

Supramolecular network-engineered transparent conductive cellulose/gelatin bioplastics for efficient Joule heating and electromagnetic interference shielding

Wen Gong^{1,2}, Jianyu Zhou^{2,3}, Yuchen Tian², Qi Zhang⁴, Puyan Huang², Run Gan⁵, Xiang He¹ (✉), Miao Miao², Xin Feng^{1,2} (✉)

¹ Department of Chemistry, College of Sciences, Shanghai University, Shanghai 200444, China

² Research Center of Nanoscience and Technology, College of Sciences, Shanghai University, Shanghai 200444, China

³ School of Materials Sciences and Engineering, Tongji University, Shanghai 201804, China

⁴ Department of Electronic Information Materials, School of Materials Science and Engineering, Shanghai University, Shanghai 200444, China

⁵ College of Food Science and Technology, Shanghai Ocean University, Shanghai 201306, China

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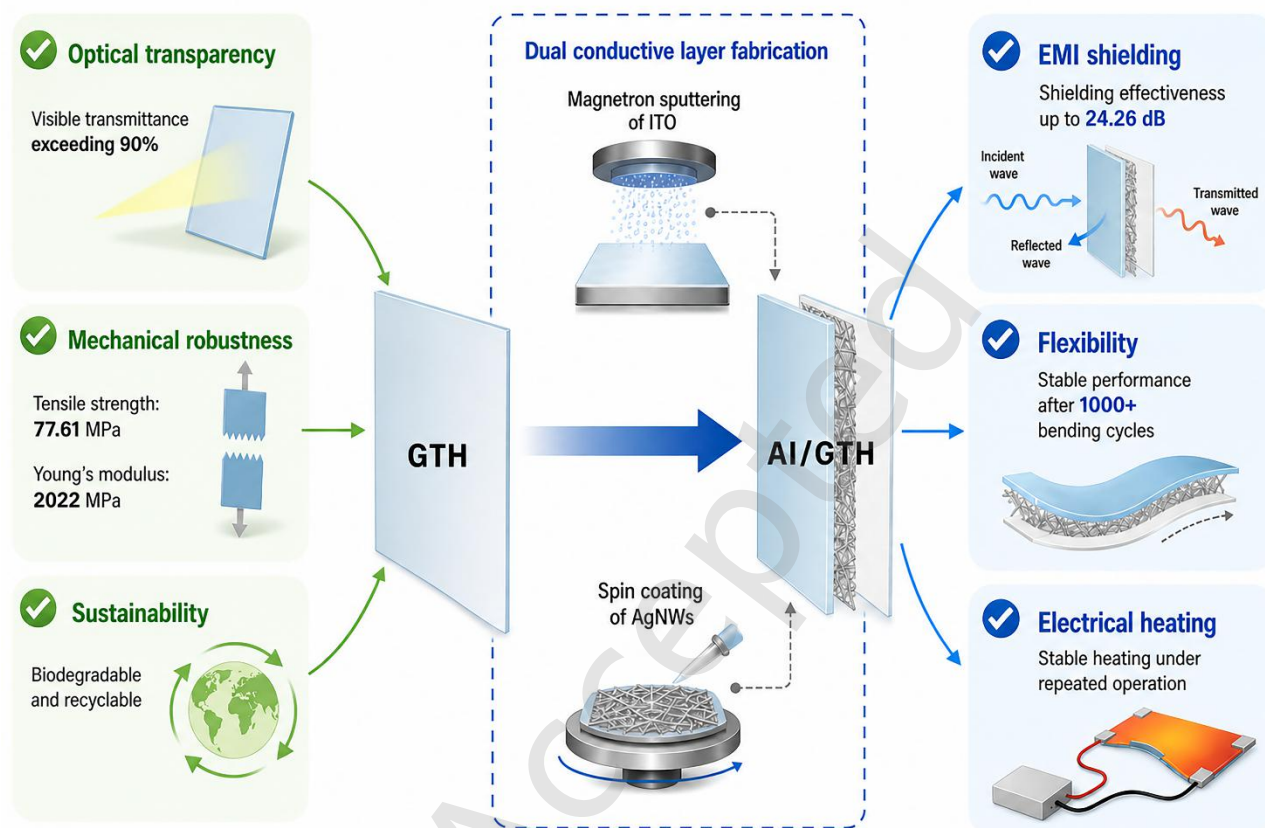
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A transparent bioplastic with excellent mechanical flexibility was fabricated via self-assembly of gelatin, TEMPO-oxidized cellulose, and hyaluronic acid through multiple hydrogen bonding. After spin-coating AgNWs and depositing ITO via magnetron sputtering, the resulting AI/GTH film integrates excellent EMI shielding and Joule heating performance, while maintaining stable functionality under repeated bending and cyclic heating, highlighting its potential for sustainable flexible electronics and multifunctional devices.

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¹ Department of Chemistry, College of Sciences, Shanghai University, Shanghai 200444, China

² Research Center of Nanoscience and Technology, College of Sciences, Shanghai University, Shanghai 200444, China

³ School of Materials Sciences and Engineering, Tongji University, Shanghai 201804, PR China

⁴ Department of Electronic Information Materials, School of Materials Science and Engineering, Shanghai University, Shanghai 200444, China

⁵ College of Food Science and Technology, Shanghai Ocean University, Shanghai 201306, China

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 Address correspondence to Xiang He, hxiang@shu.edu.cn; Xin Feng, fengxin@shu.edu.cn

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ABSTRACT: Petroleum-based plastics are indispensable in modern society, yet their non-degradable nature and toxic byproducts have led to severe environmental pollution and ecological threats. The development of bioplastics that combine high optical transparency, excellent tensile strength, and recyclability remains a significant challenge on the path toward sustainability. To address this challenge, a flexible, transparent bioplastic with exceptional mechanical performance, exhibiting a tensile strength of 77.61 MPa and a Young's modulus of 2022.02 MPa, was prepared via a straightforward self-assembly strategy using gelatin, TEMPO-oxidized cellulose nanofibril and hyaluronic acid, and as raw materials, based on a supramolecular network formed through multiple hydrogen bonding. Based on this, a dense double conductive layer was fabricated on the bioplastic surface via spin-coating of silver nanowires (AgNWs) and subsequent magnetron sputtering of indium tin oxide (ITO), which imparted high electrical conductivity ($455.4 \text{ S} \cdot \text{m}^{-1}$), outstanding electromagnetic interference shielding (24.26 dB), and efficient electrothermal heating performance. More importantly, the ITO-assisted welding effect on the AgNWs networks enhanced electrical conductivity, interfacial adhesion, bending durability, and long-term environmental stability. With these integrated functionalities, the conductive bioplastic emerges as a sustainable enabler for next-generation flexible electronics and personal thermotherapy.

KEYWORDS: bioplastic, supramolecular network, electromagnetic interference shielding, electrothermal heating

1 Introduction

Due to their light weight, durability, and low cost, plastic products have been widely applied in construction, packaging, electronics, and other fields, bringing substantial convenience to modern life [1, 2]. However, large quantities of petroleum-based plastics disposed of in landfills are resistant to degradation, thereby resulting in severe environmental pollution and posing risks to human health [3]. Consequently, the development of biodegradable and recyclable bioplastics has become an effective strategy to mitigate petroleum-based plastic pollution and to align with the principles of sustainable development [4]. Biobased plastics are fabricated using naturally derived or bio-origin feedstocks as structural matrices, thereby facilitating the development of renewable, biodegradable, and non-toxic alternatives [5]. Nevertheless, currently developed bioplastics still suffer from certain limitations, including low tensile strength, poor toughness, and insufficient transparency [6].

To enable bioplastics to meet diverse performance requirements, cellulose is typically composited with other

biomass macromolecules so that strong intermolecular interactions can be established within the polymer network, resulting in cellulose-based composite plastics with enhanced mechanical strength [7]. To date, biomacromolecules such as cellulose and proteins contain abundant functional groups ($-\text{C}=\text{O}$, $-\text{OH}$, and $-\text{NH}_2$), which enable structural integrity to be maintained through noncovalent interactions, thereby contributing to enhanced mechanical strength [8, 9]. Meanwhile, supramolecular networks are formed from high-molecular-weight polymer chains through a facile self-assembly process, in which the synergistic contribution of noncovalent interactions increases the crosslinking density, thereby improving the material toughness [10]. It has been reported that proteins, including gelatin [8], silk fibroin [11, 12], and zein [9] have been incorporated into material systems to achieve enhanced strength and toughness. Thus, while the aforementioned strategies successfully enhance the mechanical strength of bioplastics through supramolecular engineering and biomass macromolecular compositing, these materials still fall short of achieving the multifunctionality required for advanced applications.

Conventional transparent conductive films (TCFs) based on petroleum-based plastics exhibit multifunctional application potential, serving not only as electromagnetic interference (EMI) shielding materials in communication fields but also as electric heaters for thermotherapy in medical applications [13, 14]. Although these systems deliver high conductivity and optical transparency, their non-biodegradable nature and reliance on fossil resources contradict sustainability goals [15]. Moreover, the weak interfacial adhesion between the conductive layer and the plastic substrate often leads to mechanical delamination and performance degradation under bending or thermal stress. To extend bioplastics beyond mechanical strength toward multifunctional applications, integrating transparent conductive materials with bioplastics to fabricate flexible transparent conductive films that simultaneously possess EMI shielding capability, Joule heating functionality, and biodegradability has emerged as a promising direction for next-generation electronic devices [16, 17]. Silver nanowires (AgNWs) have been widely regarded as promising candidates for TCFs due to their excellent flexibility and high electrical conductivity [18, 19]. AgNWs were uniformly deposited onto the cellulose film to fabricate TCFs [20]. However, instability and oxidation susceptibility restrict their long-term use in electronic devices [21]. To address these issues, auxiliary materials (e.g., graphene [22-25], polymers [26, 27], and metal oxides [28]) were introduced onto the AgNWs network to provide protection and reduce junction contact resistance, thus enhancing TCF performance. Moreover, the indium tin oxide (ITO) can significantly enhance interfacial adhesion with AgNWs without noticeably compromising optical transmittance [29, 30].

In this work, a flexible supramolecular bioplastic (GTH) was constructed via a facile self-assembly strategy using gelatin (Gel), TEMPO-oxidized cellulose nanofibril (TOCNF), and hyaluronic acid (HA), where TOCNF served as the structural framework. The formation of the GTH bioplastic was driven by the synergistic interplay between physical chain entanglement and multiple non-covalent interactions. Subsequently, AgNWs and ITO were uniformly deposited onto the GTH surface via spin coating and magnetron sputtering. We hypothesize that the construction of a synergistic supramolecular network among the TOCNF, Gel, and HA through multiple noncovalent interactions, including hydrogen bonding, electrostatic interactions, and physical entanglement, can effectively enhance the mechanical performance of cellulose-based plastics while simultaneously imparting excellent thermal insulation capability, recyclability, and biodegradability. Moreover, the introduction of an ITO overlayer onto the AgNWs conductive layer can substantially improve the electrical conductivity and long-term stability of the film, thereby endowing the material with outstanding EMI shielding performance and superior Joule heating capability. Collectively, this study provides an effective strategy for developing high-performance, multifunctional bio-based plastics that can serve as ideal alternatives to petroleum-based plastics in practical applications, thereby aligning with the fundamental requirements of sustainable development.

2 Experimental

2.1 Materials

A solution of TEMPO-oxidized cellulose nanofibers (TOCNF, 2 wt% solid content dispersed in water, with -COO^- surface groups at a concentration of $1\text{--}2\text{ mmol}\cdot\text{g}^{-1}$, a diameter of $1\text{--}20\text{ nm}$, and a length of $500\text{--}5000\text{ nm}$) was purchased from Dongguan Songhu Shenjian Technology Co., Ltd. Gelatin (Gel, 99.5%) were bought from Shanghai Aladdin Biochemical Technology Co., Ltd. Hyaluronic acid (HA) with a weight-average molecular weight (M_w) of $433,364\text{ g}\cdot\text{mol}^{-1}$ was obtained from Shanghai Macklin Biochemical Technology Co., Ltd. Indium tin oxide target (ITO, $\text{In}_2\text{O}_3\text{:SnO}_2 = 9\text{:}1$, 99.99%) was provided by China Material Technology Co., Ltd. Silver nanowires (AgNWs, $10\text{ mg}\cdot\text{mL}^{-1}$, dispersed in ethanol) with diameters of $\sim 30\text{ nm}$ and lengths of $\sim 20\text{ }\mu\text{m}$ was bought from Nanjing XFANO Materials Technol. Co., Ltd. All reagents were used as received without further purifications.

2.2 Preparation of GTH bioplastic film

The GTH plastic was fabricated via a solution casting method. First, TOCNF (1.0 g) was dispersed in 10 mL of deionized water and magnetically stirred at $45\text{ }^\circ\text{C}$ until a homogeneous and stable aqueous suspension without visible aggregates was obtained. Subsequently, 2.0 g of gelatin and 0.1 g of HA were dissolved in deionized water under heating and then mixed under continuous stirring for 15 min to ensure uniform blending. The prepared TOCNF solution was gradually added to the above mixture and magnetically stirred at $45\text{ }^\circ\text{C}$ for 3 h . The resulting homogeneous solution was cast into a mold and dried at $60\text{ }^\circ\text{C}$ for 45 min , followed by further drying at $40\text{ }^\circ\text{C}$ for 36 h . The obtained film was denoted as GTH and stored at room temperature for subsequent use. For comparison, the films with Gel:TOCNF mass ratios of $1\text{:}2$ and $1.5\text{:}1.5$ were prepared using the same procedure and designated as G1T2H0.1 and G1.5T1.5H0.1, respectively. Additionally, pure TOCNF films, pure Gel films, and Gel/TOCNF composite films with a mass ratio of $2\text{:}1$ were prepared and labeled as TOCNF, Gel, and GT, respectively.

2.3 Fabrication of ITO/AgNWs/GTH composite film

For the fabrication of the conductive composite films, the as-prepared GTH substrate was first cut into pieces with dimensions of $20\text{ mm} \times 35\text{ mm}$. AgNWs dispersions with volumes of 0.2 , 0.3 , and 0.4 mL were then spin-coated onto the surface of the GTH films at 3000 rpm for 60 s to form AgNWs/GTH composite films. The coated films were subsequently annealed at $100\text{ }^\circ\text{C}$ for 3 min to enhance interfacial contact and electrical conductivity. Finally, ITO was deposited onto the AgNWs layer by magnetron sputtering for 15 min under a working pressure of 0.5 Pa and a DC power of 150 W , yielding ITO/AgNWs/GTH (AI/GTH) composite films. According to the deposited volume of AgNWs dispersion, the resulting composite films were designated as 0.2AI/GTH , 0.3AI/GTH , and 0.4AI/GTH , respectively. For comparison, single-component conductive films were also fabricated under identical conditions. The film obtained solely by spin-coating AgNWs was denoted as

the AgNWs/GTH, while the film deposited exclusively by magnetron sputtering of ITO was designated as ITO/GTH.

2.4 Characterization

Molecular weight analysis of hyaluronic acid was performed using an Agilent 1260 Infinity II GPC/SEC system (Agilent Technologies, USA). The sample was dissolved in aqueous solution and analyzed at 45 °C with a flow rate of 1.0 mL·min⁻¹. Separation was carried out using a series of Ultrahydrogel columns, with polyethylene glycol (PEG) standards employed for calibration, and signal detection was performed using a refractive index detector (RID). The surface and cross-section morphology of the GTH films were analyzed using scanning electron microscopy (SEM, Regulus 8230, Hitachi High-Tech Scientific Solutions Co., Ltd., China) and energy dispersive spectroscopy (EDS). The microstructure of GTH was characterized by transmission electron microscopy (TEM, JEM-1400 Flash, JEOL Ltd., Japan). The spectra of the samples in the range of 400-4000 cm⁻¹ were observed using Fourier Transmission Infrared Spectrometer (FTIR, Nicolet iS20, Thermo Fisher Scientific, USA). X-ray Diffraction patterns were characterized by X-ray diffractometer (XRD, 18kW Rigaku D/MAX 2500V, Rigaku Corporation, Japan) at 5°·min⁻¹ from 5° to 60° using Cu K α radiation. The surface composition of the samples was analyzed by X-ray photoelectron spectroscopy (XPS, ESCALAB250Xi, Thermo Fisher Scientific, USA). Raman microscope spectrometer (inVia Qontor, Renishaw, UK) was used to attain Raman spectra in the range of 100-2200 cm⁻¹ with an excitation source of 532 nm or 785 nm. The transmittance spectra of these samples were obtained on an ultra-violet and visible spectrophotometer (UV-2600,

Shimadzu Corporation, Japan). Tensile properties were carried out at a rate of 1 mm·min⁻¹ using a universal testing machine (Instron 5943, Instron, USA) with sheet dimensions of 50 mm × 20 mm under ambient conditions of 25 ± 2 °C and a relative humidity of 40%-60%. In order to ensure statistical accuracy and the capability of reproducing experimental results, each experimental measurement was performed three times. Subsequently, the average value of these measurements was calculated, and the error was evaluated as well. The storage modulus, loss modulus and tan delta of the samples were evaluated using a dynamic mechanical analysis machine (DMA 7100, Hitachi High-Tech Scientific Solutions Co., Ltd., China). The coefficient of thermal expansion of the GTH film was measured using a Thermomechanical Analyzer (TMA 7100, Hitachi High-Tech Scientific Solutions Co., Ltd., China), and the test was conducted in the range of 45-100 °C. Thermal gravimetric analysis was performed on a thermal analyzer (TGA, STA449 F5, NETZSCH, Germany) under nitrogen atmosphere at a heating rate of 10 °C·min⁻¹. Differential Scanning Calorimetry (DSC, DSC 204 F1 Phoenix, NETZSCH, Germany) of the sample was tested in a temperature range of 0 to 100 °C. The bending resistance performance of the sample is measured by a folding endurance tester (YT-CTM, Hangzhou Yante Science and Technology Co., Ltd., China) at ambient temperature. The Joule heating performance of the transparent film was evaluated by applying voltage with a benchtop multimeter (B2901A, Keysight Technologies, USA) and monitoring the film's surface temperature using an infrared thermal imaging camera (IRT, PTiL20-19040144, Uni-Trend Technology Co., Ltd., China).

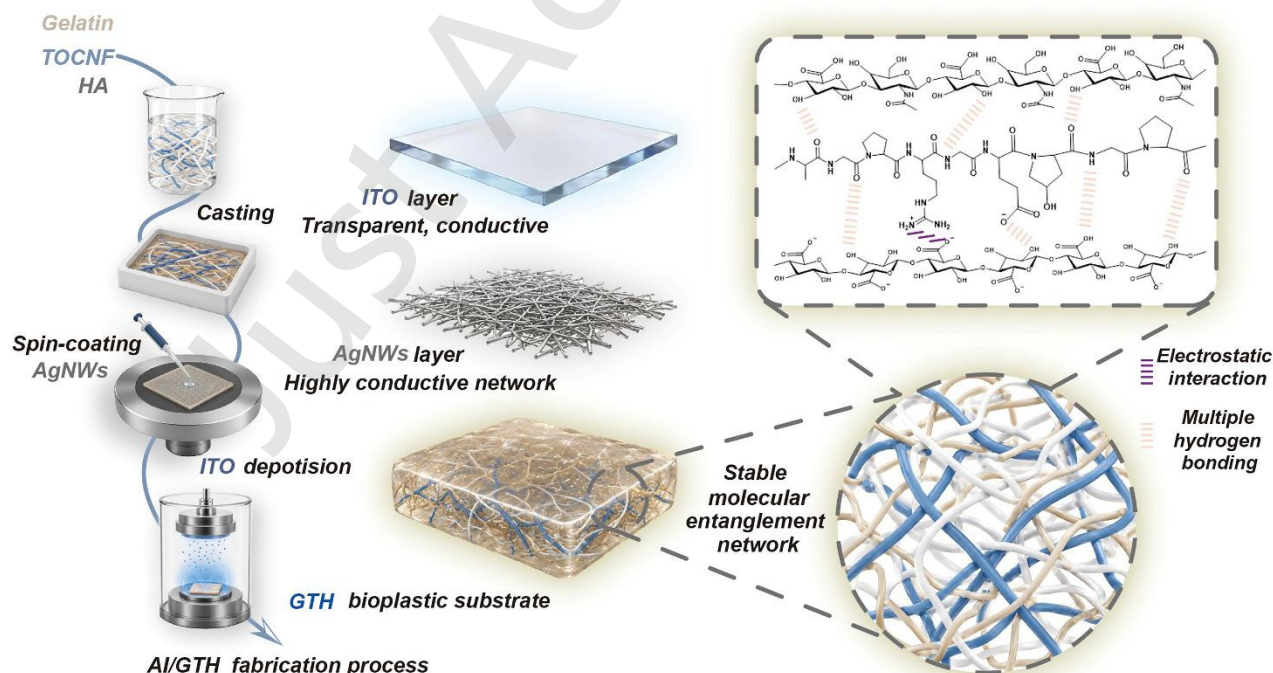


Fig. 1 Schematic illustration of the fabrication process for the Al/GTH composite films, and the non-covalent supramolecular network within the GTH bioplastic substrate.

The antibacterial activity of the GTH and Al/GTH films against *Escherichia coli* (*E. coli*) and *Staphylococcus aureus* (*S. aureus*) were evaluated using the shake flask method.

Experiments were conducted with 0.5 g samples of each film in a bacterial suspension with a concentration of 1×10^6 CFU·mL⁻¹ at 37 °C for 24 h under shaking at 200 rpm. The

sheet resistance of the samples was determined using a standard four-probe measurement system (RTS-8, Guangzhou Four Probe Technology Co., Ltd, China). The shielding effectiveness of the Al/GTH films was measured at room temperature in the X-band frequency range (8.2-12.4 GHz) using a vector network analyzer (E5063A, Agilent Technologies, USA) equipped with the waveguide method. The reflection coefficient (S_{11}) and transmission coefficient (S_{21}) were obtained directly from the vector network analyzer. The total shielding effectiveness (SE_T), reflection loss (SE_R), absorption loss (SE_A), and power coefficients (R , T , and A) were calculated according to the following equations (1) - (6). Multiple reflection loss (SE_M) within the structure were considered to be negligible. All measurements were performed at 25 ± 2 °C and 40-60% RH to accurately simulate standard ambient conditions and ensure data reliability.

$$SE_R = \log\left(\frac{1}{1 - |S_{11}|^2}\right) \#(1)$$

$$SE_A = \log\left(\frac{1 - |S_{11}|^2}{|S_{21}|^2}\right) \#(2)$$

$$SE_T = SE_R + SE_A + SE_M \#(3)$$

$$R = |S_{11}|^2 \#(4)$$

$$T = |S_{21}|^2 \#(5)$$

$$A = 1 - R - T \#(6)$$

3 Results and discussion

3.1 Structural characterization of the GTH bioplastic film

The GTH plastic was prepared by solution casting, involving thorough mixing of gelatin, TOCNF, and HA, followed by mold casting and stepwise drying. Subsequently, using the GTH bioplastic as a substrate, AgNWs dispersions were spin-coated onto its surface, followed by ITO deposition via magnetron sputtering to enhance conductivity (Fig. 1). In particular, beyond electrostatic interaction, a high-density network of multiple hydrogen bonds was established. The synergy of these interactions underpinned a stable supramolecular structure (Fig. 1). Moreover, the breaking and reformation of interchain hydrogen bonds endows the film with excellent recyclability, enabling it to retain its mechanical performance after reprocessing. EDS mapping revealed a uniform distribution of C, N, O, and Na elements across the surface of the GTH film (Fig. S1 in the Electronic supplementary material (ESM)). Microstructural characterization (Fig. S2 in the ESM) showed a porous internal network consisting of fibers with diameters ranging from 19.1 to 26.6 nm. The formation of this architecture is attributed to the uniform deposition of gelatin and HA onto the TOCNF backbone, along with pronounced chain entanglement synergistically occurring among all three components (Fig. S3 in the ESM).

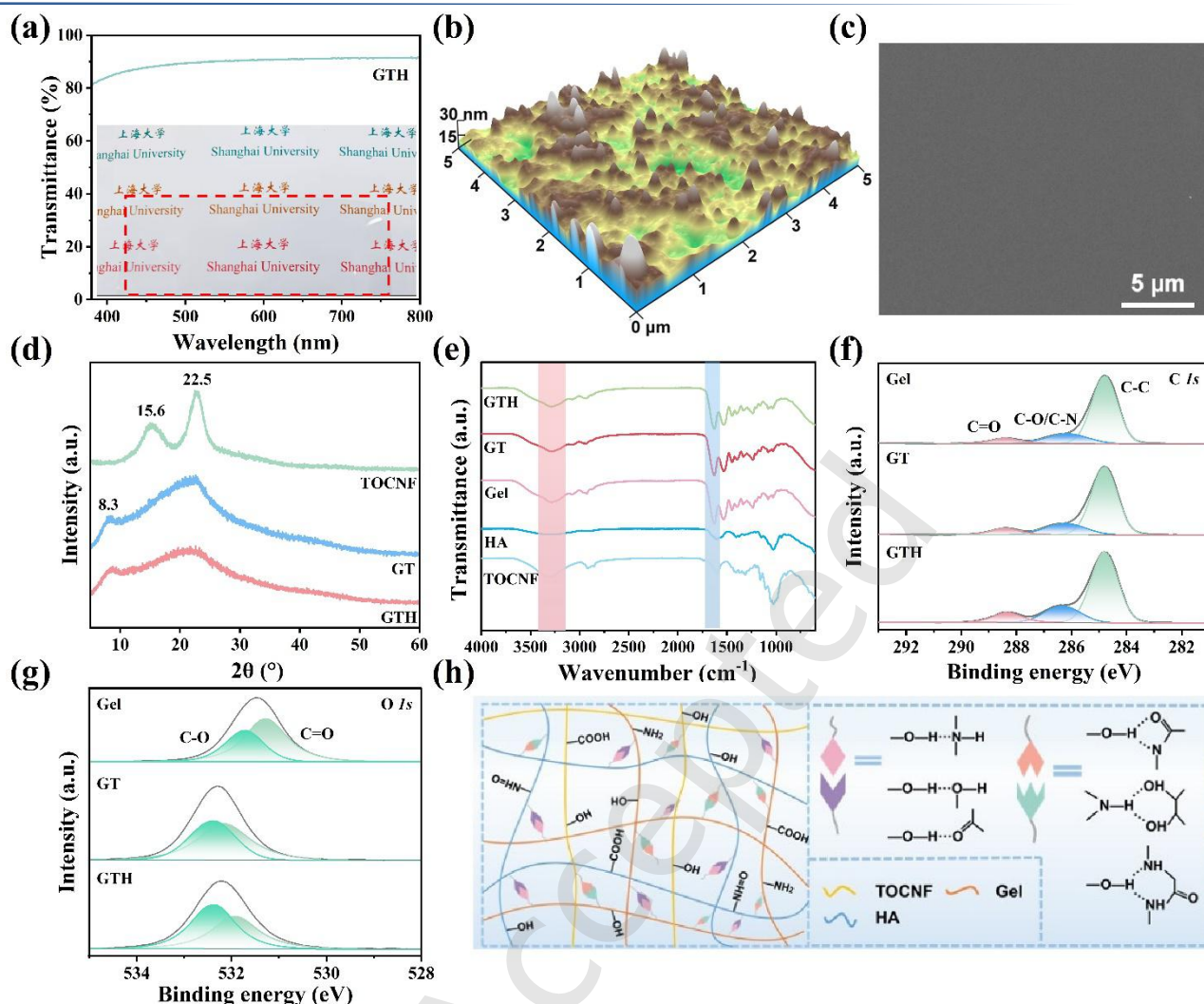


Fig. 2 (a) Transmittance spectra of GTH film. Inset: digital photograph of the GTH film covering a color photo paper. (b) AFM image of GTH film. (c) SEM image of the GTH film surface. Inset: cross-sectional view of the film fracture. (d) XRD patterns of TOCNF, GT, and GTH films. (e) FT-IR spectra of TOCNF, HA, Gel, GT, and GTH. XPS spectra of Gel, GT and GTH for (f) C 1s and (g) O 1s. (h) Schematic illustration of the hydrogen bonding crosslinking network in the GTH film.

High visible-light transparency plays a crucial role in practical applications. The color photo paper covered by the GTH film could be clearly observed with the naked eye, demonstrating the excellent optical clarity of the film (Inset of Fig. 2(a)). The transmission spectrum revealed that the GTH film exhibited a high transmittance of 90.2% at 550 nm (Fig. 2(a)). This high transparency is attributed to the smooth surface resulting from its uniform microstructure, which effectively minimizes light scattering at interfaces.[10] Indeed, the GTH film possessed a smooth and uniform surface with a measured roughness of 5.3 nm (Fig. 2(b)-(c)). XRD patterns revealed that incorporating Gel and HA into the TOCNF matrix caused the GTH composite film to exhibit a broad diffraction peak in the 2θ range of 15° - 25° , indicating a structural transition from a crystalline cellulose phase to a composite phase that contains amorphous components (Fig. 2(d)) [31-33]. This transformation is attributed to the coating effect of cellulose, as well as the formation of strong intermolecular hydrogen bonds, resulting in a composite film dominated by amorphous regions [34]. This finding was further corroborated by Raman spectroscopy (Fig. S4 in the ESM) [35]. Compared

with the GT film, the GTH film exhibited significant attenuation of the characteristic N-H band at 1349 cm^{-1} and the C-N band at 1240 cm^{-1} , indicating the formation of a denser hydrogen-bonding network upon introduction of HA (Fig. S5 in the ESM) [36, 37]. This observation was also confirmed by FT-IR spectroscopy (Fig. 2(e)). In the 3100 - 3500 cm^{-1} region, the O-H and N-H stretching vibration peaks shifted to lower wavenumbers, accompanied by a pronounced narrowing of the broad band, suggesting extensive involvement of hydroxyl and amino groups in hydrogen bonding interactions [38-40]. Additionally, the C=O stretching vibration peak exhibited a blue shift from 1597 cm^{-1} to 1631 cm^{-1} , providing further evidence for stronger and denser hydrogen bonding networks among the polymer chains [41, 42]. Moreover, the characteristic peak of carboxylate ions at 1640 cm^{-1} was observed to shift, indicating the synergistic participation of electrostatic interactions [43]. To further elucidate the chemical environment of the functional groups involved, XPS analysis was carried out (Fig. 2(f)-(g), Fig. S6 in the ESM). Compared with pristine gelatin, the GTH film showed notable binding-energy shifts in the C 1s, O 1s, and N 1s spectra [44].

Specially, the binding energy of C-O increased from 531.7 eV to 532.4 eV, while that of C=O shifted from 531.3 eV to 531.9 eV (Fig. 2(g)). Meanwhile, the relative peak area ratios of C=O and C-N increased, whereas that of C-C decreased (Fig. S7 in the ESM). These changes arise from the incorporation of TOCNF and HA, both rich in carboxyl, carbonyl, and hydroxyl groups, which induced significant alterations in binding energies and peak areas, further verifying the

hydrogen bonding interactions among TOCNF, HA, and gelatin [45, 46]. Owing to the abundant functional groups present along the supramolecular biopolymer chains, sufficient interaction sites were available for non-covalent crosslinking, thereby enabling the formation of a dense and diverse hydrogen-bonding network among the polymer chains (Fig. 2(h)).

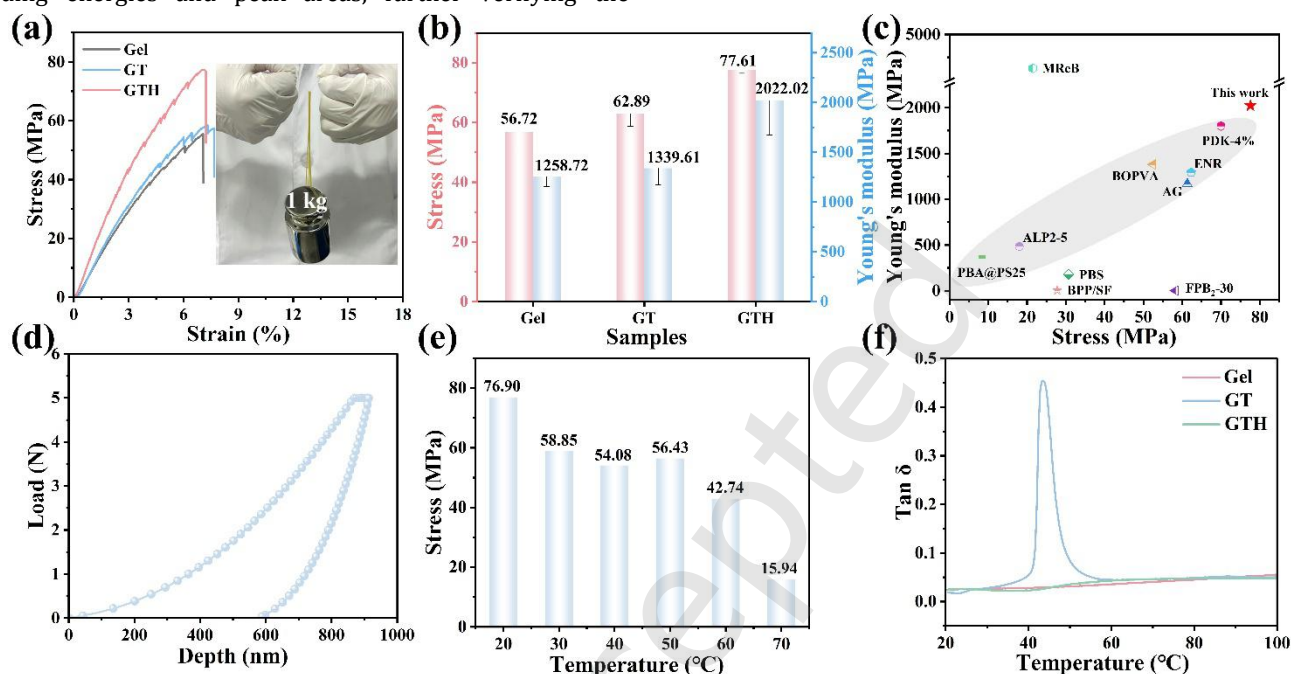


Fig. 3 (a) Tensile stress-strain curves of Gel, GT, and GTH films. Inset: photograph showing a GTH film with a cross-sectional area of 1 mm² supporting a 1 kg weight. (b) Tensile strength and Young's modulus of Gel, GT, and GTH. (c) Comparison of mechanical properties between bioplastics and non-bioplastics (gray shaded area). (d) Nanoindentation test of the GTH film with loading-unloading curves. (e) Tensile stress of GTH films at different temperatures. (f) Tan δ curves of Gel, GT, and GTH obtained from DMA.

3.2 Mechanical properties of the GTH bioplastic film

This high-density hydrogen bonding network served as a critical stabilizing foundation for the supramolecular structure, effectively maintaining the integrity of the network backbone while significantly enhancing the mechanical performance of the film. The mechanical properties of the GTH films were systematically investigated, with particular emphasis on the influence of raw material composition and mass ratio. Among the various formulations examined, the GTH (Fig. 3(a)-(b), Fig. S8-S10 in the ESM) film exhibited outstanding tensile strength, modulus, and toughness, and was therefore selected for subsequent studies. The tensile strength and Young's modulus reached 77.61 MPa and 2022.02 MPa, respectively, representing increases of 36.83% and 60.64% compared with the gelatin film. As shown in the optical image (inset of Fig. 3(a)), a GTH specimen with a cross-sectional area of 1 mm² could support a 1 kg weight without failure. The cellulose-protein-based bioplastic exhibited superior mechanical strength, significantly surpassing previously reported bioplastics and even non-bioplastics (Fig. 3(c), Table S1 in the ESM). The superior mechanical performance of GT film over Gel film is governed by the synergistic interplay between electrostatic interaction and hydrogen bonding interactions at the TOCNF-Gel interface. In the case

of GTH film, the further improvement in tensile strength is ascribed to the incorporation of HA, which facilitates the formation of a more densely packed multiple hydrogen bonding network, thereby establishing a more stable supramolecular crosslinked architecture. Many conventional bioplastics suffer from low tensile strength and intrinsic brittleness owing to strong covalent bonding and high crystallinity, which severely limits their practical application [34, 47]. In contrast, the GTH plastic displayed exceptional flexibility and could withstand 180° bending without structural damage (Fig. S11 in the ESM). Moreover, folding endurance tests demonstrated that the film could sustain over 10,000 folding cycles at a bending angle of 120° without fracture (Fig. S12 in the ESM).

Nanoindentation tests revealed that under a peak load of 5 N, the GTH film exhibited an indentation depth of 0.9 μ m, a surface hardness of 0.3 GPa, and an elastic modulus of 6.4 GPa (Fig. 3(d), Fig. S13-S14 in the ESM), values that exceed those reported for certain conventional inorganic plastics [48]. The high hardness of GTH originates from the extensive chain entanglement within its supramolecular crosslinked network, which effectively suppresses molecular chain slippage and deformation under external stress, thereby endowing the film with excellent resistance to deformation [8]. DMA was further performed to evaluate the tensile behavior of the film over a wide temperature range (Fig.

3(e)). As the temperature increased, the tensile strength gradually decreased, which is likely associated with the thermal denaturation of proteins within the polymer network and subsequent dissociation of the hydrogen-bonding network [49]. The damping factor ($\tan \delta$) is defined as the ratio of loss modulus (E'') to the storage modulus (E'). As shown in Fig. 3(f), the damping factor of GTH was significantly lower in the temperature range of 20–100 °C, indicating that the mobility of polymer chains was effectively restricted [50]. The loss modulus (E'') of GTH remained relatively low and stable, implying reduced energy dissipation due to the formation of a dense crosslinked

network (Fig. S15 in the ESM) [51]. Furthermore, the storage modulus (E') of GTH remained high and stable over the tested temperature range, demonstrating its excellent resistance to deformation (Fig. S16 in the ESM) [52]. The increased damping behavior of GT is attributed to the dynamic interfacial relaxation between TOCNF fibrils and gelatin chains, where reversible hydrogen bonds facilitate energy dissipation [53]. In contrast, HA introduces additional hydrogen-bonding and electrostatic interactions that immobilize polymer chains and reinforce the supramolecular network, thereby reducing molecular relaxation and energy dissipation [54].

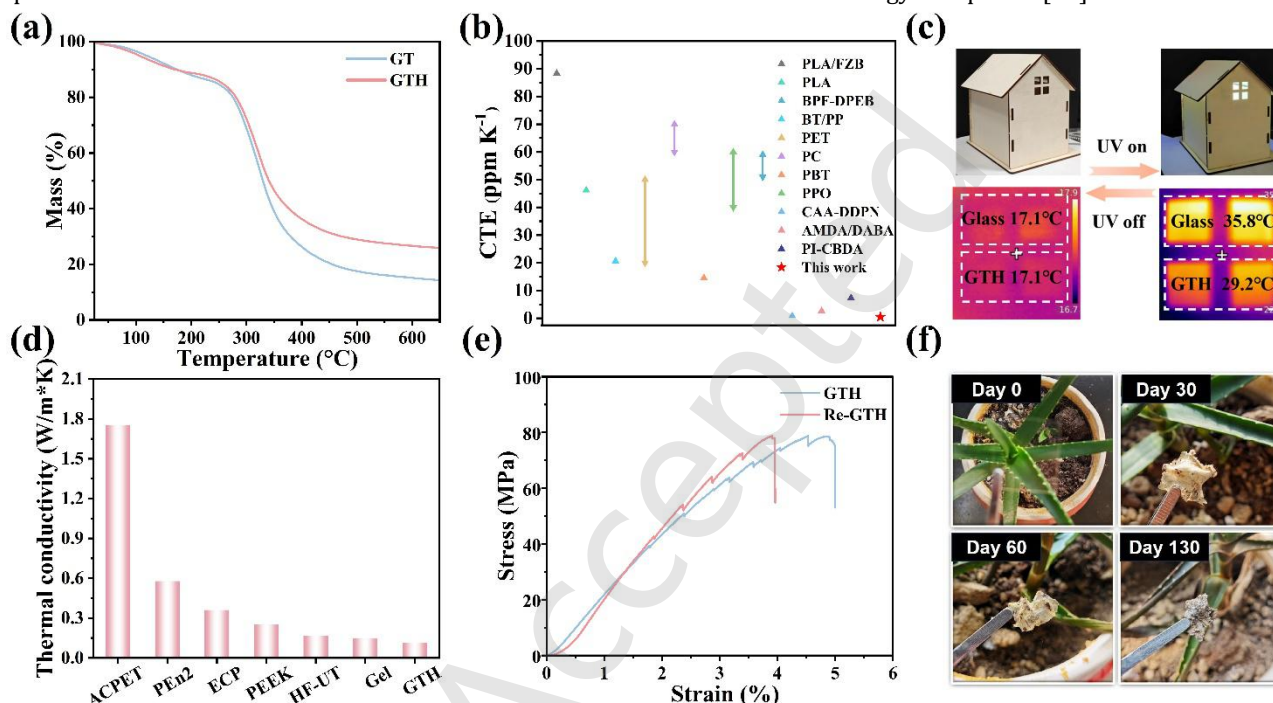


Fig. 4. (a) TGA curves of GT and GTH films. (b) Comparison of the CTE values of various materials. (c) Infrared thermal images of GTH film and bare glass before and after exposure to 365 nm UV irradiation. (d) Comparison of the thermal conductivity between the films and previously reported materials. (e) Tensile stress-strain curves of pristine GTH and recycled GTH. (f) Morphological evolution of the GTH during soil burial in winter.

3.3 Practical sustainability performance of the GTH bioplastic film

Combining excellent mechanical strength, high stability, superior thermal insulation, and inherent degradability, this bio-based plastic represents as an ideal sustainable alternative to non-degradable petroleum-based plastics. TGA revealed that GTH exhibits high thermal stability, with an initial decomposition temperature of 169.1 °C and a maximum decomposition temperature of 322.4 °C (Fig. 4(a), Fig. S17 in the ESM). The former is attributed to the evaporation of physically adsorbed water, whereas the latter corresponds to the rapid thermal dissociation of the hydrogen bonding network among the polymer chains [55]. DSC analysis further revealed a glass transition temperature (T_g) of 255.4 °C (Fig. S18 in the ESM). The film exhibits a low coefficient of thermal expansion (CTE) of $5 \times 10^{-7} \text{ K}^{-1}$, comparable to that of quartz glass [56], demonstrating negligible dimensional change upon heating (Fig. S19 in the ESM). This CTE is considerably lower than those reported for most conventional plastics (Fig. 4(b), Table S2 in the ESM). This favorable property arises from the synergistic

interplay between the internal pores, which effectively absorb thermally induced expansion stress, and the strong hydrogen bonding network, which locks the ordered arrangement of molecular chains, thereby collectively suppressing macroscopic deformation and volumetric expansion [51, 57]. Infrared thermography intuitively demonstrated that, under identical 365 nm UV irradiation, a GTH film mounted on a model house window exhibited a substantially lower temperature than ordinary glass of the same thickness, reflecting a considerably slower heating rate (Fig. 4(c)). Moreover, GTH took 12 min to warm from room temperature to 40.0 °C, three times longer than ordinary glass (Fig. S20–S22 in the ESM). The thermal conductivity of GTH was measured to be as low as $0.11 \text{ W} \cdot \text{m}^{-1} \text{ K}^{-1}$ (Fig. S23 in the ESM), which is significantly lower than that of most previously reported materials, further confirming its outstanding thermal insulation capability (Fig. 4(d)) [58–62]. The superior thermal insulation performance is attributed to the intrinsic thermal resistance of the polymer matrix and the disruption of continuous heat conduction pathways by the internal porous structure (Fig. S24 in the ESM) [63–65].

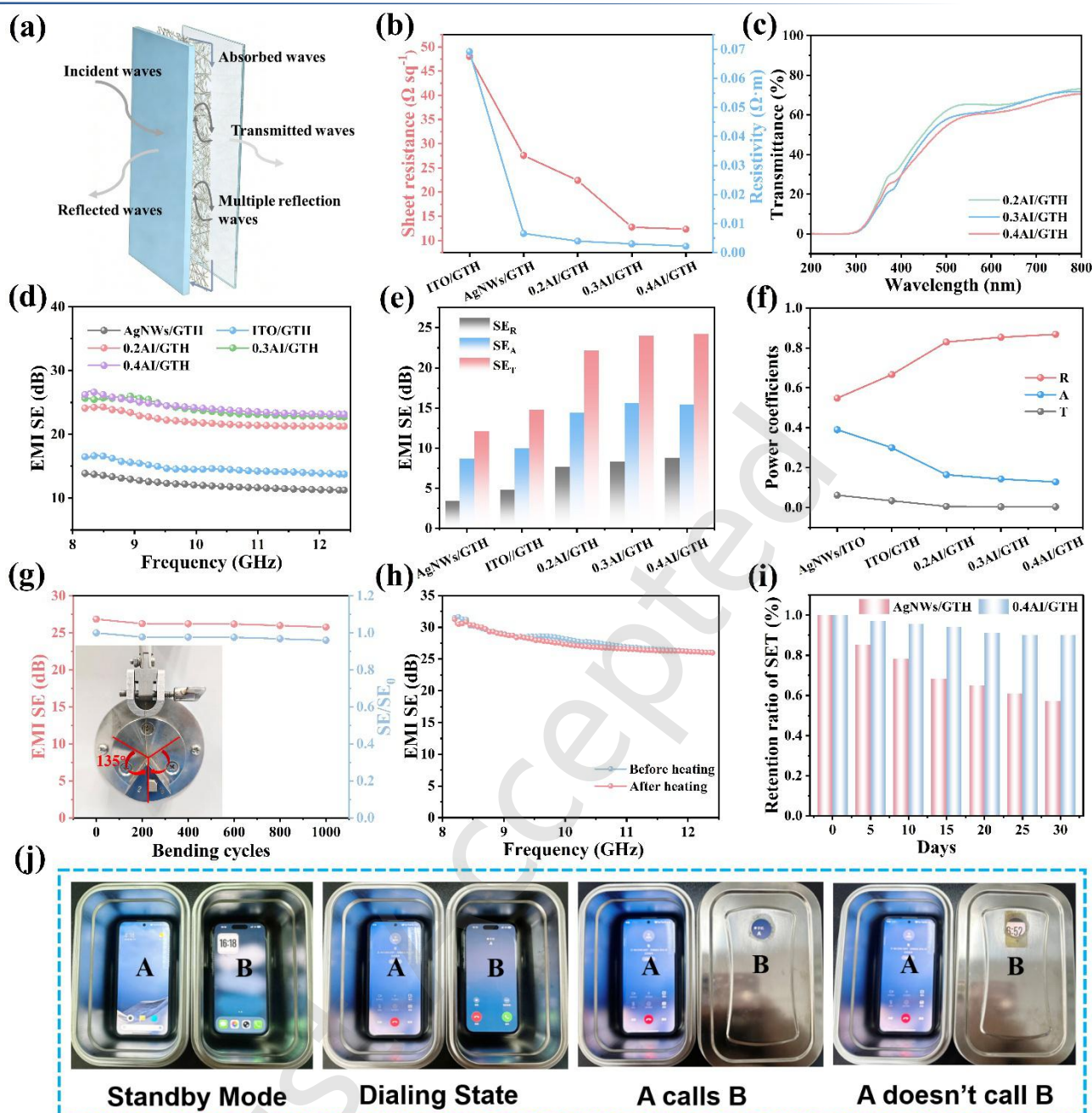


Fig. 5 (a) Schematic illustration of EMI shielding mechanism of the AI/GTH film. (b) Sheet resistance and resistivity of the ITO/GTH, AgNWs/GTH, and AI/GTH films with different loading of AgNWs. (c) Visible-light transmittance of the AI/GTH films with different loading of AgNWs. (d) EMI SE of the ITO/GTH, AgNWs/GTH, and AI/GTH films with different loading of AgNWs. (e) SER, SEA, and SET values for films with different loading contents (f) Power coefficients of films with different loading contents. (g) EMI SE and the corresponding retention ratio (SE/SE_0) of 0.4AI/GTH films as a function of the number of bending cycles, where SE_0 and SE represent the EMI SE values before and after bending, respectively. Inset: optical image of the film during the flexural durability test. (h) EMI SE of the 0.4AI/GTH composite film measured before and after thermal treatment at 40 °C for 5 min. (i) Variation of the SE/SE_0 ratio of the 0.4AI/GTH film during exposure to ambient air at room temperature. (j) Photographs demonstrating the EMI SE of the 0.4AI/GTH composite film. A mobile phone B placed inside an aluminum box shows signal reception when the box opening is uncovered, but loses signal when the opening is covered with the 0.4AI/GTH film.

In addition, the recyclability and degradability are key indicators of environmental sustainability for bioplastic materials [66]. The GTH plastics demonstrated excellent closed-loop recyclability (Fig. S25 in the ESM). After being crushed, re-dissolved in water, and recast, the recycled film showed no significant changes in either the tensile strength or optical transmittance of (Fig. 4(e), Fig. S26 in the ESM). Furthermore, the film exhibited pronounced degradability after 130 days of burial in winter soil (Fig. 4(f)), as well as

remarkable solvent resistance, with no notable changes in shape or color observed after 12 h of immersion in a range of solvents (Fig. S27 in the ESM).

3.4 EMI shielding performance of AI/GTH composite film

A double-layer conductive architecture was constructed on the GTH substrate to fabricate the AI/GTH composite film, thereby endowing it with EMI shielding capability (Fig. 5(a)). Within the conductive network of the AI/GTH film, the

intermediate AgNWs layer served as the primary conductive framework for electron transport. Meanwhile, the top ITO layer both protected and welded the AgNWs while providing an additional conductive pathway, thereby enriching the three-dimensional conductive network and synergistically contributing to the overall electrical conductivity [16]. As a result of this multilayer design, the Al/GTH film exhibited an electrical conductivity as high as $455.4 \text{ S}\cdot\text{m}^{-1}$, along with significantly reduced sheet resistance and resistivity compared with single-layer conductive counterparts (Fig. S28 in the ESM, Fig. 5(b)). This structural design not only substantially enhanced electrical conductivity but also preserved high optical transmittance and low haze, yielding superior comprehensive optoelectronic properties (Fig. 5(c), Fig. S29 in the ESM) [67]. The transparent Al/GTH film achieved an EMI shielding effectiveness (EMI SE) of 24.26 dB in the X-band frequency range (8.2–12.4 GHz) (Fig. 5(d)). The EMI shielding performance was governed by a reflection-

dominant mechanism (Fig. 5(e)-(f)). The high reflection coefficient (R) originated from the impedance mismatch at the air-ITO interface, which caused the majority of incident electromagnetic waves to be reflected at the surface (Fig. 5(a)) [68–70]. Once transmitted into the interior, the remaining waves experienced energy attenuation through several mechanisms [71]. Interactions with free electrons and dipoles in the ITO-coated AgNWs layer induce ohmic losses [72, 73]. Interfacial polarization loss was attributed to the $-\text{CO}-\text{NH}-$ moieties of gelatin and the COO^- groups of TOCNF, as well as the conductivity contrast at the interface between the conductive ITO/AgNWs and the insulating GTH matrix, which further generated additional interfacial loss [74, 75]. Moreover, the layered architecture prolonged the propagation path of the electromagnetic waves, thereby promoting multiple reflection losses at the GTH/AgNWs and AgNWs/ITO heterointerfaces [76–78].

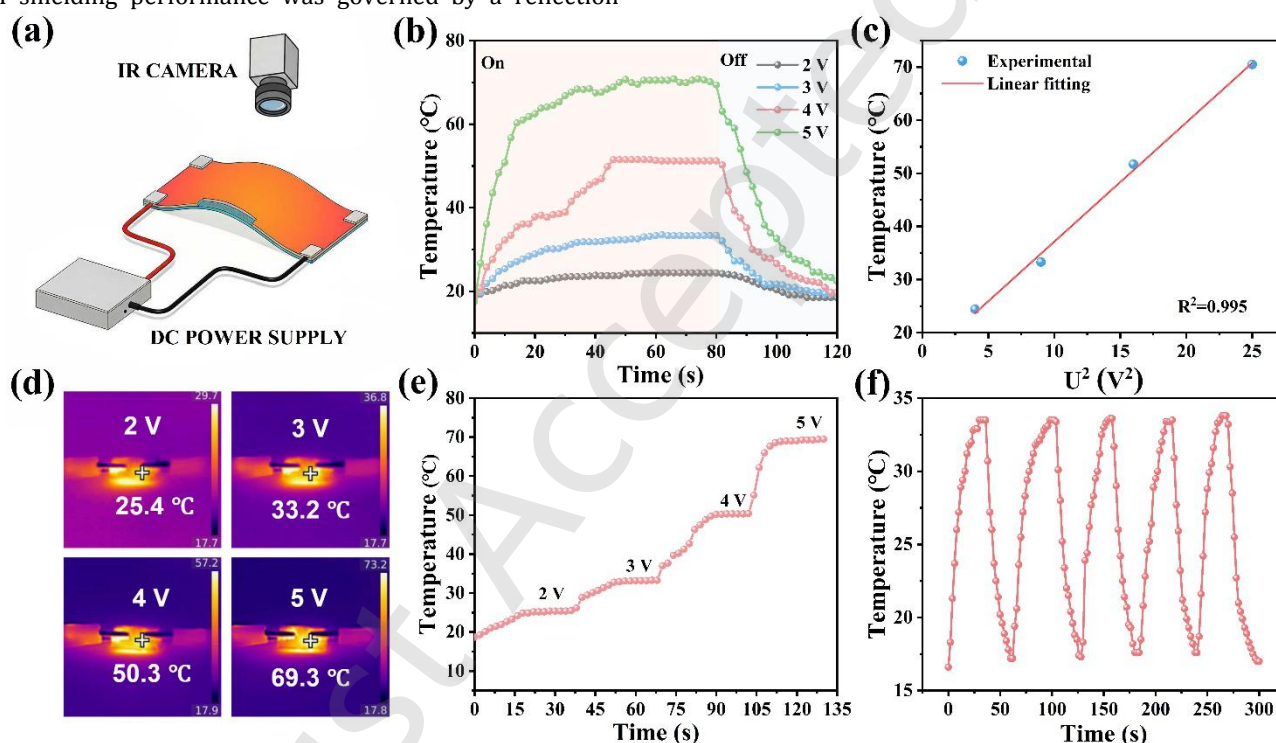


Fig. 6. (a) Schematic illustration of the experimental setup for Joule heating measurements. (b) Temperature-time curves of the 0.4Al/GTH films under different applied voltages. (c) Linear fit of the steady-state temperature as a function of the square of the applied voltage (U^2). (d) Infrared thermal images of the 0.4Al/GTH film recorded at various voltages. (e) Temperature variation of the 0.4Al/GTH film under stepwise increasing voltage inputs. (f) Cyclic temperature performance of the 0.4Al/GTH film under repeated on-off switching of a 3 V input over a 300 s test period.

The stability of transparent EMI shielding films is critically important for practical applications. The Al/GTH film retained high tensile strength (Fig. S30 in the ESM). Moreover, after 1000 bending cycles or heating processes, no significant degradation in EMI SE was observed (Fig. 5(g)-(h), Fig. S31 in the ESM). Additionally, even after exposure to air for one month, the Al/GTH film maintained satisfactory EMI shielding performance (Fig. 5(i), Fig. S32–33 in the ESM). The outstanding stability, which enables its application in electric heaters and electronic devices, can be attributed to the protective effect of the ITO layer coated on the AgNWs [29]. A mobile phone signal shielding experiment was further conducted to verify the EMI effectiveness (Fig. 5(j)). Under normal conditions, Phone A successfully called Phone

B when Phone B was in standby mode. When Phone B was placed inside an aluminum box with an open window, the call could still be connected. However, when the window of the aluminum box was covered with the Al/GTH film, Phone B failed to receive the call, indicating that signal transmission was effectively blocked.

3.5 Joule Heating and Antibacterial Performance of Al/GTH Composite Film

Owing to its high electrical conductivity and low sheet resistance, the Al/GTH composite film exhibited excellent Joule heating performance and can serve as a transparent heating panel for optoelectronic devices (Fig. 6(a), Fig. S34 in the ESM) [79]. Fig. 6(b) illustrates the temperature

evolution of the AI/GTH transparent film (15 mm × 25 mm) under different applied voltages. As the applied voltage increased from 2 to 5 V, the steady-state temperature of the film increased correspondingly, reaching a maximum value of 70.5 °C. The higher saturation temperature achieved at elevated direct current (DC) voltage is primarily attributed to enhanced inelastic collisions between accelerated electrons and phonons under the applied electric field [80]. As shown in Fig. 6(c), a linear relationship was observed between the square of the applied voltage and the steady-state temperature, consistent with Joule's law ($Q=U^2t/R$, where Q , U , t , and R denote Joule heat, applied voltage, heating time, and resistance, respectively) [69, 81]. The temperature regulation capability was further evaluated by increasing the applied voltage from 2 to 5 V (Fig. 6(d)-(e)). Real-time temperatures at various voltages were recorded using an infrared thermal imaging camera. The infrared images demonstrated stable and controllable heating behavior under different input voltages. When operated at a constant voltage of 4 V for 1800 s, only minimal temperature fluctuation was observed, indicating excellent long-term operational stability (Fig. S35 in the ESM). Moreover, during repeated voltage on-off cycles, the temperature variation closely followed the switching of the input voltage, demonstrating reliable cyclic heating performance (Fig. 6(f)) [68].

In addition to heating functionality, antibacterial properties are of paramount importance for ensuring user safety and hygiene in wearable heater applications. The AI/GTH film exhibited excellent antibacterial activity against both *E. coli* and *S. aureus* (Fig. S36 in the ESM). This antibacterial effect is ascribed to the interaction of Ag^+ ions released from the silver with thiol groups in bacterial proteins, leading to protein inactivation [82, 83].

Combining efficient heating performance, superior thermal stability, and outstanding antibacterial capability, the AI/GTH film holds great promise for application in flexible wearable electronic devices, where it can be operated at safe working voltages [67, 84].

4 Conclusions

In summary, an environmentally benign and facile self-assembly strategy was successfully employed to successfully construct a biobased plastic with a multiple hydrogen bonding network. The resulting bioplastics demonstrated high transparency (90.2%@550 nm), superior mechanical properties (tensile strength of 77.61 MPa), excellent thermal insulation (thermal conductivity of $0.11 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$), as well as recyclability and degradability. The exceptional mechanical properties are attributed to the synergistic interplay of hydrogen bonding and electrostatic interaction among gelatin, TOCNF, and HA, which facilitates the formation of a highly entangled supramolecular network. Furthermore, a transparent conductive AI/GTH film was fabricated by sequentially introducing AgNWs and ITO through spin coating and magnetron sputtering. The welding effect of ITO on the AgNWs networks further reinforced the conductive network, ensuring simultaneous improvements in both electrical conductivity and structural stability. The double-layer conductive network imparted the film with a

synergistic integration of high electrical conductivity ($455.4 \text{ S}\cdot\text{m}^{-1}$), excellent EMI shielding performance (24.26 dB), and efficient electrothermal heating capability. The AI/GTH film maintained stable performance under repeated bending, thermal treatment, and long-term storage, demonstrating exceptional durability and reliability. Overall, this work provides a sustainable strategy for designing high-performance biobased films that integrate mechanical robustness, thermal insulation, electrical conductivity, and EMI shielding functionality, highlighting their promising potential for applications in flexible electronics and wearable heating devices.

Electronic Supplementary Material: Supplementary material (please provide the brief detail of the ESM) is available in the online version of this article at <https://doi.org/10.26599/NR.2026.94909095>.

Data availability

All data needed to support the conclusions in the paper are presented in the manuscript and/or the Electronic Supplementary Material. Additional data related to this paper may be requested from the corresponding author upon request.

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Declaration of competing interest

All the contributing authors report no conflict of interests in this work.

Author contribution statement

Wen Gong: Writing - original draft, Visualization, Software, Methodology, Formal analysis, Investigation, Data curation. Jianyu Zhou: Software, Methodology. Yuchen Tian: Software, Methodology. Qi Zhang: Methodology. Puyan Huang: Software, Methodology. Run Gan: Methodology. Xiang He: Resources, Conceptualization. Miao Miao: Validation, Project administration. Xin Feng: Writing - review & editing, Supervision, Project administration, Funding acquisition, Conceptualization, Resources.

Ethics statement

Not applicable.

Use of AI statement

None.

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Electronic Supplementary Material

Supramolecular network-engineered transparent conductive cellulose/gelatin bioplastics for efficient Joule heating and electromagnetic interference shielding

Wen Gong^{1,2}, Jianyu Zhou^{2,3}, Yuchen Tian², Qi Zhang⁴, Puyan Huang², Run Gan⁵, Xiang He¹ , Miao Miao², and Xin Feng^{1,2}  

¹ Department of Chemistry, College of Sciences, Shanghai University, Shanghai 200444, China

² Research Center of Nanoscience and Technology, College of Sciences, Shanghai University, Shanghai 200444, China

³ School of Materials Sciences and Engineering, Tongji University, Shanghai 201804, PR China

⁴ Department of Electronic Information Materials, School of Materials Science and Engineering, Shanghai University, Shanghai 200444, China

⁵ College of Food Science and Technology, Shanghai Ocean University, Shanghai 201306, China

 Address correspondence to Xiang He, hxiang@shu.edu.cn; Xin Feng, fengxin@shu.edu.cn

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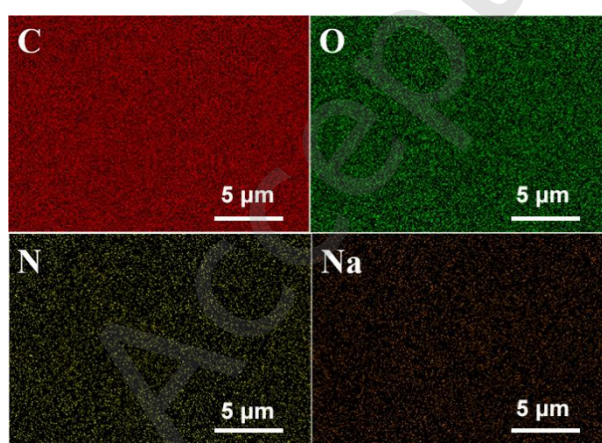


Fig. S1 EDS mapping images of the GTH film showing the distribution of C, O, N, and Na.

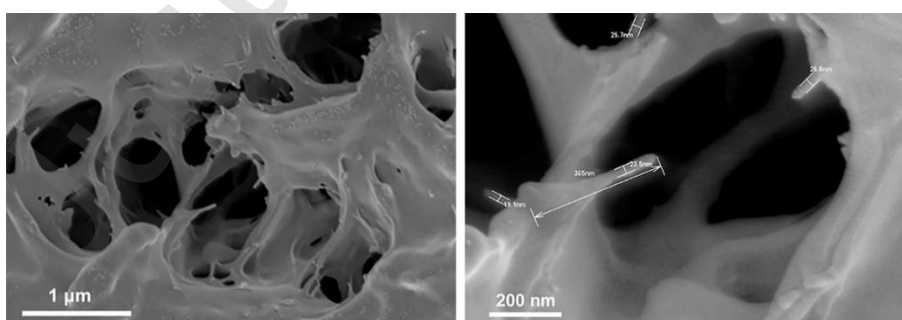


Fig. S2 Cross-sectional SEM images of the GTH film.

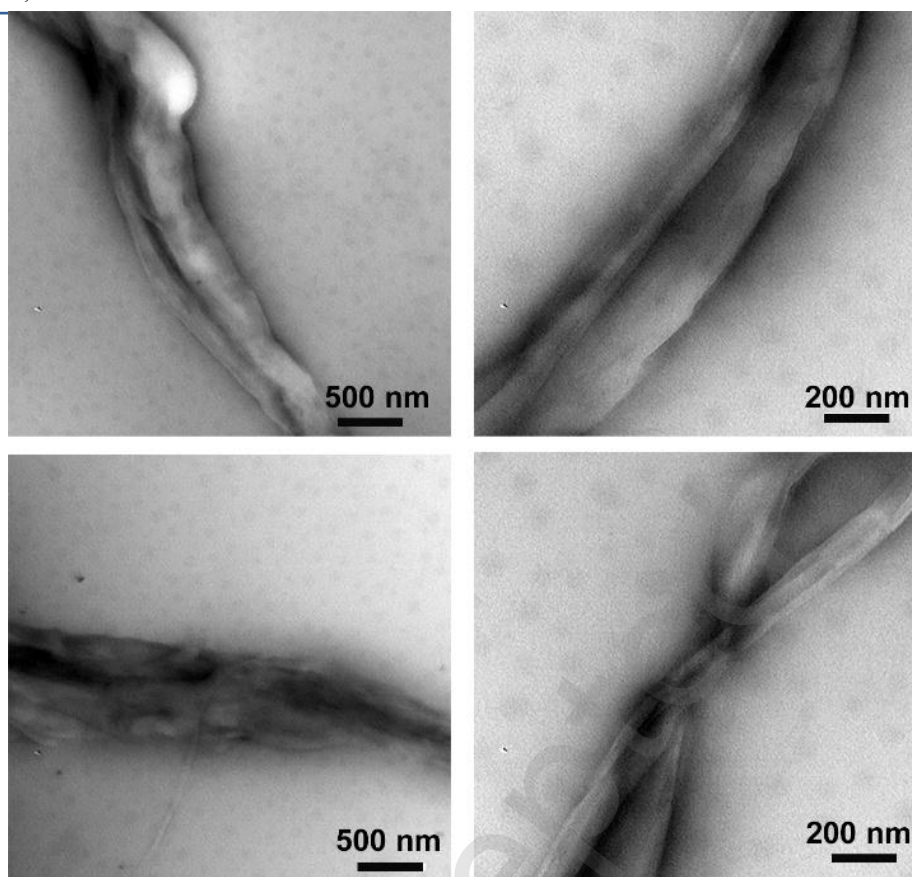


Fig. S3 TEM images of the GTH film. Scale bars: 200 nm (right two images) and 500 nm (left two images).

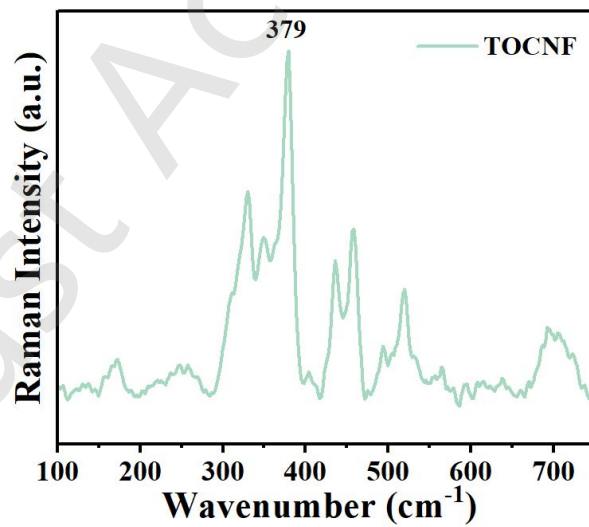


Fig. S4 Raman spectrum of TOCNF recorded with 532 nm excitation.

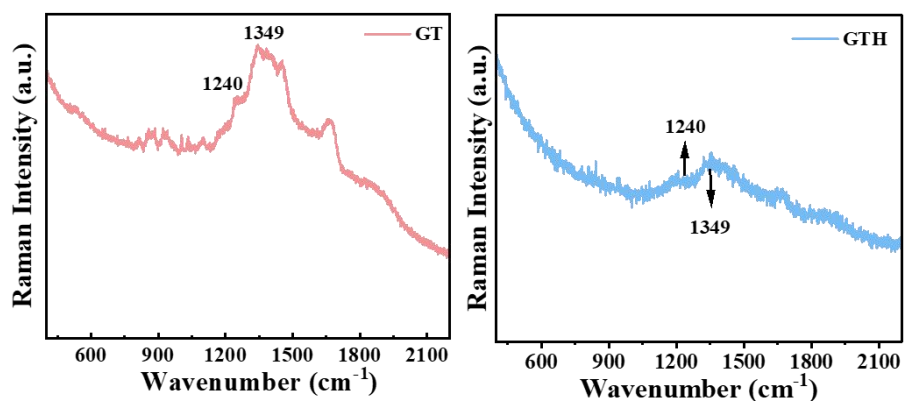


Fig. S5 Raman spectra of GT and GTH under 785 nm excitation.

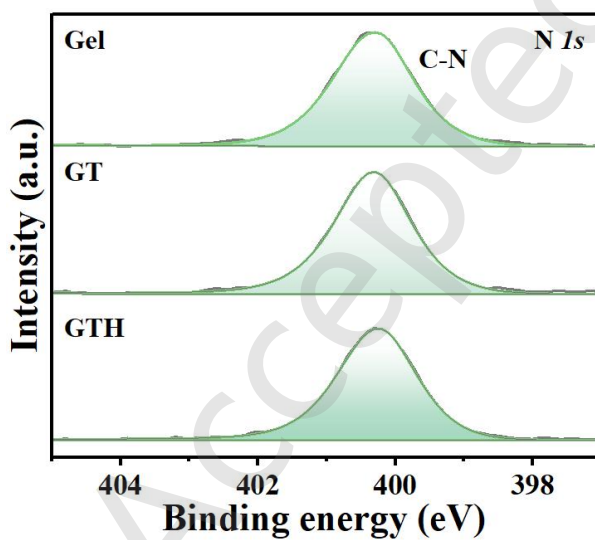


Fig. S6 High-resolution XPS N 1s spectra of Gel, GT, and GTH.

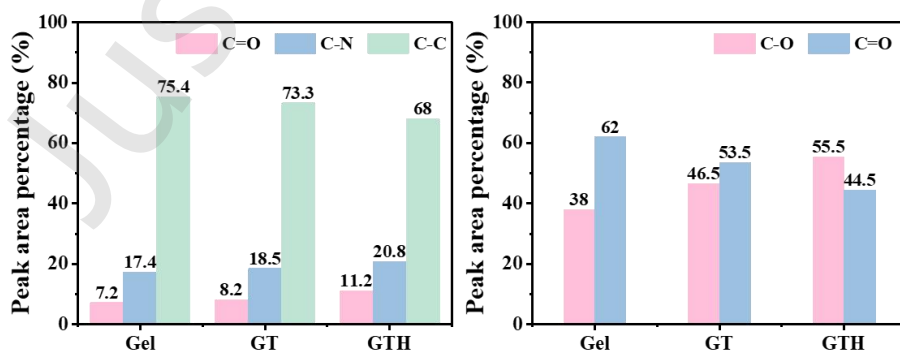


Fig. S7 Peak area variations of C=O, C=N, and C-C bonds derived from the C 1s and O 1s XPS spectra of Gel, GT, and GTH.

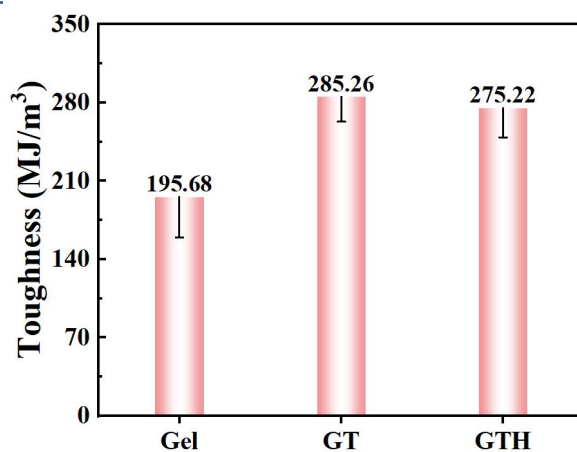


Fig. S8 Toughness comparison of Gel, GT, and GTH films.

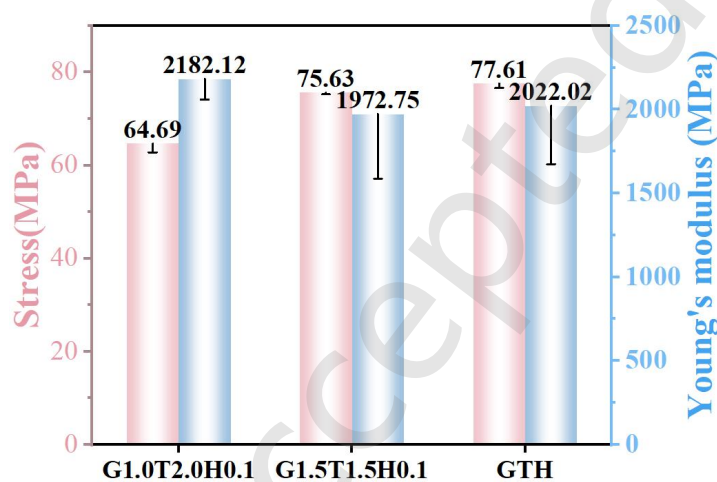


Fig. S9 Comparison of stress and Young's modulus of the G1.0T2.0H0.1, G1.5T1.5H0.1, and GTH films.

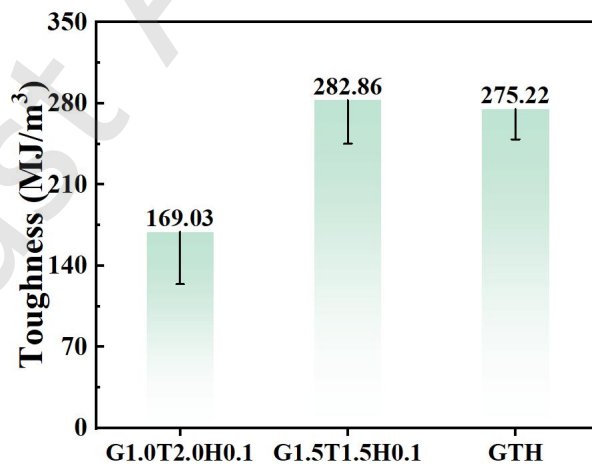


Fig. S10 Toughness comparison of the G1.0T2.0H0.1, G1.5T1.5H0.1, and GTH films.

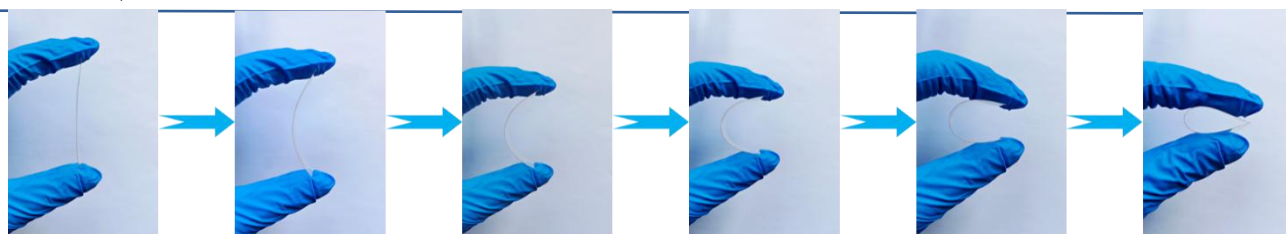


Fig. S11 Digital pictures of the GTH film captured during the compression test.



Fig. S12 Digital picture of the bending deformation of the GTH film.

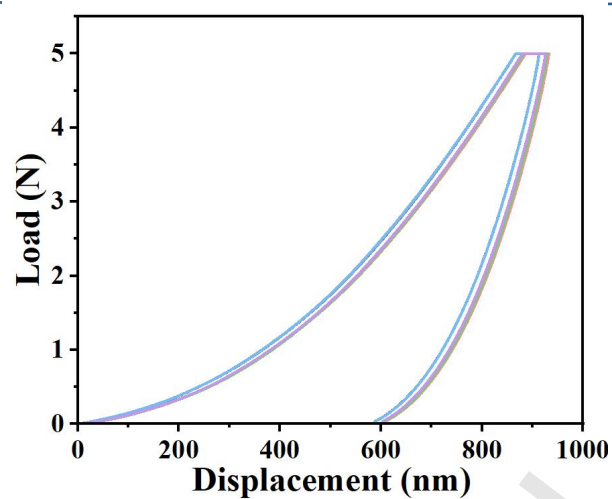


Fig. S13 Five consecutive loading-unloading cycles obtained from nanoindentation testing of the GTH film.

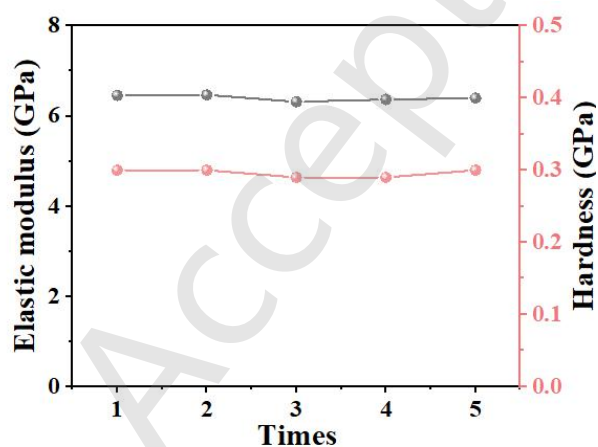


Fig. S14 Elastic modulus and hardness of the GTH film measured by five nanoindentation tests.

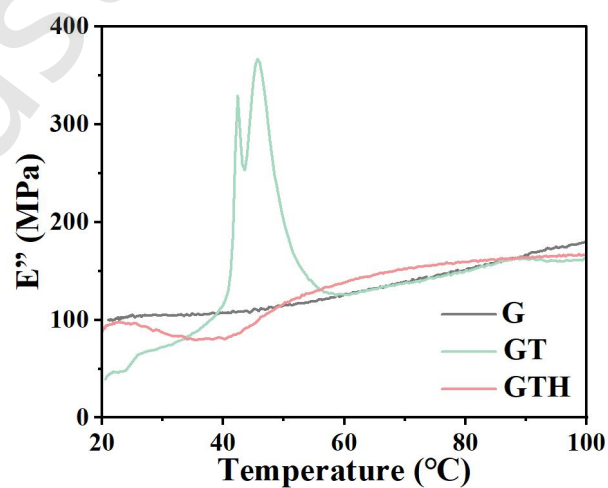


Fig. S15 Loss moduli of Gel, GT, and GTH obtained from DMA.

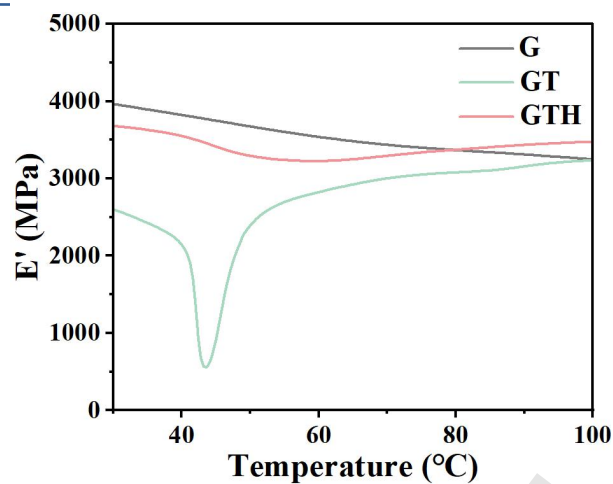


Fig. S16 Storage moduli of Gel, GT, and GTH obtained from DMA.

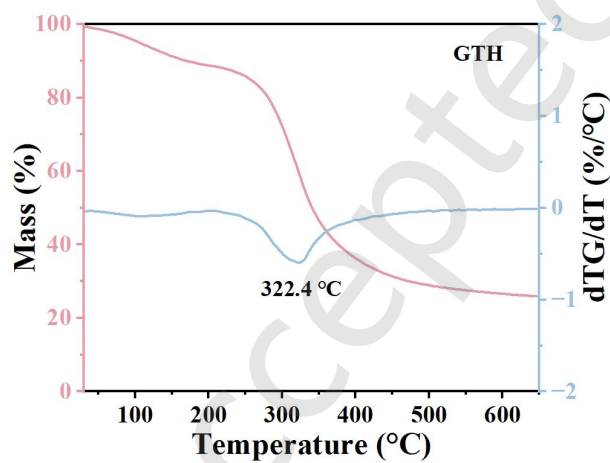


Fig S17 TG and DTG curves of the GTH film.

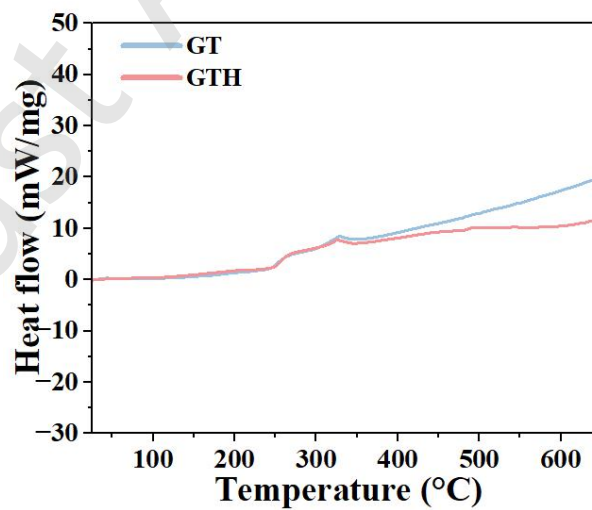


Fig S18 DSC curves of the GT and GTH films.

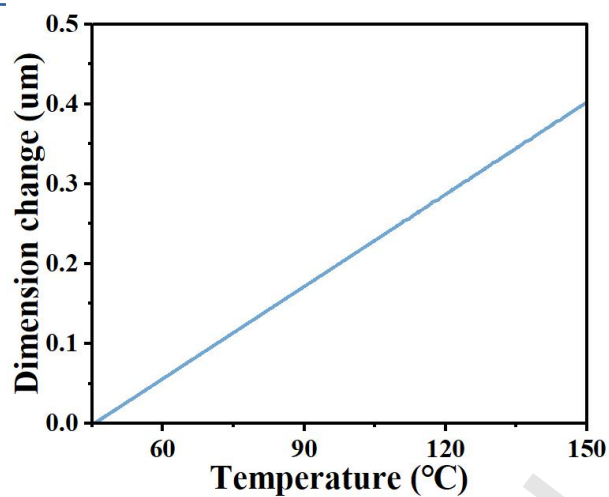


Fig. S19 Dimensional change of the GTH film as a function of temperature from 45 °C to 150 °C measured by TMA.

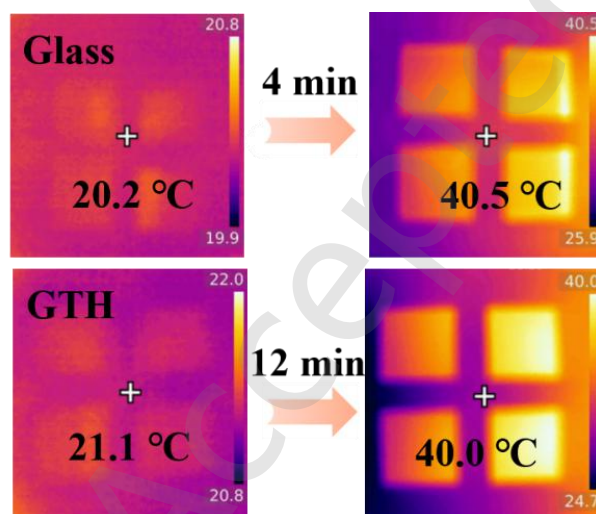


Fig. S20 Comparison of the time required for GTH film and bare glass to reach approximately 40 °C under 365 nm UV irradiation.

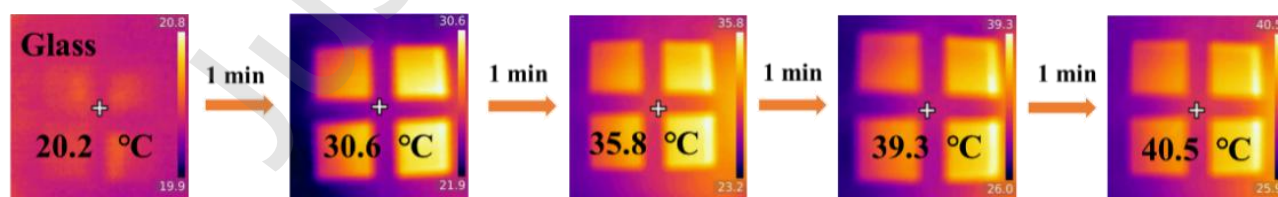


Fig. S21 Temperature evolution of bare glass under 365 nm UV irradiation up to 40.5 °C.

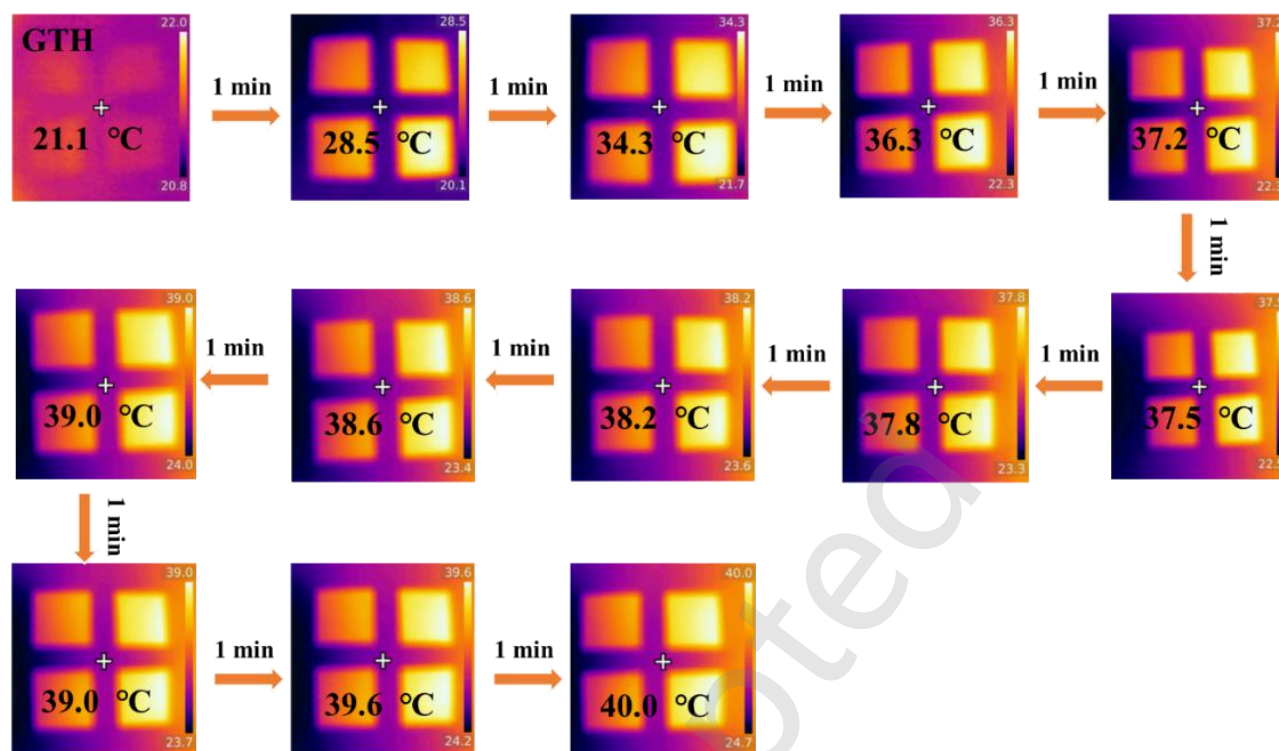


Fig. S22 Temperature evolution of the GTH film under 365 nm UV irradiation up to 40.0 °C.

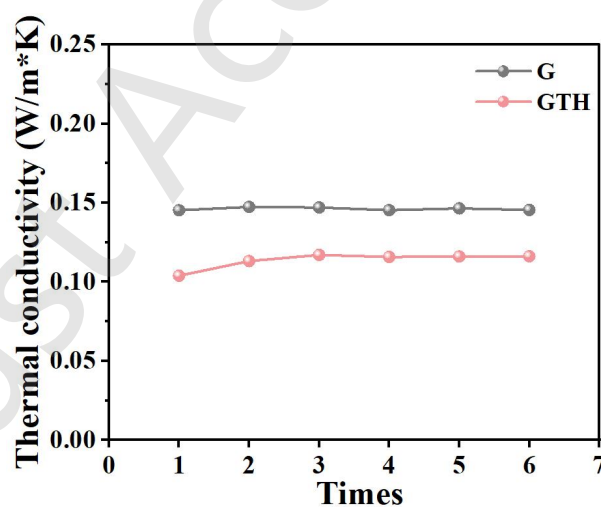


Fig. S23 Thermal conductivity values of the Gel and GTH films derived from five independent replicate measurements ($n = 5$).

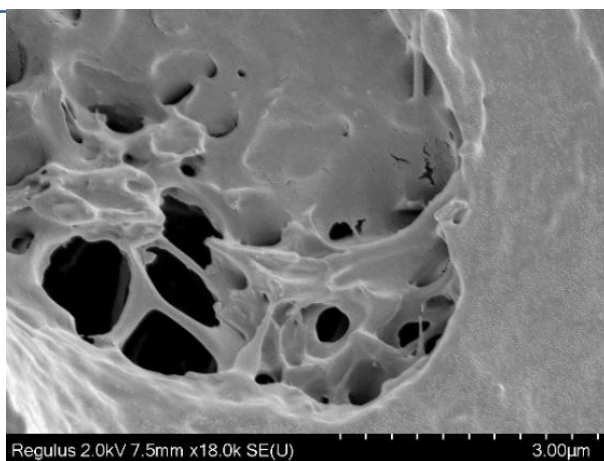


Fig. S24 Cross-sectional SEM images of the GTH film.



Fig. S25 Schematic illustration of the recycling process for the GTH film via dissolution and regeneration: the film is shredded, dissolved in water, re-cast, and finally dried to produce a recycled film.

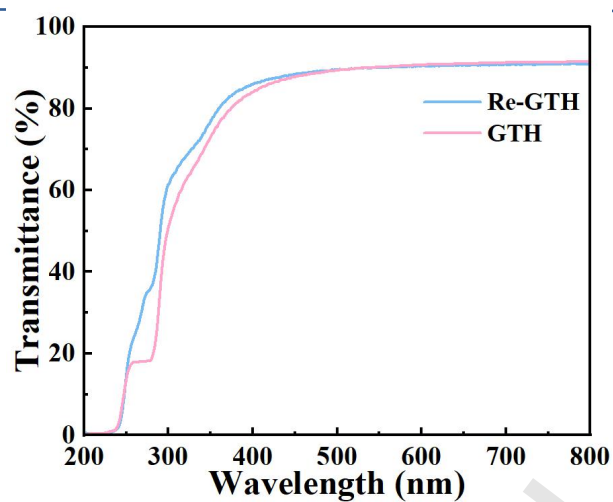


Fig. S26 Comparison of the light transmittance of pristine GTH and its recycled counterpart (Re-GTH).

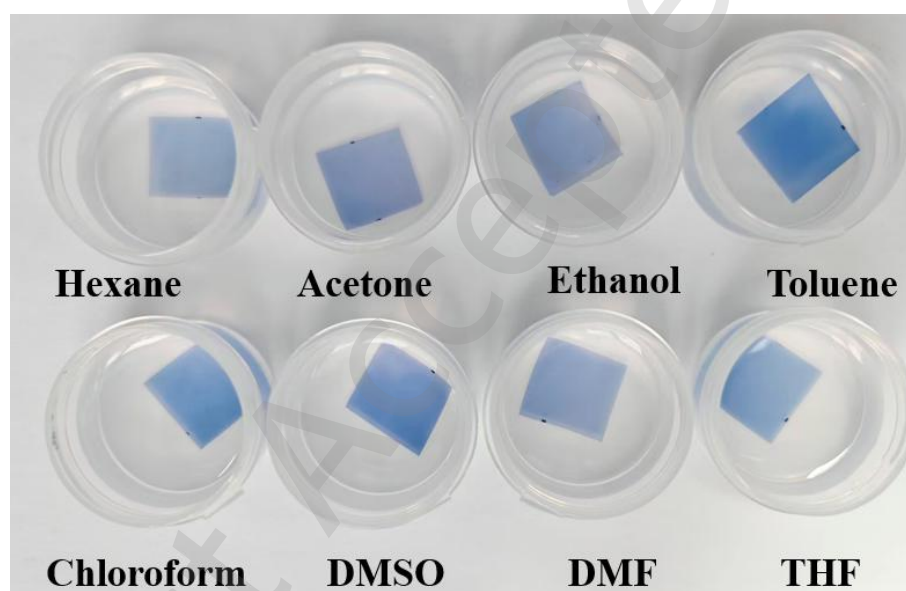


Fig. S27 Stability evaluation of GTH immersed in various solvents at 25 °C. No swelling was observed even after 12 h of immersion in hexane, acetone, ethanol, toluene, chloroform, DMSO (dimethyl sulfoxide), DMF (N, N-dimethylformamide), and THF (tetrahydrofuran).

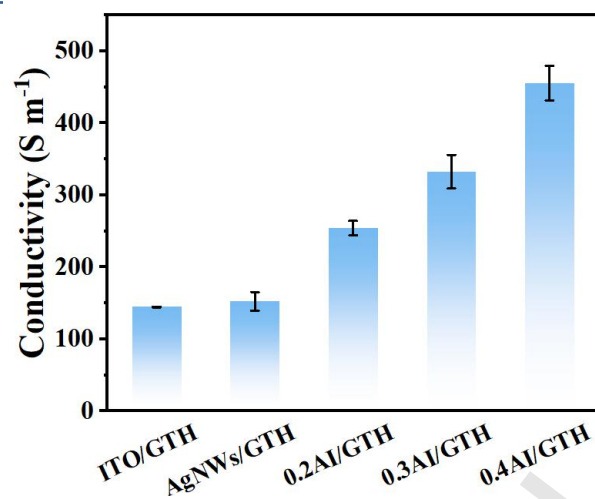


Fig. S28 Comparison of the electrical conductivity of the films with different loadings.

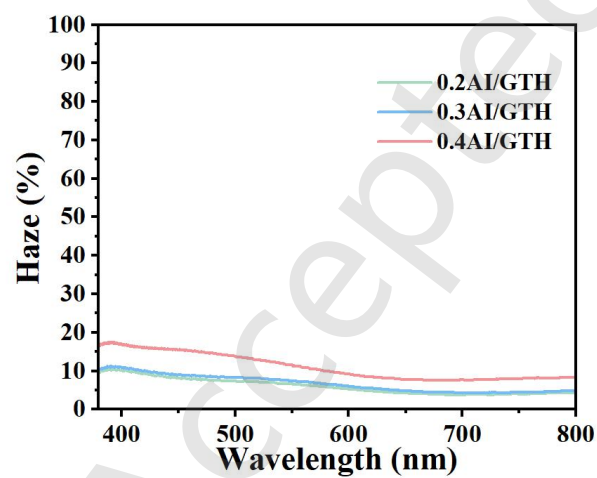


Fig. S29 Haze spectra of Al/GTH composite films over the wavelength range of 380-800 nm.

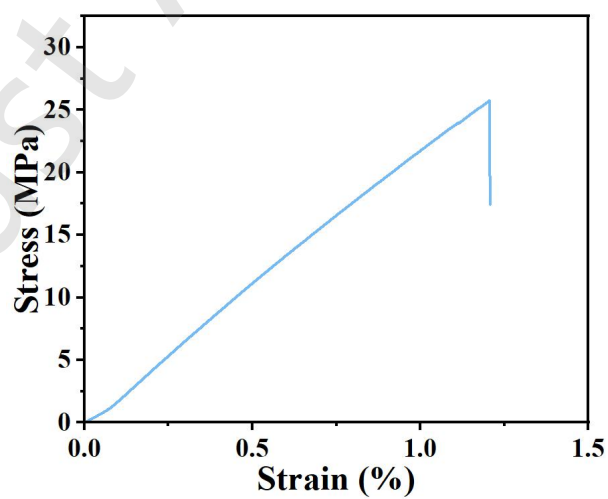


Fig. S30 Tensile stress-strain curve of the 0.4Al/GTH composite film.

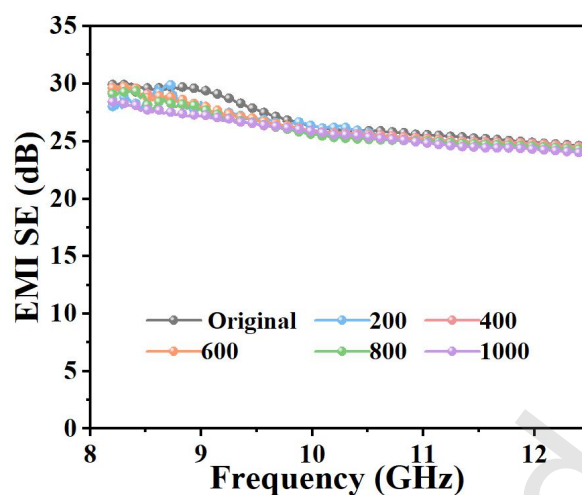


Fig. S31 EMI SE of the 0.4Al-GTH composite film after various bending cycles

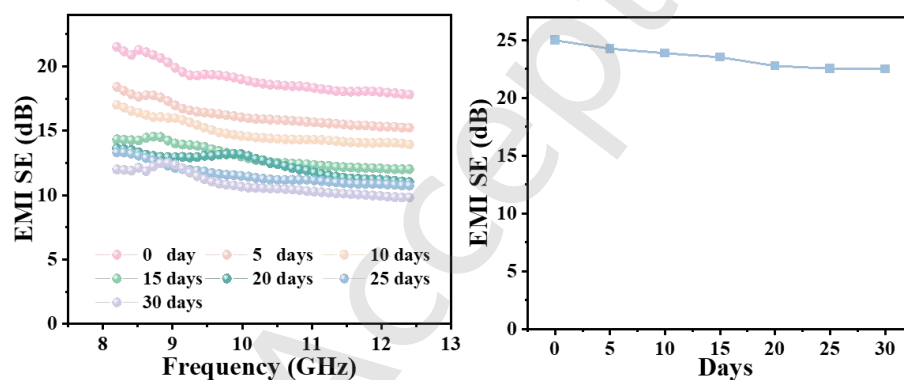


Fig. S32 EMI SE of 0.4Al/GTH composite films measured at 5-day intervals over a period of 30 days.

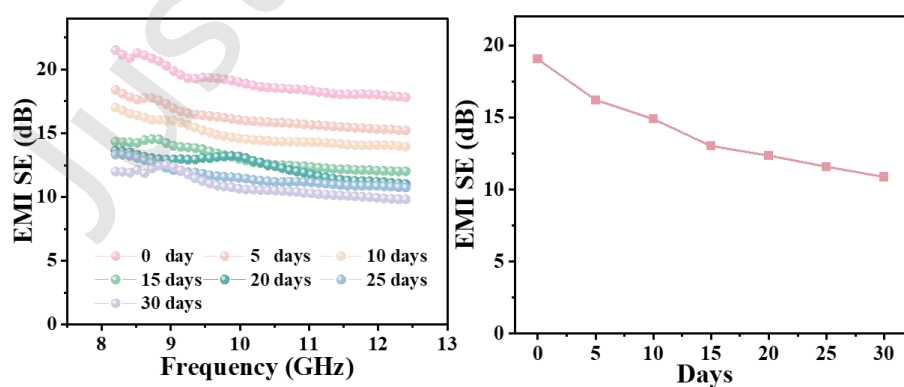


Fig. S33 EMI SE of AgNWs composite films measured at 5-day intervals over a period of 30 days.

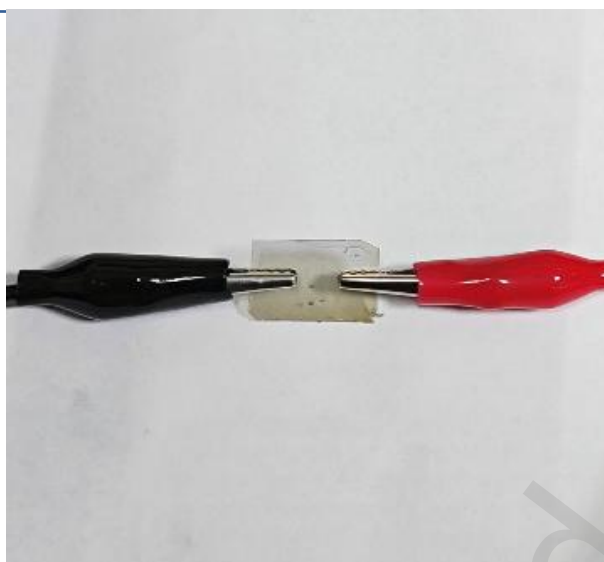


Fig. S34 Photograph of the Joule heating measurement setup for the AI/GTH film.

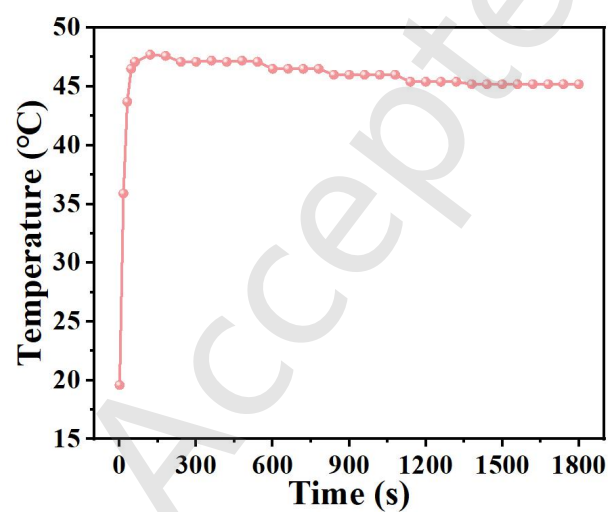


Fig. S35 Time-dependent heating stability for a long time at 4 V.

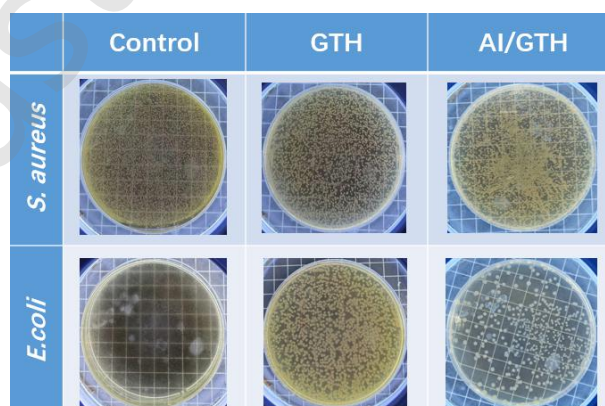


Fig. S36 Digital photographs demonstrating the antibacterial properties of the GTH film and AI/GTH hybrid film against *E. coli* and *S. aureus*.

Table S1 Summary of tensile strength and Young's modulus for representative bioplastics and non-bioplastics.

Materials	Stress (MPa)	Young's modulus (MPa)	Ref.
PBA@PS25	8.4	350.6	[85]
ALP2-5	18	484	[86]
AG	61.31	1170	[10]
PBS	30.7	178.8	[87]
BOPVA	52.4	1380	[88]
PDK-4%	70	1800	[89]
BPP/SF	27.8	3.9	[11]
ENR	62.3	1293	[90]
MReB	21.5	4630	[91]
FPB2-30	58.24	1.65	[92]
This work	77.61	2022.2	

Table S2 Comparison of the coefficients of thermal expansion (CTE) of various materials.

Materials	CTE (ppm K ⁻¹)	Ref.
PC	60-70	[93]
PPO	40-60	[93]
PET	20-50	[93]
CAA-DDPN	0.9-1.8	[51]
BPF-DABA	51-59	[94]
AMDA/DABA	2.6	[95]
PLA	46.2	[96]
PLA/FIB	88.4	[96]
PI-CBDA	7.3	[97]
BT/PP	20.6	[98]
WA121	14.61	[99]
GTH	0.5-0.55	

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