *et al.* 2024). To ensure the fineness of the finished brush handles, the raw materials were crushed and screened. Bamboo powder (BP) with a particle size distribution between 40 and 60 mesh was selected as the substrate and dried at 60 °C to constant weight to eliminate moisture influence. Chemical composition determination followed the standard analytical procedures of the National Renewable Energy Laboratory (NREL) (Sluiter *et al.* 2008). The enzyme preparation used was commercial-grade cellulase (CTec3) produced by Novozymes, with a nominal enzyme activity of 9.5 FPU/mL.

Synergistic HCME Pretreatment

A modified HCME strategy realized efficient defibrillation and *in-situ* activation while maintaining high solid content. Cheng *et al.* (2024) provided the reference frame for core parameter settings. An acetate buffer (pH 4.8) preheated to 50 °C dissolved the cellulase and was sprayed uniformly onto the dry BP. A solids content of 35% (w/w) strictly simulated an industrial high-consistency environment. The enzyme dosage was set at 0.007 g (CTec3) per gram of dry substrate. A twin-screw kneader equipped with a Sigma mixer (NH-1, Rugao Guanchen Machinery Factory, China) supported the reaction at 50 °C. To investigate the influence of treatment extent on handle performance, the experiment established five time gradients: 0 h, 0.5 h, 2 h, 4 h, and 8 h (labeled as BP-0 to BP-8, respectively). Upon completion, samples were immediately placed in a 100 °C environment for 10 min to deactivate the enzymes, then dried at 60 °C for subsequent use.

Directional Molding of Brush Handle Blanks

The specific geometric characteristics of brush handles dictated the use of a dedicated stainless steel mold. An inner diameter of 12 mm combined with a length of 200 mm defined the molding space. This experiment referenced the molding strategy in Shi *et al.* (2023). This study adopted a layered filling and axial compression process using a closed mold. This approach overcomes pressure transmission attenuation during cylindrical molding. It also improves density uniformity under high pressure. Quantitatively weighed HCME-pretreated BP was manually filled and pre-compacted. A flat vulcanizer performed the hot-pressing task. A core process parameter matrix of 180 °C, 20 MPa pressure, and 10 min holding time was established. After the hot-pressing cycle, the mold was allowed to cool naturally to room temperature under pressure; subsequent demolding yielded high-density binderless self-bonding bamboo composite (BSBC) cylindrical rods.

Precision Machining and Surface Finishing

Machinability assessment and final product fabrication drove the post-processing steps that simulated traditional brush-making crafts. Ding *et al.* (2023) guided the optimization of cutting parameters to avoid surface defects. A small computer numerical control (CNC) lathe turned the rods into standard forms with a slightly thicker center and tapered ends. A rotational speed of 1500 rpm and a feed rate of 0.5 mm/rev determined the cutting conditions. High-end stationery requires strict tactile standards. Sanding the handles with 400-, 800-, and 1200-mesh sandpaper was performed sequentially. Wool wheel polishing finally resulted in a mirror-like luster.

Characterization and Application Simulation

Multi-scale characterization methods achieved a comprehensive assessment of the microstructural and macro-performance of the BSBC. The BP-0 to BP-8 sample series was used to investigate the chemical structural evolution during HCME pretreatment. A Nicolet iS10 Fourier transform infrared (FTIR) spectrometer equipped with an attenuated total reflectance (ATR) accessory was used. The spectra were collected over a range of 4000 to 400 cm^{-1} with a resolution of 4 cm^{-1} . Cellulose crystallinity index (CrI) was determined using an X-ray diffractometer (XRD, Bruker D8 Advance, Germany) with a Cu K α radiation source ($\lambda = 1.5406 \text{ \AA}$), a scanning range (2θ) of 5° to 40°, and a scanning speed of 2°/min. The CrI values were calculated using the Segal method (Segal *et al.* 1959) based on the intensity of the (200) crystalline peak (around $20 \approx 22^\circ$) relative to the amorphous background intensity (around $20 \approx 18^\circ$). Thermal stability was evaluated *via* thermogravimetric analysis (TGA, TGA 550, TA Instruments, USA), heating samples from room temperature to 600 °C at a rate of 10 °C/min under a nitrogen atmosphere. The NREL standard procedures governed the quantitative analysis of cellulose, hemicellulose, and lignin.

A GeminiSEM 360 scanning electron microscope was employed to comparatively observe the defibrillation state of BP before and after HCME treatment. In-depth analysis of the microscopic bonding characteristics of BSBC fracture surfaces revealed the reinforcement mechanism. Gold sputtering for 60 s endowed the samples with the conductivity and clarity required for testing. An accelerating voltage of 3 to 5 kV set the scanning baseline. A SkyScan 2214 micro-computed tomography (Micro-CT) system performed quantitative characterization of material densification. The internal pore structure underwent non-destructive scanning. An operating voltage of 50 kV, a current of

80 μA , and a resolution of 4.00 μm defined the imaging precision. Three-dimensional model reconstruction and porosity calculations were executed by Avizo software following the methodological logic of Wang *et al.* (2022).

The GB/T 17657 (2022) standard established the baseline for physical and mechanical performance determination. Data for density, 24 h thickness swelling, three-point flexural strength, and internal bonding strength were obtained using these standards. Although originating from board standards, their applicability for evaluating high-density biomass composites has been frequently validated in relevant research (Li *et al.* 2022). Mechanical tests were conducted on Instron 5848 (Instron, UK) and WDW-E100D (Jinan Shijin Group, China) universal testing machines. A contact angle meter (OCA20, Dataphysics Instrument, Germany) recorded dynamic water contact angle (WCA) changes (0 to 60 s) on the material surface to evaluate wettability. Statistical significance among groups was evaluated using one-way ANOVA, followed by Duncan's multiple range test ($p < 0.05$). The detailed pairwise comparisons are reported in Supporting Information (Table S1). One-way ANOVA was used to determine whether statistically significant differences existed among multiple treatment groups, and Duncan's test was subsequently applied to identify specific group differences.

This study referenced the processing quality evaluation standards proposed by Ding *et al.* (2023). Machinability was assessed by observing microscopic surface smoothness and edge integrity after turning. Ink resistance tests validated handle durability. Finished handles underwent full immersion in standard calligraphy ink for 24 h. Capturing phenomena of surface ink penetration or swelling were used to quantify the material's resistance to colloidal ink.

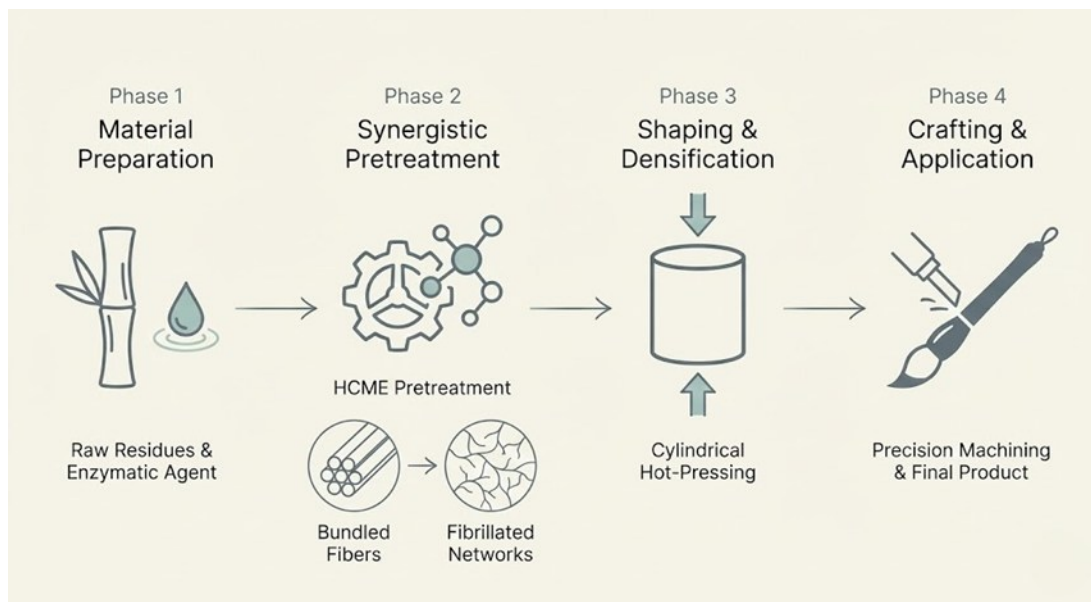


Fig. 2. The methodological workflow of the study

Figure 2 illustrates the four key preparation stages from bamboo processing residues to finished brush handles. Raw material screening and preparation occurred first. The HCME pretreatment established the material foundation. Directional hot-pressing molding within a cylindrical mold realized structural reorganization. The final form of the binderless brush handles was sculpted *via* precision lathe turning and polishing processes.

RESULTS

Microstructural Reorganization Driven by HCME

Scanning electron microscopy (SEM) images intuitively revealed the noticeable reshaping effect of HCME pretreatment on the micromorphology of bamboo powder (BP). Figure 3 presents the complete evolution. The original vascular bundles and parenchyma cell structure of Moso bamboo in Fig. 3a served as a reference. BP-0 (Fig. 3b) maintained a smooth and dense fiber bundle appearance, with blocky parenchyma cells remaining structurally intact. The quantitative proportion of bamboo fibers and parenchyma cells in the raw BP is provided in Supporting Information (Fig. S1), which confirms the coexistence of fiber bundles (60.1%) and a considerable fraction of parenchyma tissue (39.9%) as the structural basis for the subsequent micro-filler effect.

Prolonging the mechano-enzymatic synergistic treatment time drove the progressive evolution of fiber morphology. BP-0.5 (Fig. 3c) and BP-4 (Fig. 3d) show increased fiber surface roughness, signs of fiber bundle loosening and initial defibrillation, and the shedding and fragmentation of parenchyma cells. The 8-h treatment (BP-8, Fig. 3e) catalyzed the most marked qualitative change. Bamboo fibers presented strong “fibrillation” (broom-like) characteristics, where dense fiber bundles were peeled into microfibers, leading to a sharp expansion in specific surface area.

The combined effect of strong shear force and enzymatic hydrolysis pulverized the originally aggregated parenchyma cells into numerous micron-scale fragments. A multi-scale physical interlocking network was jointly constructed by these refined fiber branches and cell fragments; this established the foundation for the densified structure formed during the hot-pressing process.

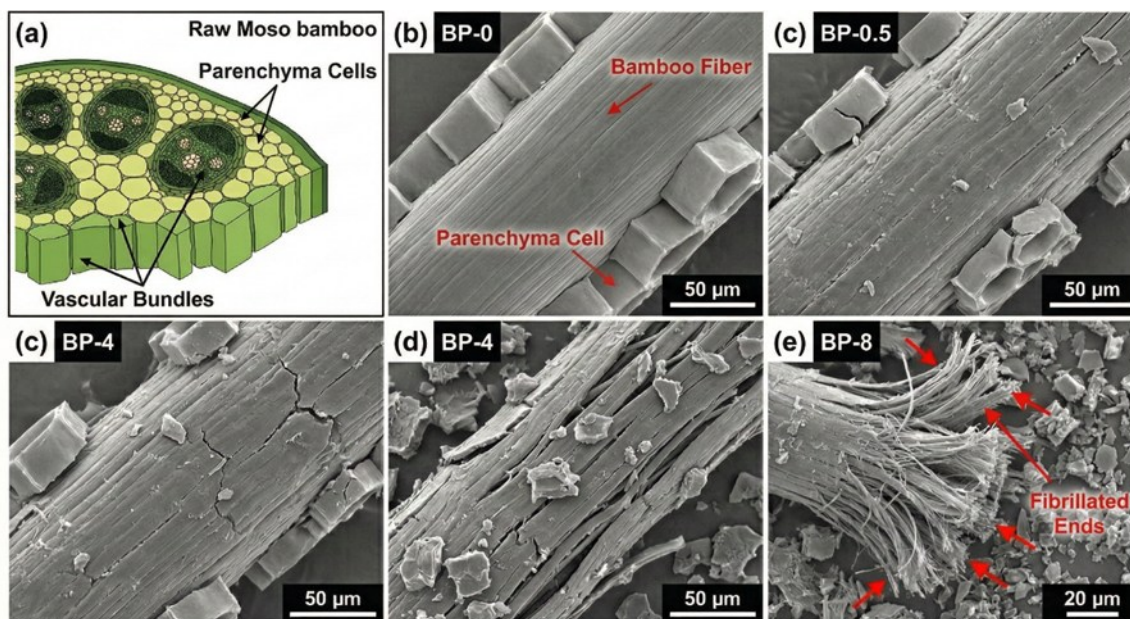


Fig. 3. SEM images revealing the microstructural evolution of bamboo powders during HCME pretreatment; (a) Schematic illustration of Moso bamboo structure; (b through e) SEM micrographs of BP treated for different durations: (b) BP-0, (c) BP-0.5, (d) BP-4, and (e) BP-8

Evolution of Chemical Components and Thermal Stability

Chemical component analysis revealed that HCME treatment induced microscopic redistribution of components while retaining the overall chemical skeleton of the bamboo. The enzymatic hydrolysis effect during HCME was further verified by measuring glucose released into the filtrate (Supporting Information, Fig. S2), confirming that cellulase activity occurred without causing excessive degradation of the structural framework. The FTIR spectra (Fig. 4a) showed that the positions of characteristic peaks remained consistent before and after treatment, indicating that the main chemical bonds of cellulose, hemicellulose, and lignin did not fracture. Quantitative analysis of chemical composition (Fig. 4b) further confirmed that the relative contents of hemicellulose, cellulose, and lignin remained basically stable with only minor fluctuations as treatment time increased. This indicates that the mild HCME pretreatment did not cause significant degradation of the main components, thereby preserving cellulose as the structural skeleton and lignin as the binder. The XRD patterns (Fig. 4c) showed that the cellulose crystallinity (CrI) of BP-8 increased slightly to 53.0% compared to BP-0. This is attributed to the preferential enzymatic hydrolysis of amorphous cellulose, which retained the high-strength crystalline regions.

However, XPS surface energy spectrum analysis (Fig. 4e, f) revealed significant surface chemical changes. The C1s XPS spectra of intermediate samples are provided in Supporting Information (Fig. S6), confirming the gradual enrichment of lignin-related C–C/C–H components with increasing treatment time. Compared to BP-0 (Fig. 4e, C1 ~37%), the relative content of the C1 peak on the surface of BP-8 (Fig. 4f), attributed to C–C/C–H bonds in lignin and extractives, rose significantly to ~53%. This trend indicates that enzymatic hydrolysis effectively disrupted the cell wall structure, prompting lignin to migrate to and enrich the fiber surface (surface enrichment), which provided the key chemical basis for subsequent hot-pressing self-bonding. Furthermore, thermogravimetric analysis (TG/DTG) curves (Fig. 4d) indicated that the thermal decomposition behavior of all samples was highly consistent. Additional thermogravimetric comparison data of the resultant BSBC are provided in Supporting Information (Fig. S3), further confirming that HCME pretreatment did not compromise the thermal stability required for hot-press molding at 180 °C. This validates that HCME treatment did not impair the thermal stability of the bamboo skeleton, and the material is fully capable of adapting to the 180 °C hot-pressing processing environment.

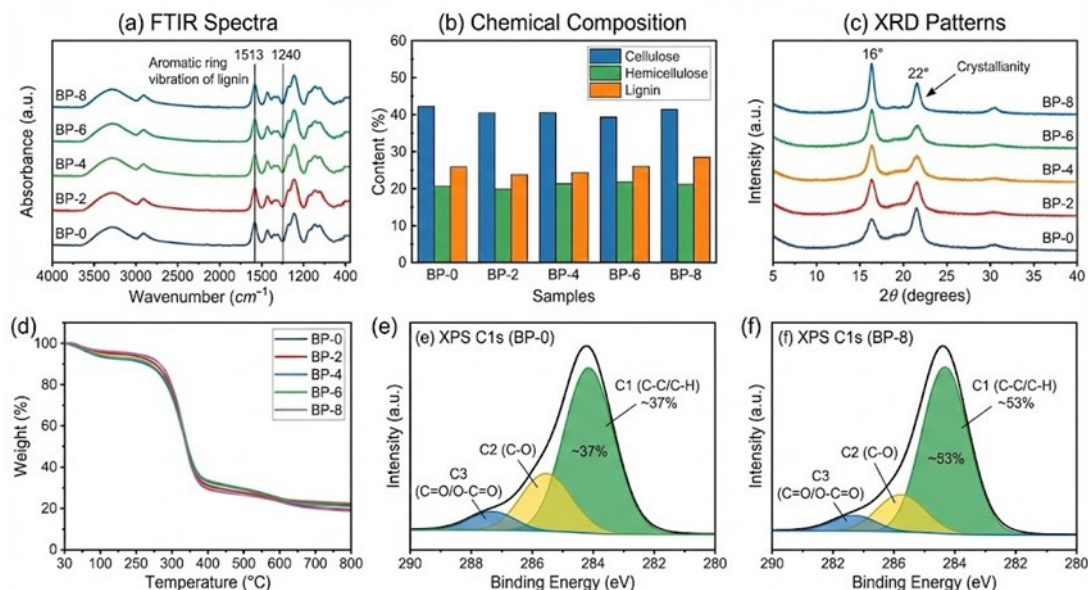


Fig. 4. Evolution of chemical components and thermal stability of bamboo after HCME pretreatment: (a) FTIR spectra showing negligible changes in characteristic peaks; (b) Relative contents of hemicellulose, cellulose, and lignin, exhibiting only minor fluctuations; (c) XRD patterns with cellulose CrI slightly increasing from 50.3% (BP-0) to 53.0% (BP-8); (d) TGA curves demonstrating that all samples maintained identical thermal decomposition behavior and high stability up to 180 °C; (e and f) XPS C1s high-resolution spectra of BP-0 and BP-8, where the increase in C1 peak area (from ~37% to ~53%) indicates the migration and enrichment of lignin on the fiber surface

Enhanced Physical Properties and Pore Densification

Table 1 records the significant improvement in the physical properties of BSBC with prolonged HCME pretreatment time. Density increased from 1.23 g/cm³ for BSBC-0 to 1.27 g/cm³ for BSBC-8. This value is unattainable by the vast majority of natural bamboo. It successfully surpassed the density threshold of precious hardwoods like *Pterocarpus santalinus* (1.1 to 1.2 g/cm³), endowing the brush handle with a superior quality of steady tactile sensation.

Micro-CT analysis provided direct verification of the densification process (Fig. 5). In the 2D slice images (Fig. 5a to c) and threshold-segmented images (Fig. 5d to f), BSBC-0 contained a large network of connected black pores. Prolonged treatment time resulted in an extremely dense and smooth cross-section for BSBC-8, with noticeable pore elimination. Representative pore characteristics of intermediate samples (BSBC-2 and BSBC-4) are shown in Supporting Information (Fig. S4), illustrating the progressive densification pathway with increasing HCME duration. The 3D reconstruction model (Fig. 5g through i) quantified this trend. Porosity remained high at 7.62% in BSBC-0 but decreased sharply to 3.27% in BSBC-8, a reduction of 57.1%.

Residual pores in BSBC-8 mostly appeared as isolated closed pores (Fig. 5i). This unconnected structure effectively blocked capillary water absorption paths, establishing the microscopic mechanism for improved water resistance. The BSBC-8 exhibited a superior hydrophobic barrier effect. The surface water contact angle (WCA) remained stably anchored above 90°. The dynamic evolution of WCA over 60 s for all samples is presented in Supporting Information (Fig. S5), further demonstrating the enhanced

hydrophobic stability induced by HCME treatment. The 24-h thickness swelling (TS) of 5.1% was far superior to the limit value for premium grade products in national standards.

Table 1. Physical Properties of BSBC Pretreated with Different HCME Durations

Sample ID	Density (g/cm ³)	24 h Water Absorption (%)	24 h Thickness Swelling (%)	Water Contact Angle (°) (at 0 s)
BSBC-0	1.23 ± 0.02	20.3 ± 1.2	10.3 ± 0.8	69.6 ± 2.1
BSBC-0.5	1.24 ± 0.01	17.8 ± 1.5	8.2 ± 0.5	71.5 ± 3.2
BSBC-2	1.24 ± 0.03	14.4 ± 1.1	6.7 ± 0.4	83.7 ± 2.5
BSBC-4	1.25 ± 0.02	11.5 ± 0.9	5.6 ± 0.3	90.6 ± 4.1
BSBC-8	1.27 ± 0.01	9.3 ± 0.6	5.1 ± 0.2	98.8 ± 1.8

Note: Data are presented as mean ± standard deviation (n = 5)

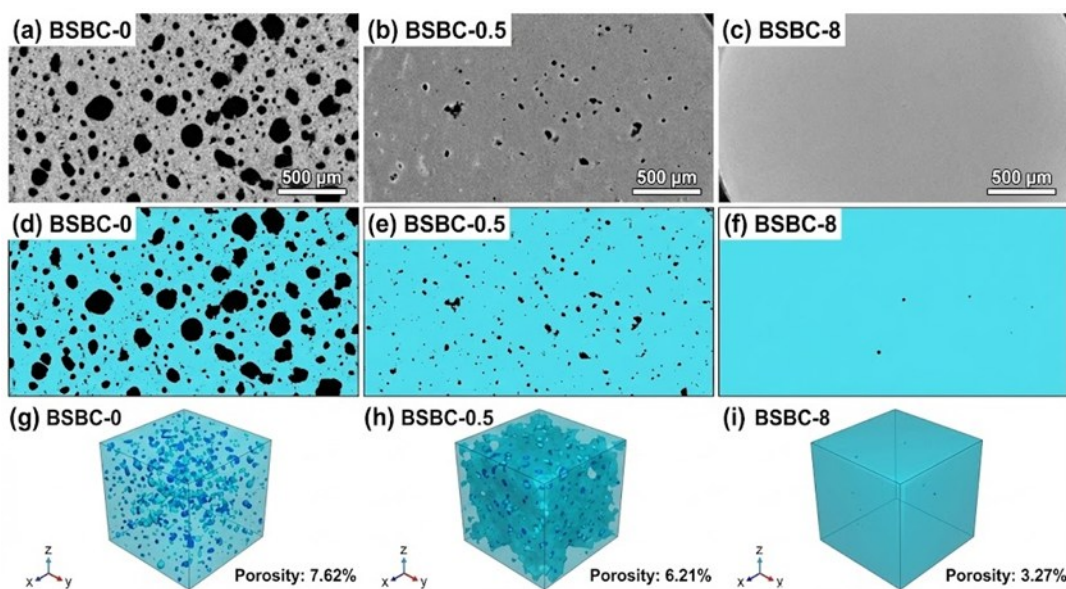


Fig. 5. Micro-CT visualization of pore structure evolution in BSBC with prolonged HCME pretreatment: (a to c) Representative 2D cross-sectional slices of BSBC-0, BSBC-0.5, and BSBC-8; (d to f) Corresponding threshold-segmented images (black regions represent pores); (g to i) 3D reconstructed pore networks with quantified porosity of 7.62%, 6.21%, and 3.27%, respectively. Scale bars: 500 μ m.

Mechanical Performance and Machinability

Mechanical performance test results indicated that HCME pretreatment significantly enhanced the material's mechanical strength. The detailed multiple comparison and statistical significance analysis (ANOVA with Duncan's test) for physical and mechanical properties is summarized in Supporting Information (Table S1), confirming the reliability of the observed improvements ($p < 0.05$). Figure 6a clearly shows that with prolonged HCME treatment time, the flexural strength of BSBC increased from 26.4 to 37.8 MPa, and the flexural modulus increased from 3.89 to 4.64 GPa, both

exhibiting significant upward trends. The stress-strain curves (Fig. 6b) further revealed a fundamental shift in mechanical behavior: unlike the longer yield stage exhibited by BSBC-0, BSBC-8 underwent brittle fracture immediately after peak stress, with no obvious necking phenomenon. This transition in fracture mode aligns with the high flexural modulus of 4.64 GPa in Table 2, indicating significantly enhanced material rigidity.

Internal bonding strength (Fig. 6d) steadily increased from 1.36 MPa to 1.84 MPa, confirming the significant enhancement of interfiber bonding forces. The SEM fracture morphology (Fig. 6c) revealed the transition in failure mode: large amounts of long fiber pull-out were visible at the BSBC-0 fracture, indicating weak interfacial bonding; BSBC-8 presented a flat brittle fracture surface, where an extremely strong interface bond formed between fibers and the matrix.

In actual processing tests, BSBC-8 demonstrated excellent adaptability to lathe turning (Fig. 6e). Compared to the rough, loose surface of BSBC-0 (rough), the turned surface of BSBC-8 was extremely smooth and dense (smooth), with no obvious pore exposure or fuzzing observed microscopically.

Finally, ink resistance tests (Fig. 6f) further confirmed that after 24 h of immersion, the raw bamboo control group showed severe ink penetration and diffusion, whereas the surface of the BSBC-8 treated sample exhibited excellent ink repellency. As shown in Fig. 6f, the untreated bamboo exhibited extensive surface staining and ink diffusion into the porous structure, whereas BSBC-8 maintained a dense and visually clean surface with only superficial staining. For clarity, ink resistance was assessed using a semi-quantitative protocol, as described in the Experimental section. Only slight wiping was needed to restore its original appearance, fully meeting the stringent requirements for high-end calligraphy utensils.

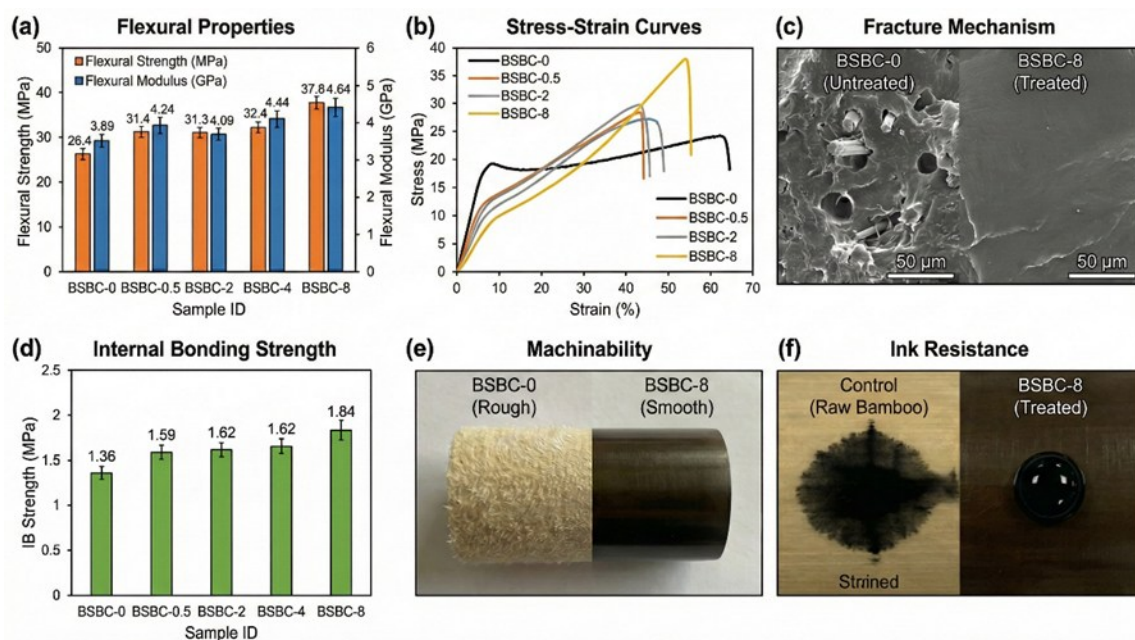


Fig. 6. Mechanical properties, fracture behavior, bonding strength, machinability, and ink resistance of BSBC samples with different HCME pretreatment durations: (a) Flexural strength and modulus; (b) Representative stress-strain curves; (c) SEM micrographs of fracture surfaces comparing BSBC-0 (untreated) and BSBC-8 (treated); (d) Internal bonding strength; (e) Machined surface comparison of BSBC-0 (rough) and BSBC-8 (smooth); (f) Ink resistance test of the raw bamboo control and BSBC-8 (treated) after 24 h immersion

Table 2. Mechanical Properties of BSBC Pretreated with Different HCME Durations

Sample ID	Flexural Strength (MPa)	Flexural Modulus (GPa)	Internal Bonding Strength (MPa)
BSBC-0	26.44 ± 2.15	3.89 ± 0.22	1.36 ± 0.12
BSBC-0.5	31.36 ± 1.84	4.24 ± 0.31	1.59 ± 0.15
BSBC-2	31.31 ± 2.03	4.09 ± 0.28	1.62 ± 0.18
BSBC-4	32.41 ± 1.56	4.44 ± 0.19	1.66 ± 0.14
BSBC-8	37.79 ± 2.31	4.64 ± 0.35	1.84 ± 0.16

DISCUSSION

This study aimed to develop a high-density, binderless bamboo-based composite suitable for brush handles, overcoming the limitations of traditional natural bamboo and modern resin-based substitutes regarding density uniformity, water resistance, and environmental safety. The results indicate that the 8-h HCME pretreatment played a critical role in enhancing densification and interfacial bonding performance. The density of 1.27 g/cm³ and strength data of 37.8 MPa was higher than the density commonly reported for binderless bamboo composites and also exceeded that of many natural hardwoods. This comparison provides a clearer basis for evaluating the performance improvement achieved in this study. Compared with recent reports on binderless bamboo composites (*e.g.*, Shi *et al.* 2023; Cheng *et al.* 2024), the present work demonstrates a further advancement in densification efficiency and property integration for cylindrical profiles. Beyond the observed mechanical improvements, the practical significance of the 8-h HCME pretreatment also resides in its industrial scalability.

From an industrial scalability perspective, the 8-h HCME pretreatment represents a practical balance between structural refinement and process feasibility. Compared with chemical-intensive delignification requiring strong acids/alkalis and extensive washing (Nagalakshmaiah *et al.* 2016), HCME operates at high solid content without bulk polymer dissolution, thereby reducing liquid waste and post-drying demand. High-consistency enzymatic–mechanical processing has been shown to preserve fiber integrity while enhancing fibrillation efficiency (Wang *et al.* 2022), indicating compatibility with existing fiber-refining and thermoforming lines. Although the 8-h duration appears long at laboratory scale, industrial application could employ parallelized or continuous high-consistency reactors, where residence time is decoupled from throughput. Thus, industrial feasibility depends less on absolute treatment time than on seamless integration into continuous densification–pressing workflows without additional chemical recovery or wastewater burdens. In this regard, HCME offers a mild, solvent-free strategy aligned with sustainable bamboo composite manufacturing (Shi *et al.* 2023; Cheng *et al.* 2024).

Key Findings and Mechanism

The unique synergistic defibrillation and micro-filler effect induced by HCME constitutes the fundamental cause of material densification. The specific contribution of cellulase to the mechanical enhancement of BSBC-8 is further illustrated in Supporting Information (Fig. S7), suggesting that enzymatic-assisted fibrillation contributed to the observed enhancement of interfacial bonding. Although Shi *et al.* (2023) prepared binderless bamboo *via* mechanical combing, its density often remained around 1.0 g/cm³, constrained by microscopic voids left by long fiber accumulation. This study observed *via* SEM (Fig. 3) that strong mechanical shear force under high solid content effectively pulverized parenchyma cells, traditionally considered "waste," into micron-scale fragments. Micro-CT analysis (Fig. 5) supports the proposed view. Parenchyma cell fragments may function as intrinsic micro-fillers. During hot-pressing, they redistribute and compact into voids within the long-fiber skeleton. The presence of these rigid cell fragments locally constrains deformation and limits the relative mobility of adjacent fiber segments, resulting in a densified and mechanically integrated network. This behavior bears resemblance to the reinforcement mechanisms reported in particle-filled composite-like systems (Chen *et al.* 2023). Although the presence of micron-scale parenchyma fragments and the concurrent porosity reduction support the proposed micro-filler effect, the exact filler volume fraction was not directly quantified in this study; therefore, the micro-filler mechanism is presented as an inference based on SEM/Micro-CT observations (see Supporting Information, Fig. S1 and Fig. S4 for supplementary context).

Beyond physical structural reorganization, the chemical selectivity of HCME treatment constitutes another key factor guaranteeing high performance. Both FTIR and TGA results (Fig. 4) confirm that despite prolonged synergistic mechano-enzymatic action, the main chemical skeleton of the bamboo did not undergo noticeable degradation. This stands in sharp contrast to traditional strong acid or alkali pretreatments, where impurity removal is often accompanied by the fracture of cellulose long chains and the destruction of crystalline structures (Nagalakshmaiah *et al.* 2016). In this experiment, HCME demonstrated a mild and precise modification advantage. The slight increase in CrI from 50.3% to 53.0% is attributed to the selective hydrolysis of amorphous regions by cellulase, as these non-crystalline regions often represent the weak points in the mechanical performance of natural fibers (Park *et al.* 2010; Liu *et al.* 2012).

The lignin *in-situ* activation and surface hydrophobization mechanism played a decisive role in this study. Compared with conventional low-solids enzymatic pretreatment approaches, the high-consistency mechano-enzymatic (HCME) system adopted here enabled efficient fibrillation/defibrillation and facilitated the exposure and surface enrichment of lignin-related components, which is beneficial for subsequent self-bonding during hot pressing (Cheng *et al.* 2024; Wang *et al.* 2022). During the hot-pressing process, this thermoplastic lignin enriched on the surface underwent a glass transition, forming a continuous hydrophobic "glaze layer." This not only effectively blocked water molecule penetration paths but also endowed the brush handle surface with natural resistance to colloidal ink (Fig. 6f), substantially mitigating the drawbacks of susceptibility to mildew and staining in traditional bamboo handles.

Furthermore, the marked improvement in machinability stems from a fundamental shift in fracture mechanism. With the sustained enhancement of interfacial bonding strength, the failure mode of the material under load and cutting gradually transitioned from ductile fracture, dominated by interfacial sliding and fiber pull-out, to microscale brittle fracture characterized by rapid failure of the fiber body (Fig. 6c). Existing studies indicate

that conventional bamboo and ordinary fiber-reinforced composites often produce defects, such as fiber tearing, fuzzing, and edge chipping, during machining due to insufficient interfacial bonding (Ding *et al.* 2023). In contrast, the micro-brittleness induced by the high bonding strength presented in this study did not manifest itself as overall material performance degradation but was confined to microscale fracture behavior under machining load. The rationality of this behavior is validated by the simultaneous improvements in flexural modulus, internal bonding strength, and ink resistance.

Under this fracture mechanism, fibers are instantly and cleanly severed during cutting. They do not undergo plastic pull-out. This reduces surface tearing and burr formation. A dense, smooth surface finish is achieved (Fig. 6e). Conversely, traditional high-toughness WPC often generates difficult-to-remove burrs and tool sticking problems during turning due to significant plastic deformation (Ding *et al.* 2023). Therefore, the high stiffness–microscale brittle fracture characteristic exhibited by this material aligns highly with the material removal mechanism that is associated with precision machining, effectively overcoming the tough and difficult to machine bottleneck long existing in biomass materials.

Scientific Implications and Practical Applications

Although scholars (Cheng *et al.* 2024) preliminarily explored HCME technology, this study further establishes the “degree of parenchyma fragmentation” as a key variable for the densification of binderless materials. These results suggest that in specific systems, parenchyma cells are not impurities requiring removal but can serve as functional structural reinforcement for achieving extreme densification. Addressing the machinability challenge, this study reveals that precise control of the 8-h enzymatic hydrolysis window establishes an optimal structural trade-off between retaining long fibers as skeletal support and generating fine fragments with filling and wrapping effects. This theoretically elucidates how to achieve precision machining of biomass materials through microstructural regulation.

While scholars (*e.g.*, Adier *et al.* 2023; Kartono Kurniawan *et al.* 2023) have extensively discussed the sustainable development of the bamboo industry, most discussions are limited to building materials or low-end daily necessities. This study demonstrates a feasible closed-loop manufacturing pathway from bamboo waste to binderless brush handles, transforming bamboo processing residues, which are typically incinerated, into high-value cultural heritage artifacts.

Limitations and Future Directions

This study still has certain limitations. First, the 8-h pretreatment time is relatively long, which increases production energy consumption to some extent. Future research could explore multi-enzyme synergistic systems (such as introducing hemicellulase or pectinase) or auxiliary physical fields (such as ultrasonic assistance) to shorten treatment time and improve production efficiency. Second, this study only validated Moso bamboo as a raw material. Considering the chemical composition differences among bamboo species, it is necessary to investigate the universality of this technology for other bamboo species and potentially other lignocellulosic residues with similar anatomical structures to establish a more comprehensive process database. Finally, although simulation tests demonstrated excellent ink resistance, long-term tracking studies are required to assess the aging behavior of the material under extreme dry-wet cycling environments.

CONCLUSIONS

1. High-consistency mechano-enzymatic (HCME) pretreatment combined with a closed-mold cylindrical molding process successfully transformed bamboo waste into high-performance binderless composite brush handles.
2. The synergistic micro-filler effect and lignin *in situ* activation markedly reduced porosity to 3.27% and achieved an ultra-high density of 1.27 g/cm³.
3. Lignin redistribution during the process constructed a natural hydrophobic barrier, which effectively limited thickness swelling to below 5.1% and provided robust ink resistance.
4. The formation of a micro-brittle network ensured superior precision machinability during lathe turning, yielding a mirror-like surface finish without fiber tearing.
5. This technical pathway establishes a new material paradigm for the high-value utilization of biomass residues and the green transformation of traditional cultural heritage carriers.

Data Availability

The data used to support the findings of this study are included within the article.

Conflict of Interest

The authors declare that there is no conflict of interest regarding the publication of this paper.

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APPENDIX

S1. Supplementary Materials and Methods

Separation of bamboo fibers and parenchyma cells

To analyze the raw material composition, bamboo powders were soaked in deionized water, stirred thoroughly for 3 to 4 min, and allowed to stand for 10 min. Utilizing the density difference between fibers and parenchyma cells, the denser fibers settled to the bottom, while the less dense parenchyma cells floated to the surface. The separated fractions were collected and dried in an oven at 60 °C to constant weight.

Determination of glucose in BP

To verify the enzymatic hydrolysis effect during HCME, 5 g of dry BP was mixed with 50 mL of ultrapure water. The mixture was filtered, and the glucose content of the supernatant was measured using an ion chromatography system (Metrohm 850, Switzerland) equipped with a Dionex CarboPac PA-200 analytical column (3mm × 250mm).

Statistical analysis

The statistical significance of differences in water resistance and mechanical performance among BSBC samples was evaluated using SPSS software. One-way analysis of variance (ANOVA) followed by Duncan's multiple range test was performed to determine significant differences at $p < 0.05$.

S2. Supplementary Figures

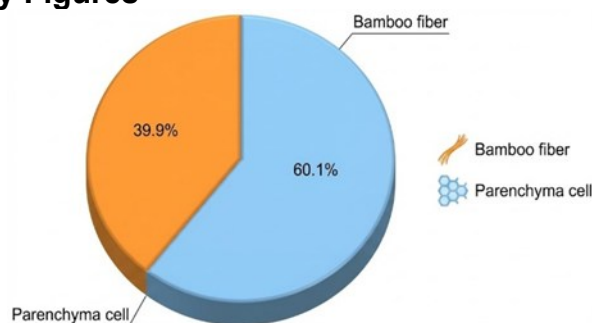


Fig. S1. The proportion of bamboo fibers and parenchyma cells in BP

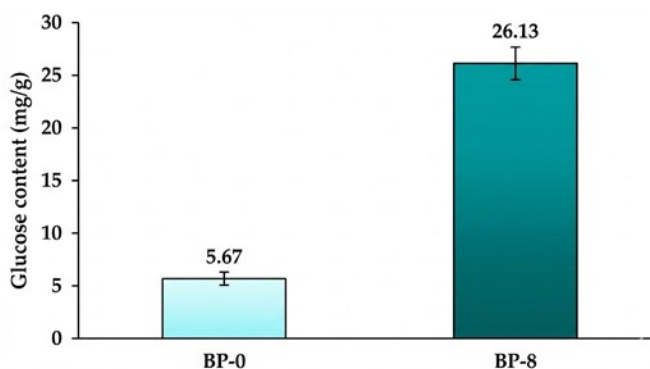


Fig. S2. Glucose content in the filtering liquids of BP-0 and BP-8

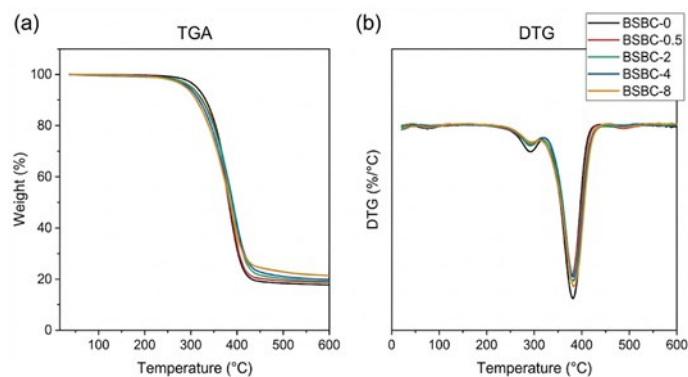


Fig. S3. Thermal stability of the resultant BSBC

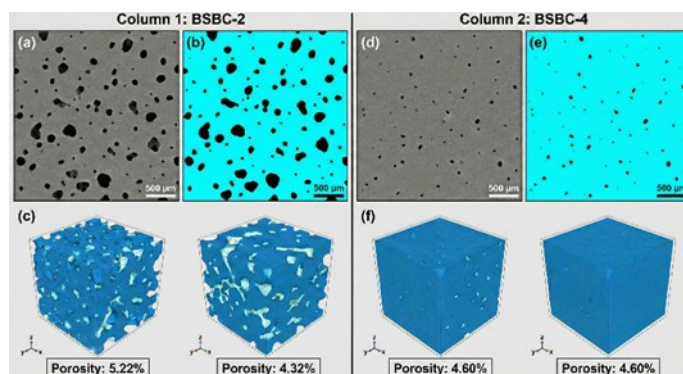


Fig. S4. Pore characteristics of intermediate BSBC samples (BSBC-2 and BSBC-4)

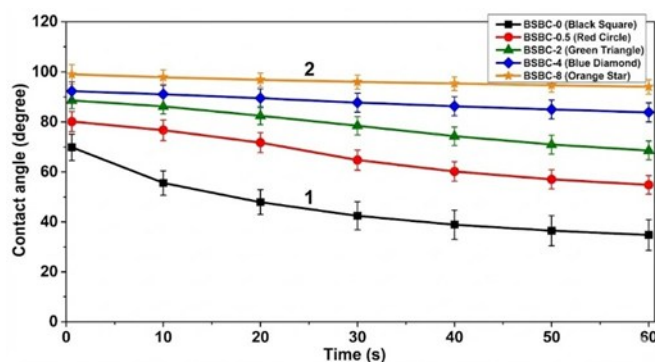


Fig. S5. Dynamic water contact angles of BSBC changes with time

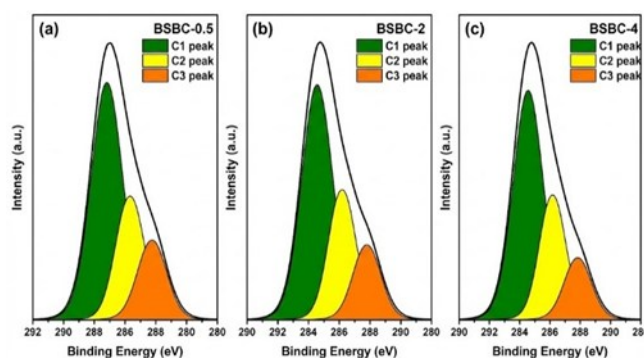


Fig. S6. The C 1s XPS spectra of intermediate BSBC samples

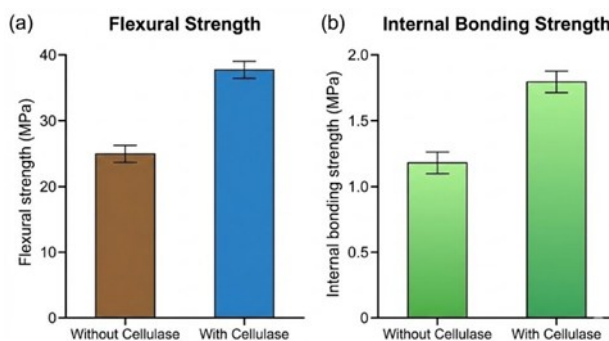


Fig. S7. Effect of cellulase on mechanical properties of BSBC-8

S3. Supplementary Tables

Table S1. Multiple Comparison and Significance Analysis between Physical and Mechanical Properties of BSBC

Group Comparisons	WA (p-value)	TS (p-value)	Flexural Strength (p-value)	Flexural Modulus (p-value)	IB Strength (p-value)
BSBC-0 vs. BSBC-0.5	0.015*	<0.001**	<0.001**	0.076	0.129
BSBC-0 vs. BSBC-2	<0.001**	<0.001**	<0.001**	0.310	0.082
BSBC-0 vs. BSBC-4	<0.001**	<0.001**	<0.001**	0.009**	0.048*
BSBC-0 vs. BSBC-8	<0.001**	<0.001**	<0.001**	<0.001**	0.003**
BSBC-0.5 vs. BSBC-2	0.002**	<0.001**	0.964	0.418	0.805
BSBC-0.5 vs. BSBC-4	<0.001**	<0.001**	0.268	0.310	0.603
BSBC-0.5 vs. BSBC-8	<0.001**	<0.001**	<0.001**	0.001**	0.0903
BSBC-2 vs. BSBC-4	0.007**	0.015*	0.287	0.076	0.784
BSBC-2 vs. BSBC-8	<0.001**	<0.001**	<0.001**	<0.001**	0.146
BSBC-4 vs. BSBC-8	0.033**	0.188	<0.001**	0.014*	0.231

Note: WA: Water absorption rate; TS: Thickness swelling rate; IB: Internal bonding strength. *p < 0.05 (Significant); **p < 0.01 (Very significant).