Eng. Sci., 2025, **37**, 1815

Engineered Science

DOI: https://dx.doi.org/10.30919/es1815

Synergistic Enhancement of Bamboo Composites Via Nano-Aluminum Oxide and Silicon Dioxide Hybrid Modification for Cooling Tower Applications

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Abstract

This article presents a preparation method for modified bamboo and explores its potential application as filler material in cooling towers. The study employs a dual-nanoparticle synergistic modification approach combined with silane-based interfacial optimization, utilizing nano-aluminum oxide and silicon dioxide modifiers to enhance the properties of bamboo. Molecular dynamics simulations reveal the reaction mechanisms between the silane coupling agent and the surfaces of the nanoparticles, which significantly improve the interfacial bonding between the bamboo and the modifiers. Scanning electron microscopy and thermal performance tests show that the modified bamboo exhibits notable improvements in thermal conductivity, thermal diffusivity, and specific heat capacity. These enhancements are attributed to the incorporation of aluminum oxide and silicon dioxide, which effectively boost the bamboo's thermal conductivity and stability. Additionally, the surface hydrophilicity of the modified bamboo increases, resulting in a reduced contact angle and stronger hydration behavior. The modified bamboo can serve as a sustainable alternative to polyvinyl chloride fillers, reducing environmental impact and promoting resource sustainability. The bamboo modification strategy proposed in this work not only improves thermal transport properties but also offers a viable solution for replacing conventional polyvinyl chloride filler materials, delivering significant environmental and economic benefits.

Keywords: Cooling tower; Modified bamboo; Thermal conductivity; Molecular dynamics.
Received: 26 June 2025; Revised: 04 August 2025; Accepted: 21 September 2025
Article type: Research article.

1. Introduction

Cooling towers are energy-efficient cooling systems with strong heat dissipation capabilities, widely employed in industrial sectors such as power generation, steel production, and chemical processing.[1] With the rapid advancement of global industrialization, optimizing the performance of cooling towers-key heat exchange components in energy-intensive industries like power and chemicals-has become critical for energy conservation and emission reduction. Packing materials, essential to heat and mass transfer in cooling towers, must possess high thermal conductivity, corrosion resistance, low weight, and environmental friendliness. Traditional fillers such as polyvinyl chloride (PVC) and metals are commonly used; however, PVC can release harmful gases and chemicals over prolonged use, posing risks to human health and the environment. Metal materials, on the other hand, are costly and prone to scaling and chemical corrosion.[2] Consequently, developing eco-friendly alternatives, especially from natural and renewable sources, has become a global research priority.

Bamboo, as a natural, renewable material with excellent mechanical properties, has emerged as a promising green alternative due to its low density, high strength, and good toughness.[3-5] Compared to the widely used fillers, bamboo is more affordable and has higher availability. (4) However, the intrinsic structure of bamboo limits its mechanical and thermal performance under extreme environments or heavy loads.[6,7] Recent advances in nanomaterial-based modification techniques offer new pathways for enhancing bamboo's functional properties. Incorporating nanoparticles such as SiO_2 and Al_2O_3 into bamboo's hierarchical porous structure can significantly improve its mechanical strength and thermal behavior.[8,9] Nonetheless, studies specifically addressing thermal conductivity enhancement remain in their early stages. In the field of thermal property enhancement, nano-SiO_2 and

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Al_2O_3 are widely used in bio-based composites due to their high thermal conductivity and chemical stability.[10-12] Yet, the interfacial bonding mechanisms between bamboo and nanoparticles remain poorly understood, and existing research often focuses on single-component modifications, lacking systematic analysis of synergistic effects from multi-component systems.

Nano-SiO_2 can form a dense hydrophobic coating on bamboo surfaces via the sol-gel method, When these hydrophobic nanoparticles are evenly distributed on the surface or shallow layer of bamboo, the contact angle can be significantly increased, endowing bamboo with excellent hydrophobicity and even super hydrophobicity, making it less susceptible to water wetting and penetration, further enhancing its waterproof, anti fouling, and anti-corrosion properties.(问题 6) while Al_2O_3 nanoparticles can be embedded into the bamboo matrix through vacuum impregnation to construct efficient thermal pathways.[13] Their combined use can overcome challenges associated with poor interfacial compatibility and mechanical strength typically seen in single-material modifications.[14,15] The interfacial bonding strength between nanoparticles and bamboo fibers is a key determinant of modification performance. Silane coupling agents such as KH-550 can modify hydroxyl groups on bamboo surfaces, promoting chemical bonding with nanoparticles and significantly enhancing thermal conduction networks.[16]

This study aims to modify bamboo using a synergistic system comprising nano-SiO_2, Al_2O_3, polyethylene glycol (PEG), and the silane coupling agent (KH550). Through molecular dynamics (MD) simulations, Fourier-transform infrared spectroscopy (FTIR), and scanning electron microscopy (SEM), the study investigates the molecular interaction mechanisms during modification and their effects on the physicochemical properties of bamboo. The findings offer a new strategy for employing bamboo as a high-performance, environmentally friendly material in cooling towers and lay the groundwork for promoting bamboo's sustainable development in architecture and engineering applications.

2. Materials and methods

2.1 Samples preparation

Moso bamboo (*Phyllostachys edulis* (Carrière) J.Houz), harvested from Taiping Forest Farm in Huangshan, Anhui Province, with an age of 4 years, had a culm diameter ranging from 100 to 120 mm and wall thickness approximately 7 to 12 mm. Bamboo sections were cut at a height of approximately 1.5 meters from the ground. Samples of dimensions 20 mm (length) × 20 mm (transverse) × wall thickness (radius) were prepared and dried to a constant weight for subsequent use.

2.2 Preparation of the modification system

First, an appropriate amount of deionized water was measured and placed into a clean beaker as the solvent. Polyethylene glycol (PEG, 10 vol%) was then added to increase the solution's viscosity and improve its flowability and penetration. Subsequently, 10 vol% of silane coupling agent (KH550) was introduced to enhance the interfacial bonding between the nanoparticles and the modifier. Precisely weighed nano-silica (SiO_2) and nano-alumina (Al_2O_3) particles were added to formulate gradient modification solutions with concentrations of 1 wt%, 3 wt%, 5 wt%, and 10 wt%. These solutions were stirred using a magnetic stirrer to ensure uniform dispersion of nanoparticles and prevent agglomeration. Continuous mechanical stirring was applied to maintain homogeneity and avoid precipitation or air bubble formation. Finally, the solution was filtered through a membrane to remove undispersed impurities, ensuring uniformity and stability.

Analytical-grade nano-SiO_2 (CAS 60676-86-0), nano-Al_2O_3 (CAS 1344-28-2), silane coupling agent KH-550 (CAS 919-30-2), and polyethylene glycol (CAS 25322-68-3) were all purchased from Macklin Biochemical Co., Ltd. and used directly without further purification.

2.3 Bamboo modification procedure

The bamboo modification process was carried out using a vacuum-pressure impregnation method. First, the bamboo specimens were placed in a pressure chamber and subjected to vacuum degassing to remove internal air, with the vacuum maintained for 30 minutes. The prepared modification solution was then introduced to ensure full contact with the samples. The pressure was gradually increased to 1.0 MPa and maintained for 30 minutes to allow deep penetration of the solution into the bamboo structure and ensure effective modification. After treatment, the bamboo samples were removed and dried in an oven at a constant temperature until reaching constant weight. Samples with different concentrations of modifier were processed in separate batches under identical treatment conditions (Fig. S1).

2.4 Characterization

Changes in microstructure were characterized using scanning electron microscopy (SEM, ZEISS Sigma 500, Germany). Chemical structure variations were analyzed by Fourier-transform infrared spectroscopy (FTIR, Thermo Fisher, USA), X-ray photoelectron spectroscopy (XPS, Thermo Fisher K-Alpha X, USA), solid-state nuclear magnetic resonance (NMR, Bruker AVANCE 400, Germany), Raman spectroscopy (DXR2xi, Thermo Fisher, USA), and X-ray diffraction (XRD, Rigaku SmartLab, Japan). The distribution of modification particles was evaluated using elemental analysis (EA, Elementar Vario EL Cube, Germany) and inductively coupled plasma spectroscopy (ICP, Agilent 4800). Thermal conductivity was measured via the transient plane source (TPS) method using a Hot Disk TPS2500S. Thermal stability was assessed using thermogravimetric analysis (TGA, TA SDT650, USA) and differential scanning calorimetry (DSC, NETZSCH DSC 214 Polyma, Germany). The reaction mechanisms in the modification solution were modeled using the Vienna Ab initio Simulation Package (VASP) with the GGA-PBE functional, as

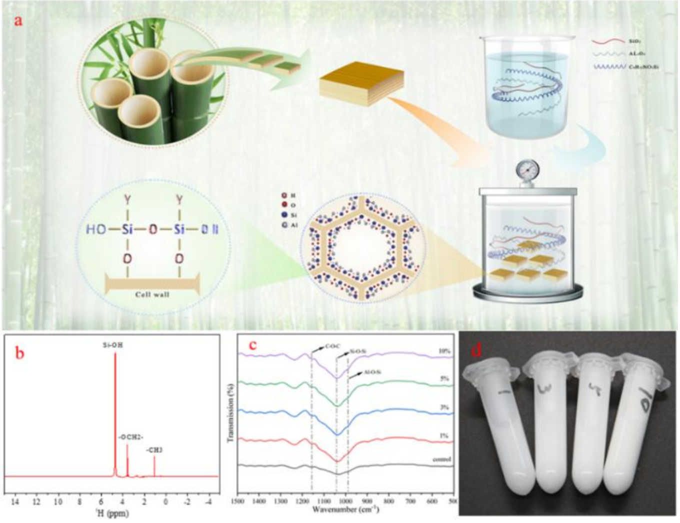

Fig. 1: a) Process of modified bamboo. b) NMR spectrum. c) FTIR. d) Modified solution.

detailed in Section Fig. S3-Fig. S6.

3. Results and discussion

3.1 Preparation process of modified bamboo

The preparation process of the modified bamboo is illustrated in Fig. 1a. The surfaces of nano-SiO_2 and Al_2O_3 particles are rich in free hydroxyl groups, which exhibit strong surface hydrophilicity. By introducing specific chemical agents, these hydroxyls can undergo chemical reactions and physical interactions, enhancing the surface activity of the particles. This modification improves their physicochemical and optical properties, which in turn enhances the performance of the treated bamboo through both physical filling and chemical bonding mechanisms.[17,18] A comprehensive analysis was conducted on both the modification solution and the treated bamboo samples. As shown in Fig. 1b, the hydrogen nuclear magnetic resonance (^{1}H NMR) spectrum of the modification solution displays characteristic peaks corresponding to Si-OH, indicating the presence of silanol groups formed by hydrolysis. Peaks associated with -OCH_2- and -CH_3 are attributed to the ethoxy groups in the silane coupling agent, confirming the occurrence of hydrolysis. The observed decrease in peak intensity further implies that condensation reactions took place during the process.

To validate the formation of condensation products, Fourier-transform infrared (FTIR) spectroscopy was employed (Fig. 1c). Vibration peaks near 980 cm^{-1} and 1040 cm^{-1}, corresponding to Si-O-Al and Si-O-Si bonds, respectively, confirm that the hydroxyl groups on the nanoparticle surfaces reacted with hydrolyzable groups of the coupling agent to form covalent bonds via condensation. Furthermore, the prepared modification solution appeared milky-white (Fig. 1d), and the addition of polyethylene glycol effectively mitigated nanoparticle sedimentation. These results underscore the need for molecular-level investigation to reveal the complex transformations and mechanisms occurring within the modification system.

3.2 Molecular dynamics simulation

To elucidate the formation mechanism of the modification solution, molecular dynamics (MD) simulations were conducted to investigate the molecular interactions and reaction processes within the solution. Initially, the silane groups in the silane coupling agent (KH550) undergo hydrolysis in aqueous solution, forming silanol (Si-OH) groups. This reaction process was monitored through MD simulations, which revealed the interaction between water molecules and silane molecules. Water molecules interact with the silicon atoms of the silane molecules via hydrogen bonds, promoting the cleavage of the Si-O bond and the formation of Si-OH groups. The changes in reaction energy and activation energy for this process were obtained through MD simulation (Fig. 2a).

Subsequently, the silanol groups undergo a condensation reaction with the hydroxyl groups on the surfaces of nano-SiO_2 and Al_2O_3, resulting in the formation of Si-O-Al or Si-O-Si bonds. Fig. 2b and 2c show the interactions between the silanol groups and the nanoparticle surfaces. The energy changes during the reaction also indicate the formation of surface hydrophilicity, hydrogen bonds, and covalent bonds. The study shows that the surface of the nanoparticles contains a large number of hydroxyl groups, which react with the silanol groups to form stable covalent bonds.[19]

Furthermore, after the reaction between the silanol groups and the nanoparticles, the formed Si-O-Al and Si-O-Si bonds facilitate further cross-linking reactions between the nanoparticles (Fig. 2d). During this process, the interactions between nanoparticles are mainly driven by van der Waals forces, hydrogen bonds, and electrostatic forces, ultimately forming stable nanoparticle aggregates.[20-22] These nanoparticle aggregates can effectively fill the void structures in bamboo, while the nanoparticles may also interact with bamboo fibers through condensation reactions, enhancing the thermal conductivity of the bamboo. Therefore, further investigation of this process is necessary to better understand the underlying mechanism.

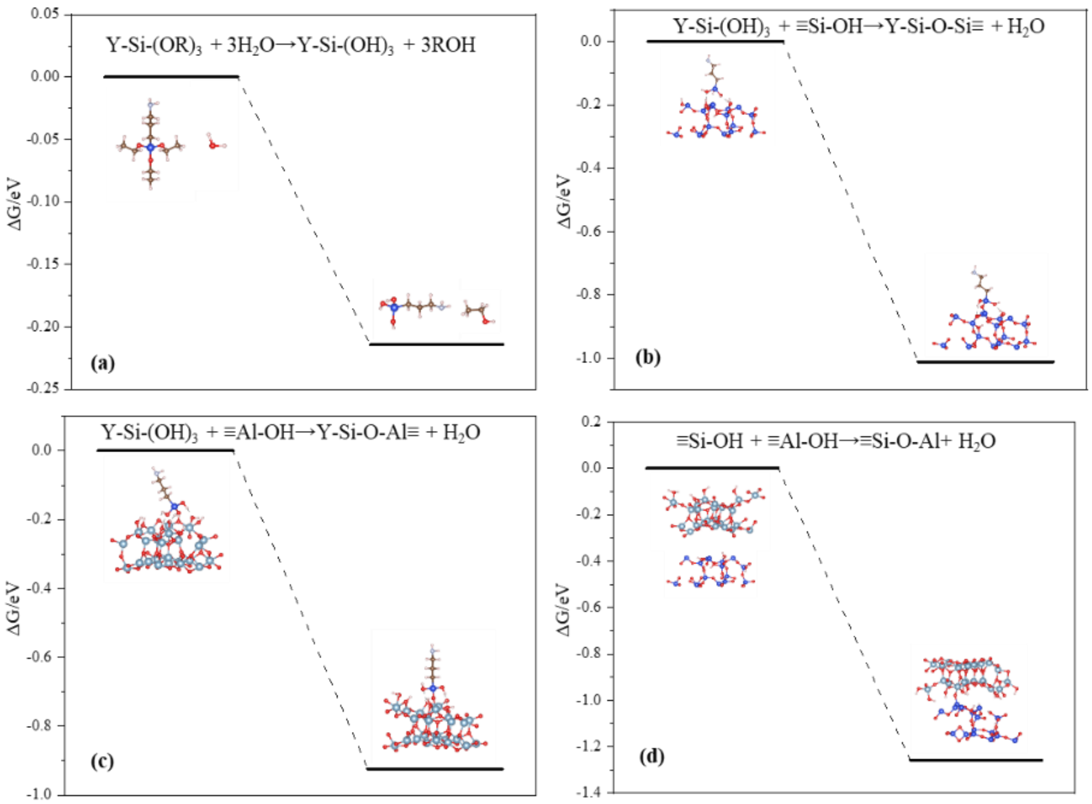

Fig. 2: a) Hydrolysis of silane coupling agent. b) Condensation reaction of silanol and nano silica. c) Condensation reaction of silanol and nano alumina. d) Hydroxyl condensation reaction on the surface of nanoparticles.

3.3 Performance Analysis

To verify the performance changes of the modified samples, both block and powder samples were tested (Fig. 1). The SEM images in Fig. 3a show that the cell surfaces are coated with a layer of light gray nanoparticles. Some of these nanoparticles aggregate through interactions (Fig. 3b), and further magnification reveals nanoparticle clusters (Fig. 3c), indicating that the modification agents have successfully adhered to the sample surface.

In the thermal conductivity tests of the samples, the thermal conductivity (Fig. 3d), thermal diffusivity (Fig. 3e), and specific heat (Fig. 3f) were investigated. It can be seen that as the concentration of the modification agent increases, the thermal conductivity, thermal diffusivity, and specific heat all show a gradually increasing trend. This is mainly due to the fact that both nano-SiO$_2$ and Al$_2$O$_3$ are materials with strong thermal conductivity. The high surface area and small size of the nanoparticles enable them to effectively transfer heat through the thermal conduction pathways in the material. The introduction of nanoparticles helps improve the thermal conductivity of the material, especially in solids, where nanoparticles can significantly increase the heat transfer channels, thereby enhancing the overall thermal conductivity of the sample.[23,24] As a good thermal conductor, Al$_2$O$_3$ particles have a thermal conductivity much higher than that of bamboo, thus enhancing the thermal conductivity of bamboo through the composite nano-Al$_2$O$_3$ particles. Moreover, the silane coupling agent (KH550) can improve the interfacial bonding between the matrix and the nanoparticles, and through reactions, it bonds with the cell surface of bamboo, enhancing the material's stability and thermal conductivity.[25,26] The introduction of these two types of nanoparticles alters the microstructure of the bamboo, allowing the sample to absorb more thermal energy over a wider temperature range. In particular, the silane coupling agent forms a chemical bond between bamboo fibers and nanoparticles, thereby improving the heat capacity of the sample during the heat absorption process.[27] Additionally, the dispersion and self-assembly properties of the nanoparticles also play a critical role in thermal conductivity. The self-assembly process of the two types of nanomaterials in the sample can form a continuous thermal conduction network, and good dispersion allows heat to be efficiently transmitted through this nanoparticle network.[28]

In the TG spectrum (Fig. 3g), it can be observed that with the increase in the modification agent concentration, the thermal stability increases. Since the temperature range for practical applications in this study is below 90°C, the reasons for the improvement in thermal stability within this temperature range were analyzed (Fig. 3h). Both nano-SiO$_2$ and Al$_2$O$_3$ have high thermal decomposition temperatures. As inorganic fillers dispersed in the bamboo structure, they form a dense network structure that enhances the bamboo's resistance to thermal decomposition at high temperatures, acting as a physical barrier during the pyrolysis process, thus delaying the onset of the thermal decomposition reaction.[29]

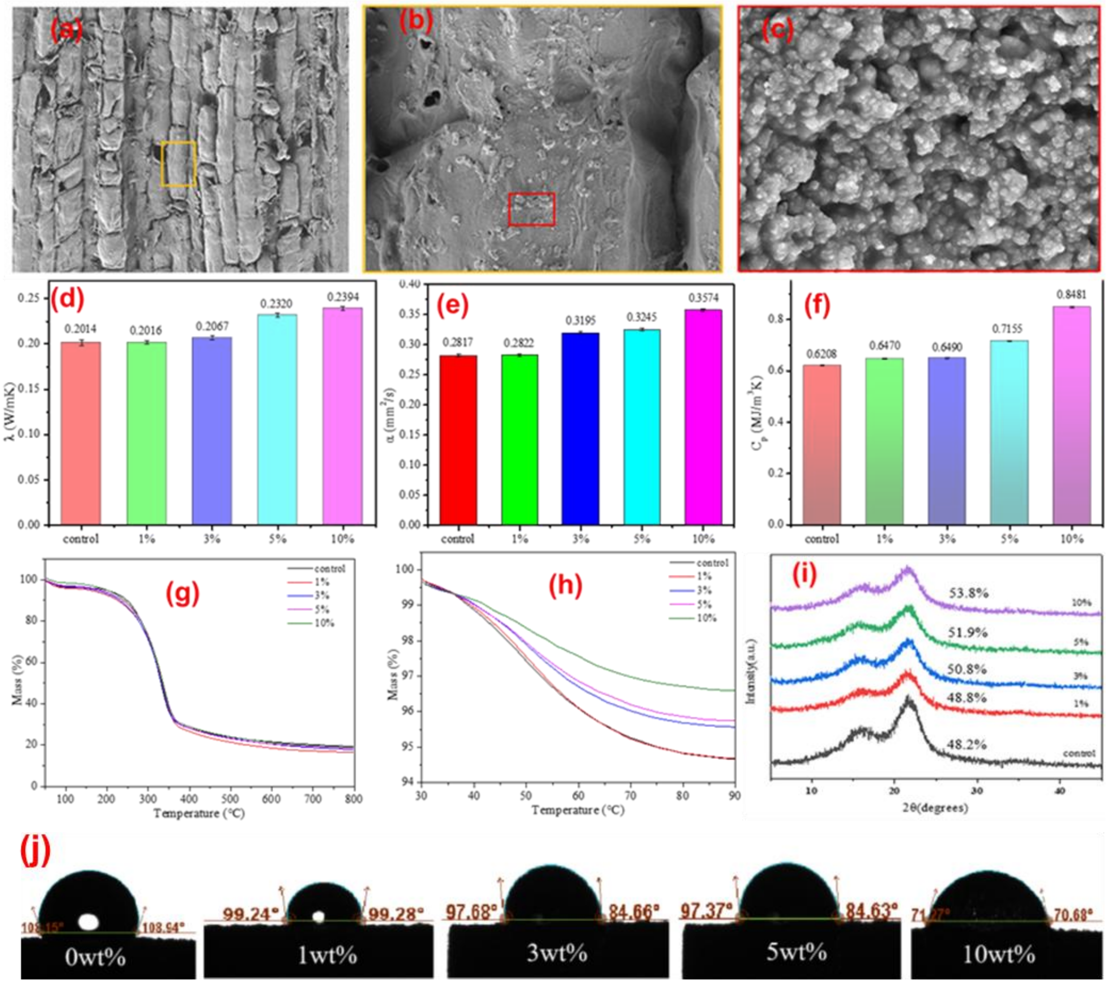

Fig. 3: a) SEM. b) Amplified image. c) Nanoparticle aggregates. d) Thermal conductivity. e) Thermal diffusivity. f) Specific heat. g) TG analysis. h) 0-90℃ TG analysis. i) XRD. j) Contact angle analysis.

In XRD analysis, the crystallinity of the samples showed a gradually increasing trend (Fig. 3i). This is primarily because nano-SiO_2 and Al_2O_3, as inorganic nucleating agents, have a good nucleating effect. They may fill or replace some of the disordered amorphous regions, promoting the ordered arrangement of the cellulose chain's non-crystalline regions in bamboo, thus improving the crystallinity of the sample.[30] The hydrophobicity of the modified samples gradually decreased (Fig. 3j). This is because the polar groups such as -Si-OH and $-NH_2$, generated after the hydrolysis of KH550, are exposed on the material surface. During the reaction, a large number of hydrogen bonds are formed, significantly increasing the surface energy and enhancing the affinity for water molecules, which leads to a reduction in the contact angle. Furthermore, the nanoparticles alter the surface structure of the bamboo at the microscopic scale, reducing surface roughness and making water droplets spread more easily, thereby weakening the hydrophobicity.[9]

3.4 Mechanism analysis

The SEM images showed that the vascular bundles of bamboo were deformed after modification. The SEM images indicate that the modified nanoparticles are uniformly distributed on the surface of the bamboo cells (Fig. 4a), demonstrating that the composite system exhibits good dispersion and interface compatibility. This clear distribution at the microscopic scale not only visually confirms the effectiveness of the modification treatment but also provides a basis for the performance enhancements observed previously, such as improved thermal conductivity and thermal stability. XPS analysis reveals a decreasing trend in the contents of C and O, alongside an increase in the amounts of Si and Al, indicating successful impregnation of nanoparticles into the samples (Fig. 4b). To further analyze the bonding mechanisms of the nanoparticles with the cell wall surfaces, peak deconvolution of the XPS spectra was performed (Fig. 2). It is evident that diffraction peaks appear in the 103-104 eV range, signifying the formation of Si-O-Si bonds; additionally, a diffraction peak in the 102-eV region corresponds to the Si-O-C vibrational mode.[31] Furthermore, a diffraction peak at 76 eV suggests that hydroxyl groups on the surface of Al_2O_3 interact with surface hydroxyls of the bamboo to form Al-O-C vibrational modes. Therefore, it can be inferred that the nanoparticles form chemical bonds with the bamboo surface via coupling agents, enhancing interfacial bonding strength.

FTIR analysis (Fig. 4c) identifies stretching vibrational

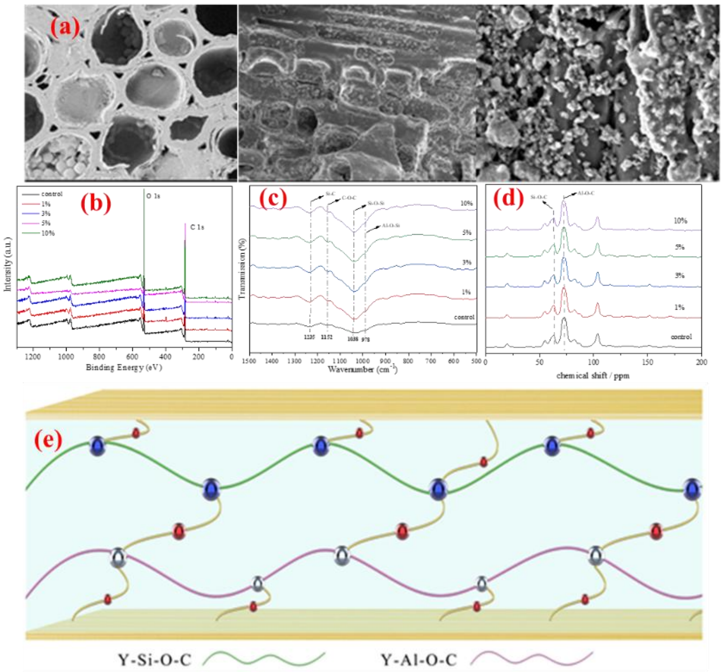

Fig. 4: a) SEM mage. b) XPS. c) FTIR. d) Solid-state NMR carbon spectroscopy. e) Modification mechanism diagram.

peaks for Si-O-Si and Al-O-Si at 1038 cm^{-1} and 978 cm^{-1}, respectively.[32] The appearance of a C-O-C bond at 1152 cm^{-1} indicates that during the modification process, nanoparticles and coupling agents penetrate into the bamboo, exposing the originally encapsulated polysaccharide chains and thereby strengthening the infrared signal of the C-O-C bond. Concurrently, the amine groups within the coupling agent undergo condensation reactions with surface hydroxyls, resulting in new ether bonds that further amplify the peaks.

Solid-state NMR carbon spectroscopy (Fig. 4d) shows an increase in peak intensity at 63 ppm and 72 ppm, suggesting that silanol reacts with hydroxyls in the bamboo through condensation, forming Si-O-C bonds, which thereby enhance the bonding strength between the bamboo and silane coupling agents. Additionally, the hydroxyls on the surface of aluminum oxide (Al_2O_3) may undergo condensation reactions with the hydroxyls on the bamboo surface to form Al-O-C bonds, indicating that the nanoparticle-coupling agent complex is covalently integrated into the supramolecular structure of the bamboo. Through model analysis (Fig. 4e), it can be concluded that the silane coupling agent/nanoparticle modification system binds to bamboo through a combination of chemical bonds and interfacial synergistic effects. A substantial number of hydroxyl groups in the modification system form hydrogen bonds with the hydroxyls on the bamboo surface; this high hydrophilicity allows deep penetration into the molecular structure of the bamboo, resulting in the formation of an organic-inorganic hybrid system. SiO_2 and Al_2O_3 react chemically with silanes via their surface-active hydroxyl groups, where the Si-O-Si and Al-O-Si bonds enhance rigidity, while the C-O-C bonds contribute toughness, achieving a "stiffness-toughness" modification effect.

This modification system, through the crosslinking action of silanes, results in the formation of stronger chemical bond connections on the surface of the bamboo, significantly enhancing its physicochemical properties.

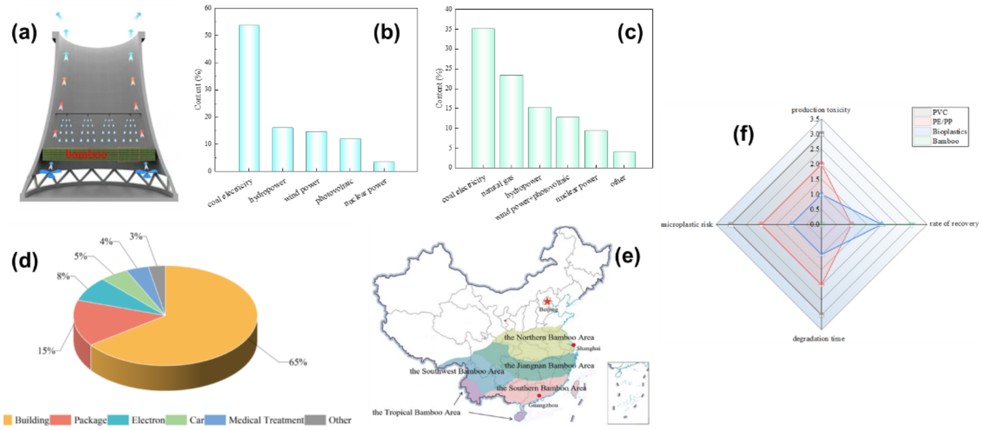

Fig. 5: a) Cooling tower. b) Types of domestic power plants. c) International power plant types. d) Main application areas of PVC. e) Distribution of bamboo resources. f) Comparison of hazards between bamboo and plastic.

3.5 Application potential analysis

Cooling towers are an essential part of the thermal cycle in power plants (Fig. 5a). The hot water (approximately 85°C) generated by the power generation system is pumped through pipes to the distribution system at the top of the cooling tower, where it is dispersed into fine droplets via nozzles or spray devices. This hot water then comes into contact with the cooling fill, while cool air enters from the bottom of the tower and rises, creating convection that effectively reduces the water temperature.

As shown in Fig. 5b and 5c, power plants are widely distributed both domestically and internationally, and can be categorized by energy type into coal, natural gas, hydroelectric, nuclear, wind, photovoltaic, and other power plants, with coal power plants being the most common type.[33,34] Among these power plants, PVC is the most commonly used cooling fill material. Due to its excellent corrosion resistance, PVC is widely applied across various fields (Fig. 5d). However, with increasing awareness of environmental protection, the environmental hazards posed by PVC during its use have drawn growing attention (Fig. 5f). Notably, issues such as its non-degradability and the potential release of toxic substances after prolonged use have emerged. Over time, aging PVC materials may release chlorine gas and other harmful substances in the power plant environment, leading to pollution of air and water bodies. Furthermore, the release of toxic hydrochloric acid gas and dioxins can pose risks to workers and surrounding ecosystems when there is prolonged exposure.[35,36]

China has a rich bamboo flora comprising 39 genera and over 800 species, earning it the title of the "Bamboo Kingdom".[37,38] Bamboo resources are primarily concentrated in the warm and humid southern regions, especially in provinces south of the Yangtze River (Fig. 5e). In the context of global energy transitions and increasingly stringent environmental protection requirements, bamboo has gained considerable attention as a sustainable material. Thanks to its natural, renewable, environmentally friendly, and durable properties, bamboo is an ideal substitute for PVC. Through modification treatments, the application of bamboo in cooling systems can not only reduce environmental pollution but also achieve sustainable resource utilization. With ongoing advancements in bamboo technology and the expansion of its application fields, the model of using bamboo to replace PVC is expected to see wider adoption in power plants.

4. Conclusion

In this study, bamboo was modified by vacuum impregnation as a composite solution of nano SiO_2/Al_2O_3, polyethylene glycol and KH550 coupling agent to explore its potential as an alternative to PVC filler for cooling towers. The results showed that the chemical bonds of silicon-oxygen-aluminum/silicon-oxygen-silicon were formed with the surface of the bamboo, and the stable covalent bond interface verified by molecular dynamics simulation increased the thermal conductivity, thermal diffusion coefficient, and initial thermal decomposition temperature of the modified bamboo. Microstructure analysis showed that the nanoparticles filled the pores of bamboo fibers and reduced the surface roughness, which reduced the contact angle of water droplets and realized the transformation from hydrophobic to hydrophilic. The bio-based material combines enhanced thermal stability with degradable properties, providing a sustainable solution for cooling tower fillers in thermal power systems and is expected to reduce plastic pollution when applied.

Acknowledgments

This work was supported by the National Natural Science Foundation of China (32101601) and Strategic Research and Consulting Project of the Chinese Academy of Engineering (2024-DFZD-64).

Conflict of Interest

The authors have no conflict of interest.

Supporting Information

Applicable.

CRediT Statement

Chunyu Yang and **Benhua Fei:** Conceived the concept. **Chunyu Yang**, **Yong Liang**, **Yuting Mao**, and **Liwen Yang:** Conducted all experiments and Data analysis. **Chunyu Yang**, **Lisheng Chen**, and **Huangfei Lv:** Wrote the initial draft, Discussed the data, and revised the manuscript. **Bin Xu**, **Huangfei Lv**, and **Yong Liang:** Supervised the work. All authors contributed to editing the manuscript.

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