

Sequential Covalent Locking of Cellulosic Eutectic Systems Enables Transparent High-strength and Luminescent Engineering Glasses

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Electronic Supplementary Information

Abstract The fabrication of functional cellulose engineering solids holds great promise for sustainable optics but is severely hindered by the processing paradox imposed by its recalcitrant hydrogen-bonded architecture. To overcome this bottleneck, an orchestrated sequential covalent molecular locking strategy has been reported to transition dynamic cellulosic eutectic melts into robust luminescent glass. Utilizing a cellulose acetate butyrate/thymol eutectic plasticization platform, primary anchoring was implemented *via* isocyanate chemistry, followed by *in situ* 3D crosslinking. This topological reconfiguration induces microscopic network densification and network vitrification, leading to enhanced tensile strength (from about 10.2 MPa to about 55.3 MPa), >90% transparency, and high hydrolytic stability. Mechanistically, the dense network establishes extreme space confinement, triggering the restriction of molecular motion to suppress nonradiative decay and activate efficient solid-state luminescence with programmable multicolor outputs. This deconstruction–locking–activation approach offers a versatile chemical method for developing next-generation high-performance biomass-derived functional materials.

Keywords Cellulosic eutectic systems; Sequential covalent locking; Restriction of molecular motion; Luminescent glass; High-strength

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INTRODUCTION

As the most abundant natural polymer on Earth, cellulose features unique β -1,4-glycosidic bonds that form highly regular linear molecular chains, which are further crosslinked into stable supramolecular architectures *via* dense intra- and inter-molecular hydrogen-bonding networks.^[1–6] While this inherent rigidity provides essential mechanical support for plant fibers, it simultaneously acts as a formidable physical barrier to high value-added processing. From a thermodynamic perspective, the exceptionally high lattice energy of cellulose causes its melting point to exceed its decomposition temperature; kinetically, the highly ordered crystalline regions resist penetration by the solvent molecules. This processing paradox results in cellulose predominantly serving as a low-content filler phase or passive solute in existing material systems.^[7–11] Subverting this hydrogen-locked physical barrier while preserving the intrinsic mechanical advantages of the cellulose backbone remains a central challenge in transforming intractable biomass into active engineering frameworks.

In the pursuit of processing breakthroughs, deep eutectic solvents (DESs) have garnered significant attention for their

exceptional capacity for hydrogen-bond dissociation.^[12–16] Consequently, traditional studies treating cellulose as a solute in a DES matrix are severely hindered by the solubility ceiling (about 20 wt%),^[17–22] limiting the performance boundaries of the resulting hybrids. To bypass this constraint and establish a completely homogeneous liquid-phase medium necessary for precise molecular anchoring without the use of specialized volatile solvents, we leveraged the intrinsic plasticization of cellulose acetate butyrate (CAB) as macromolecular hydrogen bond donors to form eutectic systems with thymol (Thy).^[23] This paradigm shifts the propelled CAB loading to approximately 80 wt%, enabling solvent-free thermoplastic processing of the biomass bulk. Nevertheless, while this deconstruction process imparts the necessary fluidity, the accompanying increase in free volume between molecular chains leads to a decline in the cohesive energy density. Consequently, the resulting materials often manifest as flexible films (strength of about 10.2 MPa), failing to meet the rigorous demands for the high hardness and strength required for engineering-grade glass.

Beyond the mechanical limitations, integrating stable and efficient optical functionality into cellulosic eutectic systems presents formidable challenges. Traditionally, luminescent cellulose has been prepared by chemically grafting specific functional groups onto a polymer backbone.^[24–26] However, these covalent modification strategies are often encumbered

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by intricate and time-consuming synthesis protocols and use of organic solvents. In contrast, leveraging the inherent functional richness and designability of DESs offers a greener and more versatile platform for modular assembly of cellulose-based luminescent materials. Recent efforts have utilized fluorescent DES-mediated pathways to functionalize cellulose nanocrystal assemblies or to enable luminescence through physical doping.^[27,28] Nevertheless, the performance of these hybrid systems remains highly sensitive to the dynamic and flexible nature of the plasticizing matrix, which limits their application in the field of rigid engineering materials. Furthermore, the intrinsic hygroscopicity of the traditional DES/cellulose hybrids renders their optical properties highly susceptible to atmospheric moisture and solvent leaching. Therefore, reconciling the green processing advantages of DES with a transition into an environmentally immune, ultra-rigid framework remains a pivotal challenge for the realization of high-performance cellulosic luminescent materials.

In this study, a robust and sustainable molecular design was achieved using a stepwise chemical-anchoring strategy. This intrinsic CAB/Thy eutectic fluidization converts the intractable biopolymer into a highly accessible liquid-phase reaction template, transitioning the subsequent isocyanate anchoring from an inefficient heterogeneous solid-liquid boundary into a uniform, high-yield homogeneous addition process. This approach is initiated by primary anchoring through nucleophilic addition between a 2-isocyanatoethyl acrylate (IEA) and the residual hydroxyl groups on the CAB backbone, followed by *in situ* incorporation of glycidyl methacrylate (GMA) to construct a dense 3D topological crosslinked network. Through this sequential locking mechanism, a precise transformation from a dynamic eutectic melt to rigid transparent glass was successfully facilitated. Macroscopically, this structural reconfiguration induces a dramatic leap in tensile strength from about 10.2 MPa to about 55.3 MPa; microscopically, a radical collapse of the system's free volume is triggered. The resulting exceptionally compact space-confinement environment effectively seals the non-radiative dissipation pathways of luminescent precursors, thereby activating highly efficient radiative transitions under ambient rigid conditions. Engineered cellulosic luminescent glass (CLG) combines high optical transparency, robust hydrolytic stability, and tunable multicolor emission, thereby offering a promising conceptual framework for the development of sustainable functional optical materials.

EXPERIMENTAL

Materials

Cellulose acetate butyrate (CAB, butyryl content 50%–54%), thymol (Thy, 98%), 2-isocyanatoethyl acrylate (IEA, 98%), and functional naphthalic anhydride derivatives, including 1,8-naphthalic anhydride (NA, 98%), 3-hydroxy-1,8-naphthalic anhydride (NA-OH, ≥98%), 4-bromo-1,8-naphthalic anhydride (NA-Br, ≥95%), 4-chloro-1,8-naphthalic anhydride (NA-Cl, 95%), 4-nitro-1,8-naphthalic anhydride (NA-NO₂, 95%), and 4-amino-1,8-naphthalic anhydride (NA-NH₂, 95%), were purchased from Shanghai Macklin Biochemical Co., Ltd. Glycidyl methacrylate (GMA) (97%) and the photoinitiator diphenyl(2,4,6-trimethylbenzoyl)phosphine oxide (TPO, 98%) were obtained from Sigma-

Aldrich. All the reagents were used as received.

Preparation of the CAB/Thy Eutectic Matrix

CAB and Thy were mixed in a fixed mass ratio of 4:1. The mixture was heated at 120 °C under continuous stirring for 2 h until a clear, transparent eutectic mixture was formed, indicating the intrinsic plasticization of the CAB framework.

Primary Chemical Anchoring

The IEA was introduced into the CAB/Thy mixture. For luminescent samples, a specific naphthalic anhydride derivative (0.2 wt% of the total mass) was added simultaneously. The system was stirred at 120 °C for an additional 2 h to ensure the quantitative nucleophilic addition between the isocyanate groups of IEA and the residual hydroxyl groups on the CAB chains, effectively anchoring the reactive vinyl groups and luminophores onto the CAB backbone.

Secondary Network Formulation

GMA and the photoinitiator, TPO (1 wt% of the total mass) were added to the mixture. The mixture was further stirred at 120 °C for 2 h to achieve a homogeneous monomer-polymer melt.

Photopolymerization and Vitrification

The hot homogeneous melt was cast into a preheated polytetrafluoroethylene (PTFE) mold. The samples were then subjected to UV irradiation (365 nm, 100 mW/cm² for 5 min). Rapid photopolymerization of the acrylate and methacrylate groups triggered the final vitrification, resulting in a hard, transparent CLG.

Characterizations

The chemical evolution of the stepwise anchoring process was monitored using Fourier transform infrared spectroscopy (FTIR) (Bruker Vertex 33). The disappearance of the characteristic isocyanate band at 2266 cm⁻¹ and epoxy signals were tracked. ¹H-NMR spectra were obtained on a Bruker AVANCE III HD 400 spectrometer. Solid-state ¹³C-NMR spectra were obtained using a Bruker AVANCE NEO 600 spectrometer. The amorphous nature of CLG was confirmed using X-ray diffraction (XRD). Thermogravimetric analysis (TGA) was performed from 40 °C to 600 °C at a heating rate of 10 °C/min under a N₂ atmosphere to evaluate thermal stability. Surface hydrophobicity was quantified using water contact angle (WCA) measurements (Dataphysics OCA15EC). The surface morphologies of the samples on the microsubstrates were measured *via* tapping-mode atomic force microscopy (AFM, Bruker Multimode 8) using tapping MPP-rotated cantilevers with silicon probes (model: RTESP, part MPP-11100-10). Dynamic mechanical analysis (DMA) was conducted in tensile mode on a rheometer (HAAKE Mars 60, Thermo Fisher Scientific, Germany) to determine the storage (G') and loss (G'') moduli and the glass transition temperature (T_g). Based on the kinetic theory of rubber elasticity, the topological crosslinking density (ν_e, mol/m³) was calculated quantitatively using the following Eq. (1):^[29,30]

$$\nu_e = \frac{G'}{3RT} \quad (1)$$

where G' is the storage modulus of CLG in the rubbery plateau region at T_g+30 °C, R is the gas constant (8.314 J/(mol·K)), and T is the absolute temperature (the temperature at which G' is taken).

Optical transmittance was measured using a UV-Vis spectrophotometer (Agilent Cary 60) in the range of 400–800 nm.

Steady-state photoluminescence and excitation spectra were recorded to determine the emission characteristics of different NA-X-doped CLG samples. Steady-state photoluminescence (PL) and excitation spectra were recorded to determine the emission characteristics of the different NA-X-doped CLG samples. The absolute photoluminescence quantum yield (Φ_{PL}) values were experimentally measured using a high-sensitivity fluorescence spectrometer equipped with an integrating sphere accessory (FluoroMax-4 equipped with a Quanta-phi integrating sphere; HORIBA Scientific, Japan). Time-resolved photoluminescence (TRPL) measurements were utilized to record the fluorescence decay curves and determine the average fluorescence lifetimes (T). Using the experimentally measured absolute Φ_{PL} and the derived T , the radiative rate constant (k_r) and non-radiative rate constant (k_{nr}) were quantitatively calculated according to the standard photophysical Eqs. (2) and (3):^[31,32]

$$k_r = \frac{\Phi_{\text{PL}}}{T} \quad (2)$$

$$k_{\text{nr}} = \frac{1 - \Phi_{\text{PL}}}{T} \quad (3)$$

Positron annihilation experiments were performed using a fast-fast coincidence system (ORTEC fast-fast composite lifetime spectrometer, United States of America (USA)) with a ^{22}Na positron source and an activity of approximately 10 μCi . The membranes were cut into slices of 1.0 cm $\times$ 1.0 cm, with thicknesses of approximately 1.2 mm. A total of 10^6 cells were collected from each sample. After acquiring the positron annihilation lifetime spectra, the spectra were analyzed by fitting with LT software, yielding the lifetime components and their corresponding intensities.

Tensile properties were assessed using an INSTRON 5565 machine (USA) at a strain rate of 10 mm/min. Nanoindentation tests were performed to evaluate the surface hardness. The adhesive performance was quantified *via* lap-shear tests on various substrates, such as poly(methyl methacrylate) (PMMA), poly(vinyl chloride) (PVC), poly(ethylene terephthalate) (PET), polycarbonate (PC), poly(laurylamide) (PA12), and polystyrene (PS), following the ASTM D1002 standard. All tests were conducted in triplicates to ensure reproducibility.

RESULTS AND DISCUSSION

Stepwise Construction Strategy and Bio-inspired Logic of CLG

The molecular design of this study is rooted in the intrinsic plasticization framework of CAB/Thy. As a highly efficient small-molecule hydrogen-bond regulator, Thy deconstructs the inherent dense physical interactions of natural cellulose by establishing dynamic H-bonds with CAB segments. This facilitated the solvent-free transformation of high-loading biomass components from solid mixtures into homogeneous, transparent eutectic melts (Fig. S1 in the electronic supplementary information, ESI). Nevertheless, such dynamic physical networks are encumbered by intrinsic challenges, including low cohesive energy density, insufficient mechanical strength, and excessive free volume, which hinder the stable integration of the functional units. Consequently, we propose a stepwise chemical anchoring strategy aimed at reconfiguring network topology through

covalent locking.

This design was fundamentally inspired by the precision of natural bioluminescent systems, such as bioluminescent fungi, which leverage sophisticated microenvironments within enzyme active sites to stabilize reaction intermediates and catalyze high-efficiency chemiluminescence (Fig. 1a).^[33,34] We recognized that the artificial construction of a rigid microenvironment, which exerts spatial confinement on luminophores, could similarly suppress nonradiative dissipation. By mimicking this biological confinement paradigm, we sought to integrate high-efficiency photoluminescence functionality into our synthetic framework. In this system, the CAB backbone not only serves as a macromolecular structural support but also provides residual hydroxyl sites for subsequent chemical anchoring. 1,8-Naphthalic anhydride derivatives (NA-X) act as luminescent precursors that are highly sensitive to the local microenvironment, and their activation in quantum efficiency is critically dependent on matrix rigidity.

The stepwise construction pathway is illustrated in Fig. 1(b) and Table S1 (in ESI). Initially, a CAB/Thy cellulosic eutectic melt was used as the reaction medium. Subsequently, IEA was introduced, leveraging the high reactivity of its $-\text{NCO}$ groups for nucleophilic addition to the hydroxyl groups of the CAB backbone. This process pre-organizes polymerizable vinyl groups around the CAB chains while simultaneously achieving covalent grafting of luminescent precursors. Finally, GMA and the photoinitiator TPO were incorporated to construct a dense 3D topologically crosslinked network *via* UV-induced radical polymerization (Fig. S2 in ESI). This strategy facilitates precise evolution from a dynamic physical melt to a hard, transparent vitreous material. To verify that this sequential architecture is paramount to achieving network densification rather than simple comonomer mixing, we directly contrasted this multistep pathway with a conventional one-step polymerization control. The structural necessity of this sequential pathway was directly corroborated by solid-state ^{13}C NMR, *in situ* photo-rheology, and strain-stress curves (Figs. S3, S4 and Table S2 in ESI). The sequential prepolymer achieved a curing storage modulus plateau of about 10^7 Pa (versus about 10^5 Pa for the one-step control) and enhanced ^{13}C -NMR cross-polarization peak intensity, directly confirming that primary covalent anchoring prevents comonomer phase separation and establishes a rigid, highly confined topological vitrified network.

To complement this macro-topological validation and trace the molecular transformations along the sequential pathway, step-by-step chemical group conversions were systematically monitored using FTIR and ^1H -NMR spectroscopy (Figs. S5 and S6 in ESI). No new chemical bond signatures were observed during the initial formation of the eutectic system, confirming the physical interaction-dominated nature of the intrinsic plasticization process. As the reaction progressed, the characteristic $-\text{NCO}$ peak at 2266 cm^{-1} completely disappeared^[35,36] validating the successful conversion of active functional groups and orchestrated construction of the crosslinked network.

A series of gradient-crosslinked samples were prepared by varying the mass ratio of the base matrix to the comonomers (IEA/GMA). The core scientific value of this construction route

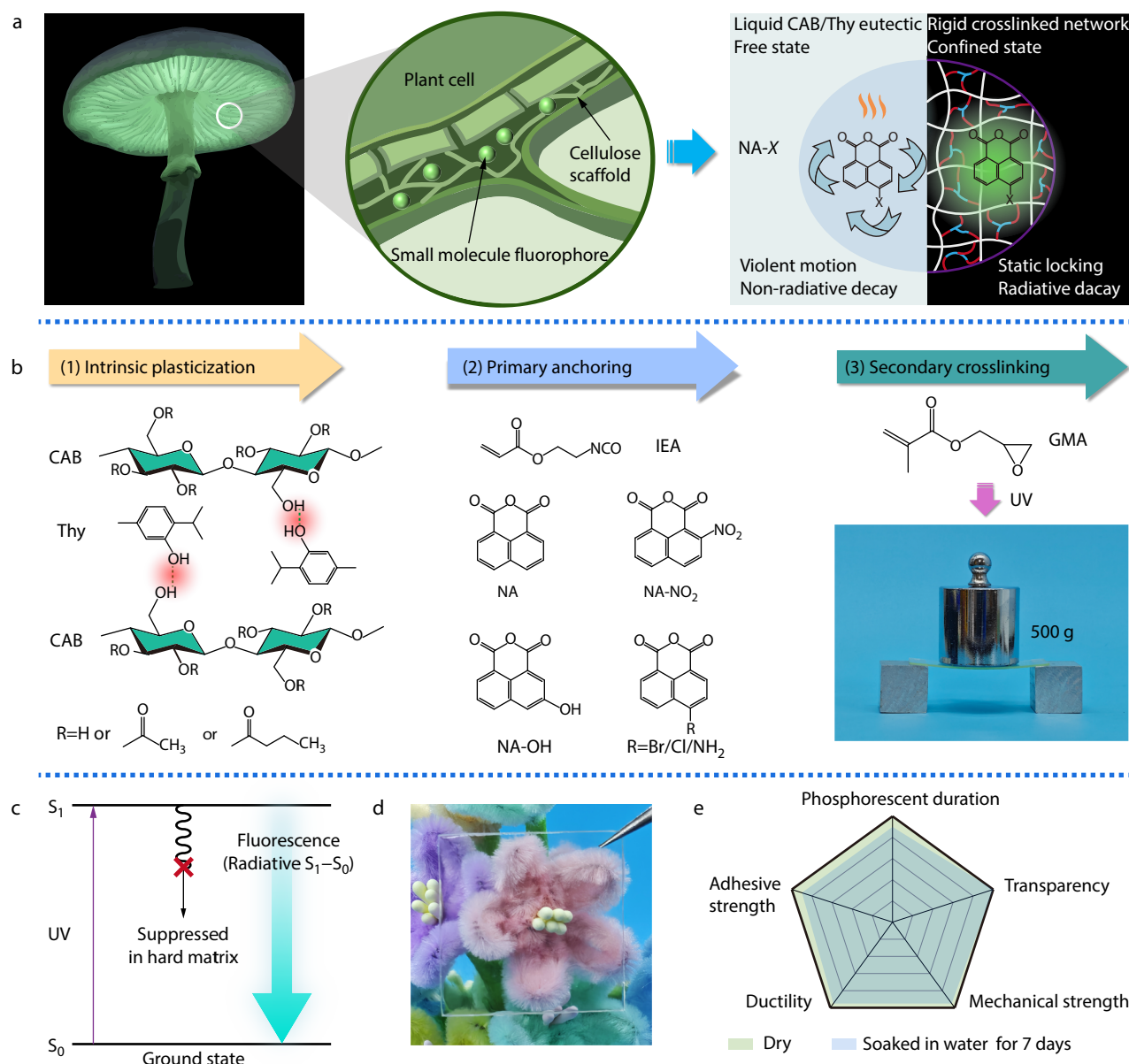


Fig. 1 Bio-inspired design, stepwise synthesis, and comprehensive performance of robust CLG. (a) Bio-inspiration and design philosophy: schematic illustration of the luminescent mechanism inspired by bioluminescent fungi, where the luminescent precursor (NA-X) is activated through space confinement within a rigid polymer matrix; (b) The intrinsic plasticization of CAB is achieved via H-bonding with Thy, followed by the sequential incorporation of functional monomers (IEA, NA-X, and GMA). The final material is obtained via UV-triggered *in situ* polymerization. Optical photograph showing the high stiffness of the material supporting a 500 g weight without noticeable deformation. (c) Schematic representation of the NA-X molecules trapped in the rigid crosslinked network, facilitating radiative transition via the RMM effect; (d) Optical photograph demonstrating the superior transparency of the fabricated CLG; (e) Radar chart of multidimensional performance: evaluation of the comprehensive properties including transparency, mechanical strength, ductility, adhesive strength, and luminescence. The evaluation highlights the exceptional hydrolytic stability maintained even after a 7-day immersion in water.

lies in its departure from the inhomogeneity typical of simple physical doping. Instead, it utilizes a stepwise approach to establish a compact microenvironment where mechanical robustness and luminescence are synergistically enhanced. Fig. 1(c) reveals the physical essence of this luminescent activation: in liquid or highly dynamic matrices, NA-X molecules possess high degrees of freedom for internal rotation and vibration, causing excited-state energy to dissipate primarily through non-radiative pathways (quenching). In contrast,

within the crosslinked rigid network, the restriction of molecular motion (RMM) effect, underpinned by space confinement, effectively suppresses these nonradiative dissipation pathways, thereby triggering stable solid-state luminescence.

This multistage locking mechanism endows the resulting CLG with exceptional comprehensive properties. The XRD analysis confirmed the maintenance of a stable amorphous structure (Fig. S7 in ESI) with superior optical transparency (Fig. 1d and Fig. S8 in ESI). Benefiting from the inhibition of

polar rearrangement by the covalent network, the material exhibits remarkable hydrolytic stability (Fig. 1e), maintaining its transparency, mechanical modulus, and luminescent performance even after 7 days of immersion in water. Furthermore, CLG exhibited negligible swelling and mass loss in a concentrated acid (37 wt% HCl), strong alkali (20 wt% NaOH), and saturated NaCl solution (Fig. S9 in ESI). Combined with the excellent thermal stability confirmed by TGA (Fig. S10 in ESI). These results demonstrate the universal value of the stepwise chemical anchoring strategy for constructing high-performance, environmentally adaptable biomass-based optical engineering materials.

Modulation of Topological Network Evolution and Macroscopic Mechanical Properties via Sequential Covalent Locking

To systematically elucidate the evolution mechanism of the physical properties of the material under the sequential covalent locking strategy, we evaluated the topological network morphology and mechanical response for various component ratios. Fig. 2(a) shows the mechanical disparity between the single-monomer control systems and the synergistic dual-monomer system. Systems incorporating only IEA or GMA failed to surpass the performance bottleneck of the physical network, indicating that a single reaction pathway, whether limited to backbone anchoring or free monomer polymerization, cannot simultaneously satisfy the demands for increased crosslinking density and network integrity. In contrast, the synergistic intervention of IEA and GMA, through an "anchor-first, crosslink-later" modality, induces a qualitative leap in mechanical performance. With increasing comonomer content, the material undergoes a precise transition from ductile plastic (CLG₁₀₀, about 10.2 MPa) to engineering-grade rigid glass (CLG₆₀, about 55.3 MPa) (Fig. 2b, Fig. S11 and Table S3 in ESI). This increase in strength is attributed to the significantly enhanced cohesive energy density of the system. The comprehensive static evaluation firmly validates the immediate structural robustness of the sequentially locked CLG series, and translating this functional biomass platform into broad commercial optoelectronic components and load-bearing applications will naturally warrant further multidimensional investigations. To establish a more exhaustive reliability matrix for long-term engineering deployment, evaluating advanced dynamic viscoelastic behaviors, such as cyclic fatigue propagation and sustained tensile creep resistance, represents a highly valuable and necessary direction for our ongoing work. Integrating these long-term service metrics with the current impressive static strength and rigidity will provide a complete engineering blueprint for governing the lifecycle predictability of these sustainable vitrified networks.

Dynamic mechanical analysis (DMA; Fig. 2c) and differential scanning calorimetry (DSC; Fig. S12 in ESI) consistently showed a systematic increase in the glass transition temperature (T_g) as the degree of covalent locking intensified. This provides robust evidence that the dense covalently crosslinked network effectively suppresses long-range relaxation of macromolecular CAB segments. The stress relaxation experiments (Fig. S13 in ESI) provide direct physical evidence that CLG₁₀₀, which relies primarily on dynamic hydrogen bonding, undergoes rapid chain slippage under stress, leading to precipitous energy dissipation. Conversely, CLG₇₀, bol-

stered by a covalent framework, exhibited a significantly slower stress decay and superior structural integrity. Atomic force microscopy (AFM) phase imaging (Figs. 2d and 2e) revealed a homogeneous morphology across all samples, with no signs of macro-phase separation. The notably higher brightness in the phase map of CLG₇₀ (where brighter regions represent higher hardness) further confirmed the mechanical uniformity and rigidity imparted by the crosslinked network.

The leap in macroscopic performance is rooted in the evolution of microscopic architecture. Small-angle X-ray scattering (SAXS, Fig. 2f) results indicated a significant rightward shift of the characteristic scattering peak for CLG₇₀ relative to CLG₁₀₀, with the scattering vector magnitude q -value increasing from about $0.125 \text{ \AA}^{-1}$ to about $0.183 \text{ \AA}^{-1}$. Based on the relationship characteristic spatial distance $d \approx 2\pi/q$, the characteristic correlation length (interchain spacing) within the system is markedly reduced, reflecting the mesoscopic structural domain compaction and network densification induced by the sequential reaction. To unambiguously quantify the sub-nanometer free-volume evolution beyond the mesoscopic SAXS correlation metrics, positron annihilation lifetime spectroscopy and bulk density measurements were conducted (Fig. S14 and Table S4 in ESI). Applying the Tao-Eldrup model to the long-lived ortho-positronium (o-Ps) pick-off lifetime τ_3 ,^[37] the average free-volume cavity volume (V_f) exhibits a distinct shrinkage from $155.2 \text{ \AA}^3$ ($R_h = 3.33 \text{ \AA}$ in CLG₁₀₀) down to $118.5 \text{ \AA}^3$ ($R_h = 3.05 \text{ \AA}$ in CLG₇₀). Coupled with a reduction in void intensity (I_3 from 33.70% to 30.46%), the overall relative free-volume fraction underwent a substantial 31.0% collapse upon network vitrification. This sub-nanometer volumetric compaction increases the local electron density—as evidenced by a drop in average lifetime (τ_{average} from 1107.82 ps to 929.14 ps)—and a macro-density elevation to 1.13 g/cm^3 . This multi-scale structural densification constructs a compact space-confinement environment, which not only yields a macroscopic rigid skeleton but also provides an ideal physical capsule for the subsequent RMM of luminescent materials.

Furthermore, network densification substantially enhances the engineering reliability. As shown in Fig. 2(g), the static water contact angle increases progressively with crosslinking density, indicating that the reduced surface energy and restricted segment mobility effectively bolster the intrinsic hydrophobicity of the material. Accordingly, CLG₇₀ exhibits negligible fluctuations in its stress-strain response after a 7-day underwater immersion compared with its initial state (Fig. 2h), proving that the highly integrated covalent network effectively seals the infiltration pathways for water molecules. Variable-temperature mechanical tests (Fig. S15 in ESI) demonstrated that the material maintained a substantial load-bearing capacity across a broad temperature range, reflecting excellent thermomechanical adaptability. Nano-indentation tests (Fig. 2i) and falling-ball impact experiments (Figs. 2j and 2k) further show its potential as a high-performance protective coating. Compared to untreated substrates prone to plastic deformation or brittle failure, the CLG coating effectively buffers the impact kinetic energy and suppresses crack propagation. This series of experimental evidences reveals the essence of the sequential covalent locking

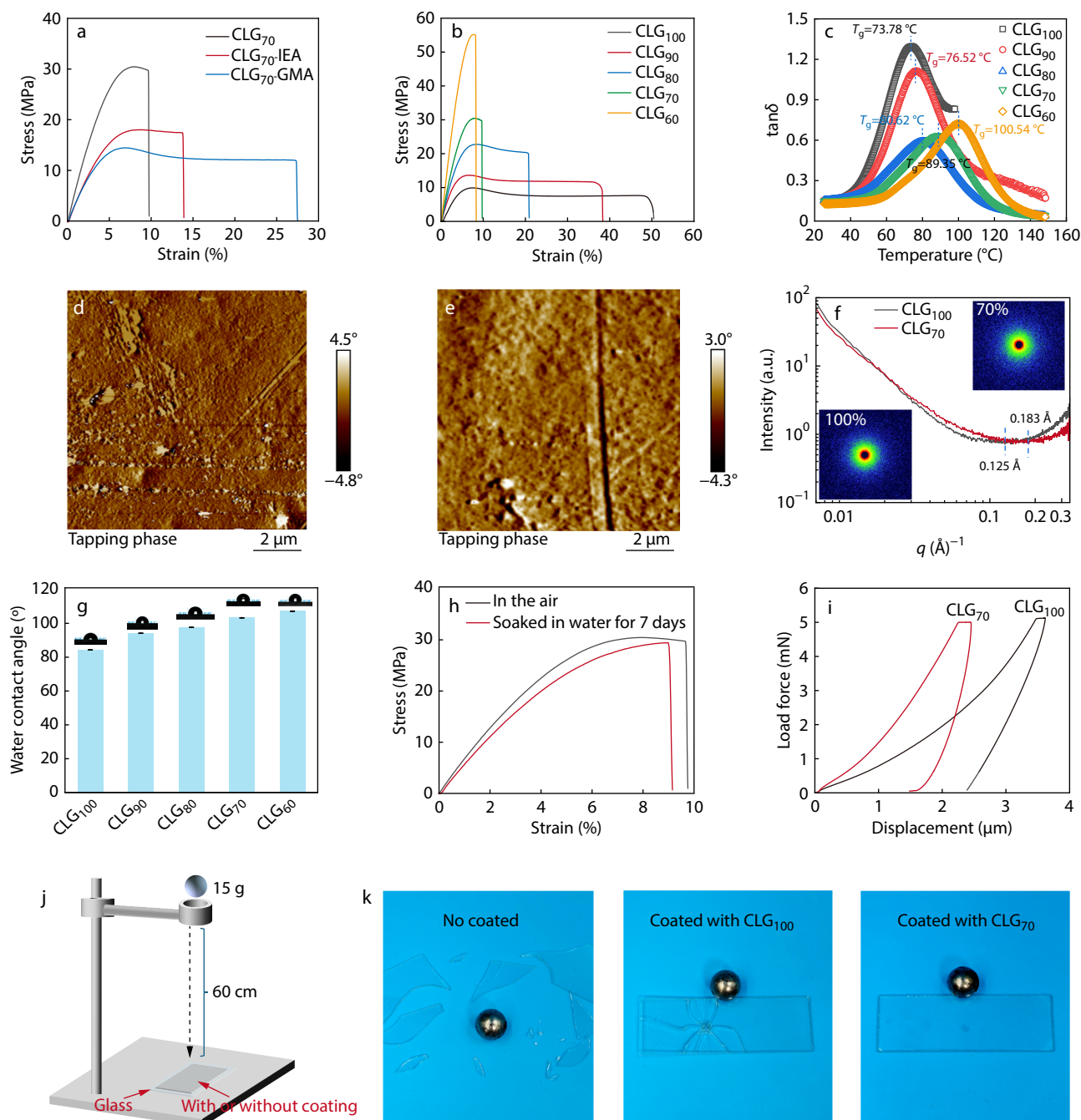


Fig. 2 Modulation of multi-scale mechanical properties and environmental stability of CLGs *via* topological network regulation. (a) Comparison of stress-strain curves between single-monomer control systems and the synergistic IEA/GMA comonomer system; (b) Stress-strain curves of various formulations obtained by varying the mass ratio of the base matrix (CAB/Thy) to comonomers; (c) DMA revealing variations in $\tan\delta$ and the significant elevation of T_g ; (d, e) AFM phase images of CLG₁₀₀ and CLG₇₀ samples; (f) SAXS patterns characterizing the contraction of internal structural correlation lengths upon crosslinking; (g) Static water contact angles of different samples; (h) Comparison of strain-stress curves for CLG₇₀ samples tested in ambient air and after a 7-day immersion in water; (i) Nano-indentation load-displacement curves; (j, k) Falling-ball impact tests, visually demonstrating the improved substrate protection provided by the CLG₇₀ coating compared to the control group.

strategy: it achieves a continuous evolution from a dynamic physical network to a high-performance engineering glass by constructing a high-density topological network in a step-wise manner, while preserving the processability of the cellulosic eutectic system. This structural transformation not only endows the material with exceptional strength, hardness, and environmental resistance but also establishes a physical four-

dimensional network for achieving efficient and stable light-activated functions at the molecular scale.

Rigid Matrix-induced RMM and Tunable Multi-color Emission

Having established a high-strength, high-transparency CLG matrix, its core scientific value lies in whether the rigid crosslinked

architecture can provide an effective space-confinement environment for chromophores, thereby activating otherwise inhibited solid-state luminescence. To this end, we designed and synthesized a series of 1,8-naphthalic anhydride derivatives (NA-X) with various substituents as model luminescent units, which were integrated into the CAB/Thy/IEA/GMA topological network *via* a stepwise chemical anchoring strategy.

Fig. 3(a) illustrates a striking fluorescence turn-on effect: both the pristine NA-Br powder and liquid CLG prepolymer exhibit a quenched state under UV light (365 nm). However, upon UV-triggered vitrification of the system, the resulting rigid CLG samples instantaneously emitted intense visible fluorescence. This contrast provides robust evidence that the activation of luminescence is not merely a consequence of luminophore incorporation but is deeply dependent on the

rigid microenvironment established after photopolymerization. Corroborated by the FTIR evidence (Fig. S16 in ESI), the covalent anchoring mechanism effectively locks NA-X molecules within the dense 3D topological grid. This extreme spatial compression triggers the RMM effect, which significantly suppresses the intramolecular rotation and vibration of chromophores, thereby effectively sealing non-radiative transition pathways and prioritizing radiative decay for the excited-state energy.

The CLG platform exhibits not only potent activation capacity but also exceptional molecular-to-macroscopic color delivery. As shown in Fig. 3(b), broad-spectrum multicolor emission, ranging from deep blue to green, is achieved by precisely tailoring the substituents on the NA-X backbone (e.g., $-\text{NH}_2$, $-\text{OH}$, $-\text{NO}_2$, $-\text{Br}$, $-\text{Cl}$). This color programma-

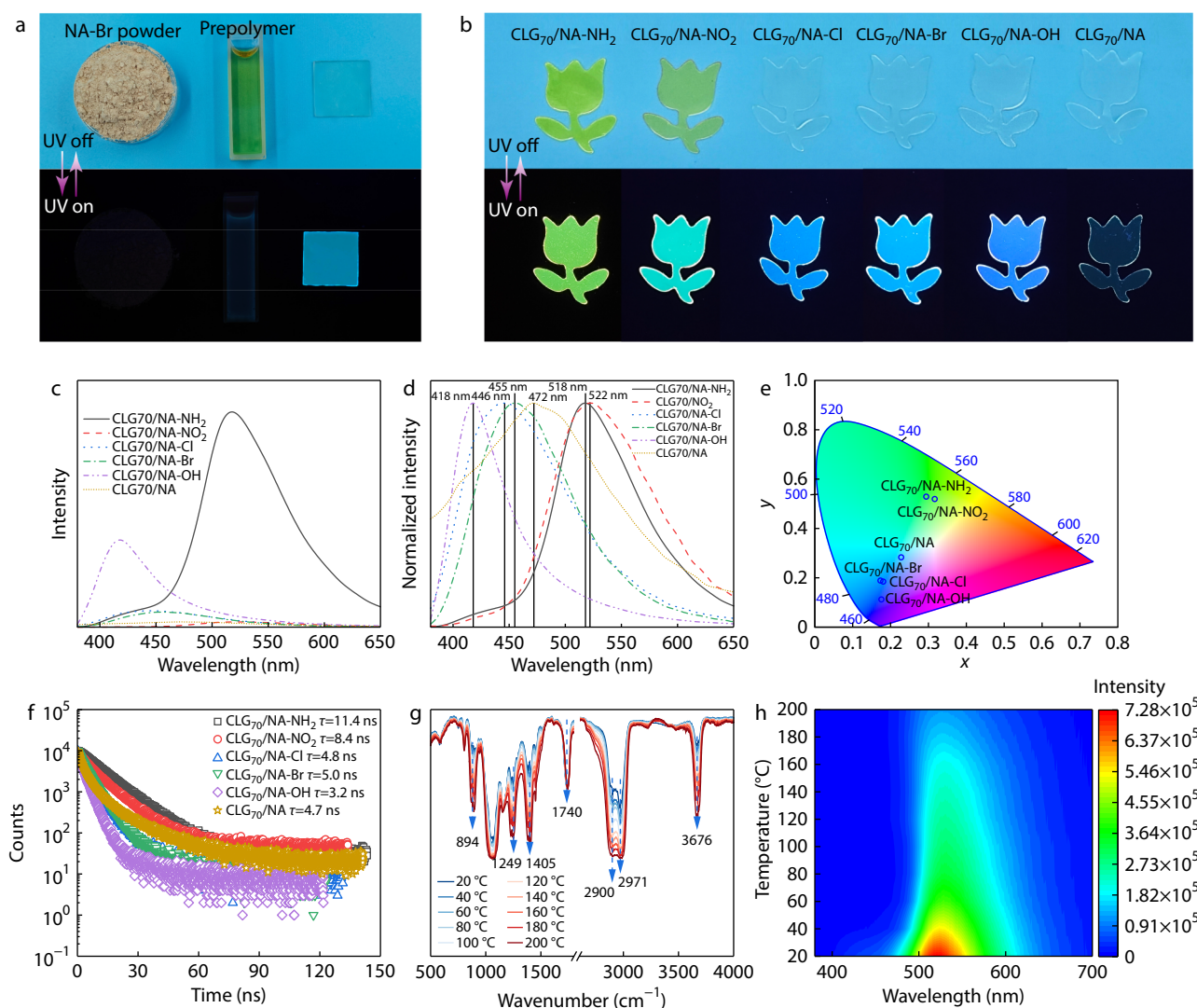


Fig. 3 Validation of rigid-network-dominated luminescence *via* RMM and tunable multi-color fluorescence. (a) Visual comparison: pristine NA-Br powders and liquid CLG prepolymers exhibit no noticeable luminescence, while the CLG sample emits intense blue fluorescence under 365 nm excitation; (b) Comparison of CLG samples incorporated with various NA-X derivatives under ambient light (top) and UV light (bottom), demonstrating multi-color emission tunability; (c) Comparison of fluorescence intensities for CLG samples with different NA-X; (d) Solid-state fluorescence emission spectra of various CLG samples; (e) CIE chromaticity coordinates; (f) Time-resolved fluorescence decay curves of different CLG samples under 365 nm UV irradiation; (g) Variable-temperature FTIR spectra of CLG₇₀/NA-NH₂; (h) Variable-temperature fluorescence intensity patterns of CLG₇₀/NA-NH₂.

bility stems from the systematic modulation of molecular conjugation, internal charge transfer characteristics, and local polar responses owing to the varied electronic structures of the substituents. The fluorescence spectra in Figs. 3(c) and 3(d) quantitatively reveal these electronic effects, where different NA-X variants exhibit distinct emission efficiencies and peak shifts under identical space-confinement conditions. The CIE chromaticity diagram (Fig. 3e) further confirms the extensive gamut coverage of this system, demonstrating that the logic-based programming of biomass-based optical materials can be realized through simple luminescent plug-in replacement without altering the host matrix or processing protocols.

To seek further dynamic evidence of the space-confinement effect, we analyzed time-resolved photoluminescence decay curves (Fig. 3f). Combined with absolute quantum yield (Φ_{PL}) measurements, the radiative (k_r) and nonradiative (k_{nr}) rate constants were rigorously extracted (Table S5 in ESI). Crucially, a quantitative inverse correlation is established between network topology and non-radiative decay: as the crosslinking density (v_c) elevates from 34.53 mol/m³ (CLG₈₀/NA-NH₂) to 89.52 mol/m³ (CLG₆₀/NA-NH₂), k_{nr} undergoes a monotonic reduction from 0.0825 ns⁻¹ to 0.0511 ns⁻¹ (alongside a k_r gain from 0.0175 ns⁻¹ to 0.0323 ns⁻¹), quantitatively proving that topological compaction suppresses non-radiative energy loss. Direct vibrational evidence for this spatial confinement was validated *via* static comparative FTIR spectroscopy across the gradient glasses (Fig. S17a in ESI). With increasing v_c , the characteristic absorption bands of the carbonyl stretch (C=O, from 1741 cm⁻¹ to 1732 cm⁻¹) and ester backbone (C—O, from 1250 cm⁻¹ to 1230 cm⁻¹) exhibit systematic red shifts, confirming that denser networks impose tighter spatial confinement to restrict segment vibrations. Furthermore, variable-temperature FTIR analysis (25–200 °C, Fig. 3g) revealed an anomalous intensity enhancement of the C=O stretch (1740 cm⁻¹), C—O stretch (1249 cm⁻¹), cellulosic ring deformation (894 cm⁻¹), and C—H stretching (2900/2971 cm⁻¹). Unlike conventional soft polymers that undergo thermal absorbance decay, this thermal intensity gain confirms that these specific skeletal and carbonyl modes are deeply frozen under ambient spatial confinement, directly underpinning the suppression of nonradiative dissipation pathways. Complementing the temperature-dependent rheology (Fig. S17b in ESI) and fluorescence (Fig. 3h) experiments provide critical thermo-responsive evidence; as the temperature increases, the microscopic rigidity of the local network diminishes, releasing these frozen modes and partially restoring the intramolecular rotational degrees of freedom, which results in a monotonic decrease in fluorescence intensity. This thermosensitivity provides a reciprocal validation of the hypothesis that the rigid network serves as the luminescence engine.

Multidimensional Optical Integration and Engineering Scenario Demonstrations

By leveraging its exceptional mechanical robustness, high transparency, and programmable luminescence, we further explored the application potential of CLG materials in multidimensional visual optics and complex engineering environments. First, CLG demonstrates a formidable capacity for con-

structing high-resolution digital pixel arrays and logic displays (Fig. 4a). Owing to the highly tunable emission colors of the system, luminescent motifs with distinct emission characteristics can be precisely arranged within a single matrix. This information-encoding capability is ascribed to two pivotal factors: (i) the superior rheological plasticity of the prepolymer during the processing stage, which allows for intricate patterned topological arrangements, and (ii) the molecular-scale locking of luminophores by the rigid network upon photopolymerization. This locking mechanism ensures high-definition pixel boundaries and uniform luminescent distribution, essentially eliminating the pattern blurring caused by the migration or physical diffusion of small-molecule luminophores.

Second, the excellent optical homogeneity and structural stability of the material provides a foundation for applications in optical waveguiding and 3D structural molding. As shown in Fig. 4(b), the laser light was transmitted continuously within the CLG filaments *via* total internal reflection. This efficient light-guiding capability stems from the uniform vitreous network of the system, which suppresses scattering losses caused by phase separation, microvoids, and aggregates. This demonstration provides a visionary proof of concept, showing the potential of sustainable topological networks in organic waveguiding applications. Concurrently, the high crosslinking density endows the material with excellent shape-retention capability, enabling its transformation into complex 3D geometric structures *via* molding processes, thereby paving the way for the development of biomass-based structure-function integrated optical components.

Furthermore, this study verified the reliability of CLG as an advanced anti-counterfeiting printing ink (Fig. 4c, Movie S1 in ESI). Compared to traditional fluorescent inks that rely on physical doping, the superiority of the CLG system lies in the covalent anchoring of chromophores within the confined network. This chemical stability imparts extreme leaching resistance to severe deformation or extreme chemical environments. Extreme environmental challenge experiments (Fig. 4d) show that digital pixel arrays maintained nearly unaffected fluorescence intensity and pattern integrity even after continuous immersion in water for 7 days (Fig. S18 in ESI).

Interfacial Engineering and High-performance Adhesive Applications

Leveraging the exceptional initial flow-wettability of the CAB/Thy eutectic system and the dense network derived from *in situ* IEA/GMA crosslinking, CLG has significant potential as a high-performance structural adhesive. The formidable macroscopic lap-shear performance of CLG adhesives across diverse engineering substrates originates from a synergistic combination of excellent bulk cohesive energy and favorable multiscale interfacial interactions. Statistically, the fluid-like wetting kinetics of the CAB/Thy eutectic melt ensured highly conformal boundary contact and effective capillary penetration into the micro-roughness of the substrate. Upon *in situ* UV irradiation, rapid crosslinking locks this wetted interface into a robust mechanical interlocking network. Concurrently, the abundance of ester groups, urethane linkages, and residual hydroxyls within the vitrified network facilitates the establishment of dense dipole-dipole interactions and hydrogen-bonding arrays with the polar segments of PMMA, PC, PVC, and PET substrates, effectively preventing interfacial delamination under mechanical

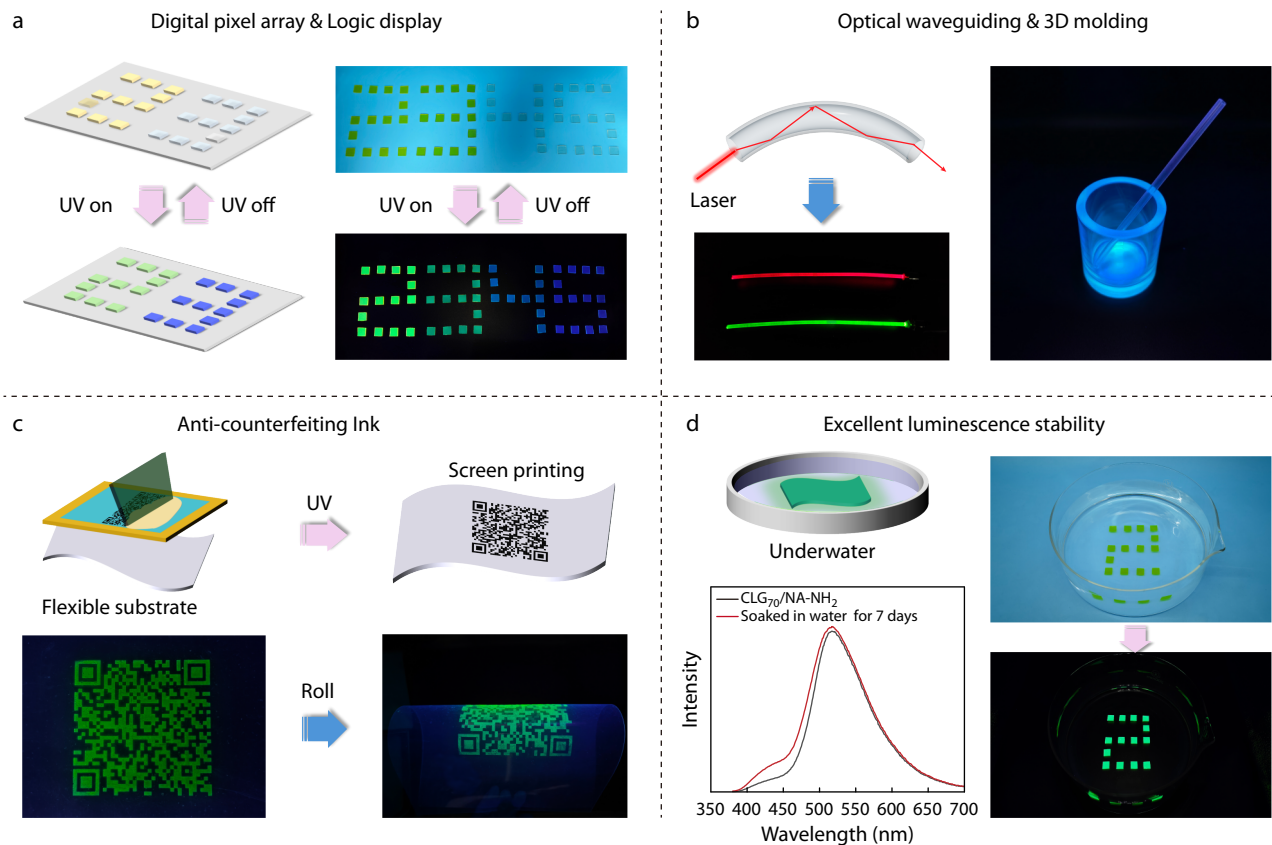


Fig. 4 Multidimensional applications of CLG. (a) Schematic of digital pixel arrays and logic displays; (b) Demonstrations of applications in optical waveguiding devices and 3D molded structures; (c) Example of a foldable anti-counterfeiting QR code printed with fluorescent ink; (d) Comparison of fluorescence intensity before and after a 7-day water immersion, demonstrating exceptional hydrolytic stability under ambient light (top right) and UV light (bottom right).

load.

Fig. 5(a) intuitively validates the superior interfacial load-bearing capacity of the material. Following simple lap-shear bonding and UV-photopolymerization between two PMMA plates, the resulting joint robustly supported the weight of a 97-kg adult. This extreme load test not only reflects the formidable internal cohesive strength of the CLG but also demonstrates its excellent energy dissipation capacity and interfacial fracture toughness under stress. Fig. 5(b) further explores the temperature-responsive behavior of the adhesive strength. As expected, a systematic decrease in bonding strength was observed with increasing temperature (from 25 °C to 100 °C), attributed to enhanced polymer segmental mobility, which leads to a reduction in the viscoelastic modulus and weakening of interfacial cohesion. Nevertheless, CLG maintains substantial residual strength even at elevated temperatures, highlighting the pivotal role of the crosslinked network in maintaining interfacial stability.

Notably, CLG exhibits a remarkable adaptability to aqueous environments. Benefiting from the effective barrier effect of the dense network against water molecules, the material achieves efficient underwater bonding across a diverse range of polar and nonpolar substrates (including PMMA, PVC, PET, PC, PA12, and PS, Fig. 5c). A comparison of lap-shear strengths between ambient air and a 7-day water immersion (Fig. 5d) reveals exceptionally high strength-retention ratios.

This superior hydrolytic stability originates from the shielding effect of the dense topological network, which effectively prevents swelling-induced disruption of interfacial polar bonds by water molecules.

To establish the practical engineering lifespan of the CLG platform for optoelectronic components and load-bearing adhesives, a comprehensive environmental aging assessment was systematically conducted.^[38–40] After prolonged underwater soaking for 21 days, the vitrified matrix sustained its structural integrity while retaining about 50% of its initial luminescence (Fig. S19 in ESI). Remarkably, subjecting the glass to 10 rapid thermal cycles (25–100 °C) and accelerated UV weathering (365 nm, 100 mW/cm² for 72 h) induced an about 30% and about 40% enhancements in fluorescence intensity, respectively. This performance gain is driven by thermal annealing and photo-induced post-crosslinking, which release internal processing stresses and further compact the topological network to intensify space confinement. Furthermore, the glass displayed stable optical properties under damp-heat conditions (40 °C, 85% RH for 48 h) and maintained stable underwater adhesive shear strength over 21 days of continuous immersion (Fig. S20 in ESI). These results collectively demonstrate the superior long-term environmental durability of the CLG architecture under severe operational stresses. Crucially, the juxtaposition of robust structural adhesion and programmable optical properties estab-

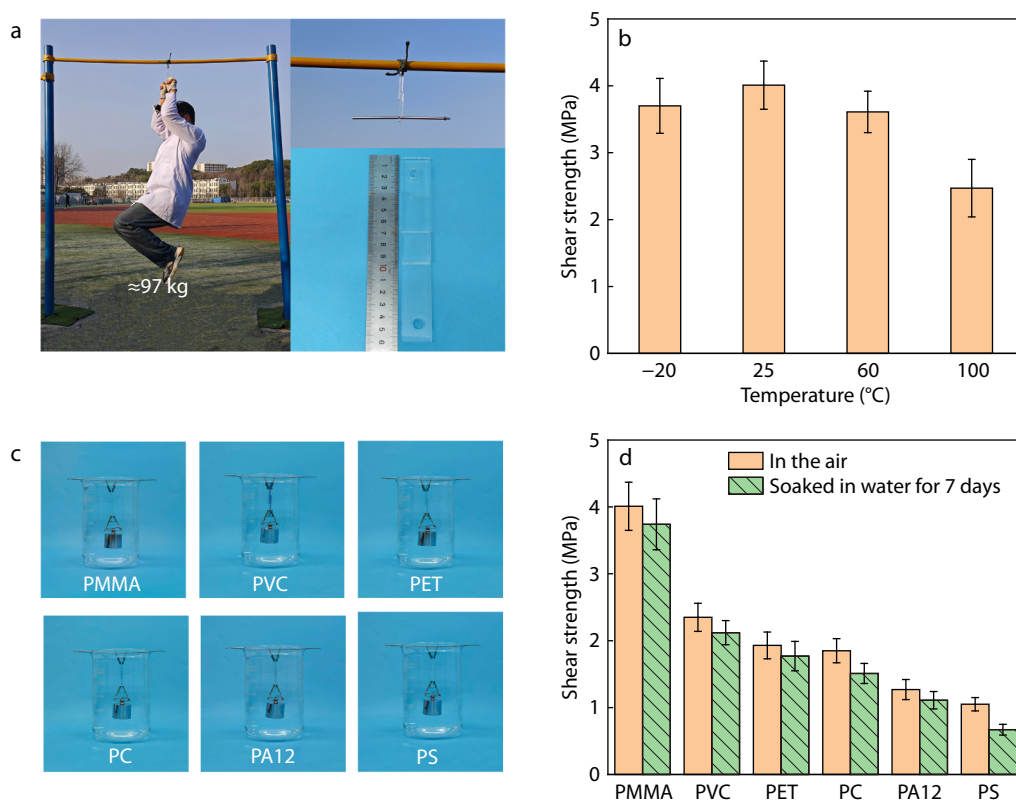


Fig. 5 Applications as high-performance adhesives. (a) Visual demonstration of the material's adhesive load-bearing capacity; (b) Variations in adhesive strength on PMMA substrates at different temperatures; (c) Demonstration of underwater bonding capability on diverse substrates; (d) Lap-shear strengths for various substrates tested in ambient air and after a 7-day water immersion.

lished in this study may address a pivotal challenge in modern advanced optoelectronics: the need for adhesive-free, seamless device assembly. By functioning simultaneously as both a high-strength mechanical joint and an active optical transmission/display medium, the engineered CLG eliminates traditional multi-interfacial scattering losses, offering a general conceptual strategy for integrated photonics and tamper-evident security packaging.

CONCLUSIONS

We demonstrate a sequential covalent molecular locking strategy that transforms dynamic cellulosic eutectic melts into transparent, robust, and luminescent engineering glasses (CLGs). By combining intrinsic plasticization, primary isocyanate anchoring, and subsequent UV photopolymerization, a dense 3D cross-linked topological network was established under solvent-free conditions. Multi-scale evaluations reveal that this sequential process drives sub-nanometer free-volume compaction, as evidenced by a 31.0% collapse in the relative free-volume fraction, and elevates the topological crosslinking density up to 89.52 mol/m³. Consequently, the vitrified glass achieved a high tensile strength of 55.3 MPa and a Young's modulus of 1.12 GPa. Photophysically, the dense network imposes strict spatial confinement on the chromophores, effectively restricting molecular motion and suppressing nonradiative decay rate constants (k_{nr} , reduced to 0.0511 ns⁻¹), thereby activating bright solid-state photoluminescence with tunable multicolor outputs. Furthermore, the material displayed high environmental durability,

preserving its photophysical integrity and wet-bonding capabilities under prolonged water immersion (21 days), thermal shock cycling, and accelerated UV weathering. Overall, this study presents a promising deconstruction–locking–activation conceptual framework that offers a practical methodology for engineering sustainable high-performance biomass functional materials.

Conflict of Interests

The authors declare no interest conflict.

Electronic Supplementary Information

Electronic supplementary information (ESI) is available free of charge in the online version of this article at <http://doi.org/10.1007/s10118-026-3832-1>.

Data Availability Statement

Data supporting the findings of this study are available from the corresponding author upon reasonable request.

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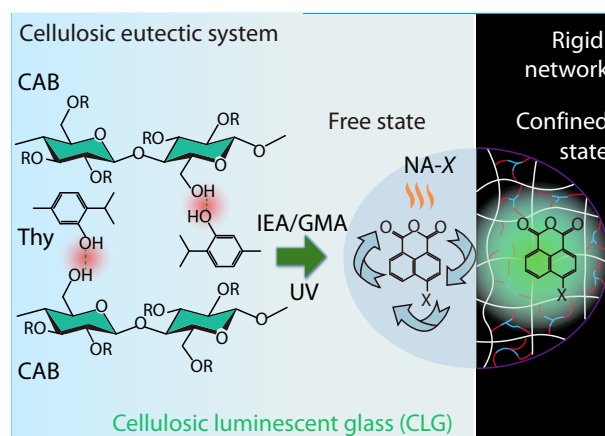
Graphical Abstract

Sequential Covalent Locking of Cellulosic Eutectic Systems Enables Transparent High-strength and Luminescent Engineering Glasses

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Sequential covalent locking transforms cellulosic eutectic melts into transparent, high-strength, luminescent engineering glasses. The dense topological network restricts molecular motion, activating efficient solid-state photoluminescence with high hydrolytic stability.



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