

## RESEARCH ARTICLE

# Advanced Paper-Based Three-Dimensional Electronics Fabricated via Freeform Direct Laser Writing

Shuhong Yang<sup>1</sup> | Liu Liu<sup>1</sup> | Bujingda Zheng<sup>2</sup> | Qiang Yan<sup>3</sup>  | Jian Lin<sup>2</sup>  | Caixia Wan<sup>1</sup> 
<sup>1</sup>Department of Chemical and Biomedical Engineering, University of Missouri, Columbia, Missouri, USA | <sup>2</sup>Department of Mechanical and Aerospace Engineering, University of Missouri, Columbia, Missouri, USA | <sup>3</sup>Forest Products Laboratory, USDA Forest Service, Madison, Wisconsin, USA

**Correspondence:** Caixia Wan ([wanca@missouri.edu](mailto:wanca@missouri.edu))

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## ABSTRACT

This study presents a borax–water-activated cellulose nanofiber (BWCNF) material that can be readily transformed into robust three-dimensional (3D) electronics through molding and freeform direct laser writing (FDLW). Compared with untreated cellulose nanofiber (CNF), BWCNF exhibits 135% of the tensile strength and 186% of the extensibility. Molded wavy structures also show excellent shape retention, with negligible force–strain hysteresis (<0.5%) over 1000 stretch–release cycles at 40% strain. Laser-induced graphene (LIG) can be patterned directly on these 3D BWCNF structures, yielding a sheet resistance as low as 54  $\Omega$   $\text{sq}^{-1}$  and a D-to-G Raman band intensity ratio of 1.5. The resulting devices include wave-structured piezoresistive strain sensors with gauge factors (GF) as high as 44.5 at 48% strain ( $R^2 > 0.97$ ) and low response hysteresis (6%–9%), capacitive touch sensors for directional control (approximately 300% relative capacitance change per  $0.8 \times 0.8$  cm electrode), and light-emitting diode (LED) arrays assembled on a 3D paper model. These results demonstrate that borax–water activation enables robust paper-based substrates for advanced 3D electronics.

## 1 | Introduction

Laser-induced graphene (LIG) can be readily synthesized on various carbon-rich precursors (e.g., polyimide, lignin, and cellulose) through direct laser writing (DLW) [1–5]. Owing to its high specific surface area, electrical conductivity, thermal stability, and chemical resistance, LIG has been widely used in advanced electronic devices, including sensors and supercapacitors [1, 6, 7]. Most LIG-based devices use two-dimensional (2D) structures, which offer excellent flexibility and wearability [8–10]. However, planar devices cannot readily provide the geometries needed for higher levels of component integration. In contrast, three-dimensional (3D) architectures use vertical space more efficiently, enabling greater system complexity and multifunctionality [11–13]. For example, tactile sensors designed with intricate 3D geometries can detect surface changes with

higher spatial resolution and accommodate larger mechanical deformations, improving performance in applications such as health monitoring and robotic control [14–17].

Cellulose nanofiber (CNF) from cellulosic biomass has emerged as a renewable and sustainable material [18–20]. Due to its exceptional mechanical strength, thermal stability, and optical properties, CNF has been widely used in applications spanning food safety, environmental monitoring, and healthcare [21–26]. To create 3D paper-based electronics, techniques such as origami and kirigami, which involve folding and cutting, are commonly used [27–29]. However, these approaches often result in discontinuous surfaces that hinder the formation of physically integrated LIG electrodes, limiting their use in piezoresistive sensing. Consequently, many origami- and kirigami-based paper devices rely on capacitive sensing and offer limited strain-sensing

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range and sensitivity [30, 31]. In contrast, 3D molding is a promising technique for constructing continuous 3D models with higher production efficiency, but it requires substrates that combine high formability with sufficient mechanical robustness to preserve the molded geometry [32]. This requirement is particularly critical for deformation-driven devices (e.g., sensing or energy harvesting), where recovery of the original geometry in each loading/unloading cycle underpins signal repeatability and reliability. CNF is well known for its high Young's modulus and can be shaped into 3D forms [33], yet molded CNF geometries show limited shape retention because the hydrogen-bonded fibril network gradually relaxes under sustained deformation. This limitation narrows its utility in deformation-dependent functions, highlighting the need for junction engineering that stabilizes the molded 3D geometry while preserving mechanical resilience under cyclic loading.

In this study, we introduce a borax–water-activated cellulose nanofiber paper (BWCNF) produced through sequential hot pressing and water activation. This treatment reorganizes the borax–CNF network to create a mechanically reinforced yet moldable substrate with high shape stability and resilience. Compared with untreated CNF, BWCNF shows a 135% increase in tensile strength and a 186% increase in extensibility, enabling robust molded 3D architectures under cyclic loading. BWCNF also supports in situ LIG formation and direct patterning of continuous conductive traces on nonplanar surfaces. Using focus-tracked freeform direct laser writing (FDLW), we fabricated representative 3D devices and evaluated their electrical performance, demonstrating a practical pathway toward paper-based 3D electronics for health and environmental monitoring, human–machine interfaces, and the Internet of Things (IoT).

## 2 | Materials and Methods

### 2.1 | Materials

CNF was purchased from the Process Development Center at the University of Maine. Custom 3D plastic molds were fabricated from polylactic acid filament (MatterHackers) by 3D printing, and commercial aluminum molds (Luxshiny) were used as received. All chemicals were purchased from Fisher Scientific (Hampton, New Hampshire, USA) and used as received.

### 2.2 | CNF Film Preparation and Characterization

Six sample types were prepared in this study (Table S1). The pure CNF film was prepared by vacuum filtering a 0.5 wt% nanocellulose suspension through Fisher Q5 filter paper (diameter, 5 or 12.5 cm) in a Büchner funnel, followed by air drying. The dried CNF film was immersed in a 5 wt% borax solution for 5 min, and then hot-pressed at 25°C or 100°C under 1 kN for 30 min. The resulting samples were labeled BCNF-X, where X denotes the hot-pressing temperature. BCNF-100 was then immersed in deionized water for 10 s and air-dried either on a flat surface or in molding tools under 1 kN for 3D shaping. The resulting water-activated material was labeled as BWCNF. Samples subjected to repeated hot pressing/water activation cycles are denoted as BCNF-100' and BWCNF', respectively. Tensile tests were conducted using

a TestResources 100 Series (TestResources Inc., Shakopee, MN, USA) at a stretching rate of 0.5 mm/min. Young's modulus was determined from the slope of the initial linear region of the stress–strain curve, and tensile strength was defined as the maximum stress prior to fracture. Stretching–release cycling tests were performed on a 3D BWCNF wavy structure (Figure S1) at 40% strain for 1000 cycles using an EasyMESUR F305 (Mark-10 Corporation, Copiague, NY, USA) at a stretching rate of 250 mm/min. The surface morphology of CNF films was investigated by scanning electron microscopy (SEM) (FEI Quanta 600 FEG, FEI Company, Hillsboro, Oregon, USA) equipped with a Bruker QUANTAX 200 energy-dispersive X-ray spectroscopy (EDS) system with an XFlash silicon drift detector (SDD) operated at 5 kV and 100 pA. Fourier transform infrared (FTIR) spectra were collected using a Nicolet 4700 FT-IR spectrometer (Thermo Fisher Scientific, Waltham, Massachusetts, USA) over 500–4000 cm<sup>−1</sup> at room temperature, with a resolution of 1 cm<sup>−1</sup> and 128 accumulated scans. Crystalline structure was analyzed using a Rigaku Ultima IV X-ray diffraction (XRD) system (Rigaku Americas Corporation, Woodlands, Texas, USA). The diffracted intensity of Cu K $\alpha$  radiation ( $\lambda = 0.1542$  nm; 40 kV and 44 mA) was measured over a  $2\theta$  range of 10°–50° with a step size of 0.02°. The crystallinity index (CrI) was calculated from the intensity of the (200) peak ( $I_{200}$ ,  $2\theta \approx 22.6^\circ$ ) and the intensity minimum between the peaks at (2 0 0) and (1 1 0) ( $I_{am}$ ,  $2\theta \approx 18.7^\circ$ ) according to Equation (1) [34].

$$CrI = \frac{I_{200} - I_{am}}{I_{200}} \times 100\% \quad (1)$$

Thermal stability was evaluated using an SDT-Q600 simultaneous thermogravimetric analysis/differential scanning calorimetry (TGA/DSC) analyzer (TA Instruments, New Castle, Delaware, USA) under nitrogen from 50°C to 800°C at a heating rate of 30°C min<sup>−1</sup>.

### 2.3 | LIG Synthesis and Characterization

Three different 3D molds and the resultant BWCNF structures, including their geometries and LIG electrode configurations, are shown in Figure S1. LIG was directly synthesized on BWCNF models via FDLW using a custom 3D freeform direct laser-writing platform (450 nm wavelength, maximum power of 5 W) that allows continuous and uniform LIG patterning on nonplanar and curved 3D surfaces (details in the Supporting Information) [35, 36]. Laser powers of 0.6–2.1 W were applied, while the defocus distance (2 mm) and scan rate (200 mm/min) were kept constant. Raman spectra of LIG were obtained on an inVia confocal Raman microscope (Renishaw, West Dundee, Illinois, USA). The morphology of LIG was examined using the same SEM system described above. For moisture-resistance testing, the LIG–BWCNF samples were dipped in a 4 wt% PVDF solution for 10 s, dried at room temperature for 24 h, and immersed in water for 48 h, followed by standard sample preparation for cross-sectional SEM imaging. High-resolution transmission electron microscopy (HRTEM) images were acquired on a Thermo Scientific Spectra 300 microscope operated at 300 kV. The LIG powder was scraped off from the surface of laser-scribed BWCNF films and sonicated in ethanol for 10 min before HRTEM imaging. A Pro-4 four-point

resistivity system (Lucas Labs, Gilroy, California, USA) was used to measure sheet resistance ( $R_s$ ).

## 2.4 | Fabrication and Evaluation of 3D Electronics

To develop strain sensors, copper wires were attached to the ends of the LIG electrodes using silver paste (Ted Pella Inc., Redding, California, USA). A Keithley 6514 electrometer (Keithley Instruments, Cleveland, Ohio, USA) was employed to continuously monitor the sensor's resistance changes, while an EasyMESUR F305 (Mark-10 Corporation, Copiague, New York, USA) was used to apply strain at different levels (i.e., 12%, 24%, 36%, and 48%), with a stretching rate of 20 mm/s. To develop the touch sensor, an additional layer of poly(vinylidene fluoride) (PVDF, 2.5 wt%) was applied to the surface of the LIG electrodes, serving as a dielectric layer. Finger-touch signals were transmitted to MATLAB through a XIAO ESP32S3 signal-processing module (Seed Studios Inc., Nanshan, Shenzhen, China). Light-emitting diodes (LEDs) were attached to the 3D conductive pathway with silver paste and operated at 10 V direct current (DC). The gauge factor (GF) was calculated as the ratio of the relative resistance change ( $\Delta R/R_0$ ) to the applied strain ( $\epsilon$ ), as expressed by Equation (2).

$$GF = \frac{\Delta R/R_0}{\epsilon} \quad (2)$$

Unless otherwise specified, all characterization and device performance measurements were conducted under ambient conditions at 20°C–25°C and 25%–40% relative humidity.

## 3 | Results and Discussion

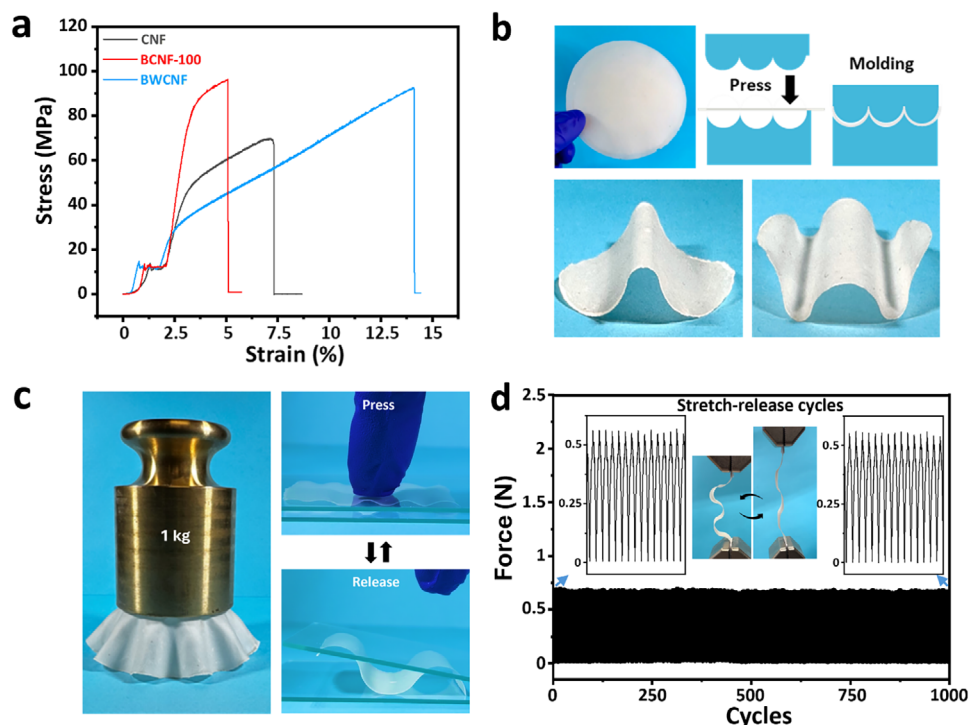
### 3.1 | BWCNF Properties and Moldability

Compared with CNF and BCNF-100, BWCNF exhibits a markedly smoother surface morphology (Figure S2) and significantly enhanced mechanical properties, with tensile strength and extensibility increased by 35% and 86%, respectively, relative to CNF (Figure 1a, Figure S3). Through molding, BWCNF can be readily shaped into diverse 3D structures with excellent shape retention and stability (Figure 1b,c, Video S1). In contrast, CNF structures showed poor shape retention, whereas BCNF-100 was too brittle for effective molding. However, once BCNF-100 undergoes a rewetting–redrying transition, it converts into BWCNF that can be effectively molded into 3D structures with high robustness and structural resilience. Load-bearing tests further confirmed the superior stability of BWCNF (Figure 1c). Despite a film thickness of only  $0.110 \pm 0.003$  mm (Table S2), the 3D BWCNF model withstood loads up to 1000 g without structural failure, in contrast to the collapsed CNF model (Video S2). This capability is attributed to balanced mechanical properties: a moderate Young's modulus ( $\sim 72$  MPa) provides sufficient structural rigidity for the hollow, arch-like geometry to resist collapse while maintaining flexibility. Moreover, under 1000 stretch–release cycles at 40% strain, the wavy BWCNF structures maintained a consistent force of approximately 0.7 N with negligible hysteresis (below 0.5%), confirming excellent fatigue resistance (Figure 1d). These results show that BWCNF can be molded into robust, durable, and flexible 3D structures for practical paper-based applications.

To understand BWCNF's superior mechanical performance, the interactions between borax and CNF under hot pressing and subsequent water activation were further investigated. FTIR spectra were analyzed for CNF, BCNF-25, and BCNF-100 (Figure 2a and Table S1). Pure CNF exhibited a distinct O–H stretching vibration at  $3343\text{ cm}^{-1}$ , along with characteristic cellulose peaks at  $1431$ ,  $1318$ ,  $1162$ ,  $1059$ , and  $898\text{ cm}^{-1}$  corresponding to symmetric  $\text{CH}_2$  bending,  $\text{CH}_2$  wagging, C–O–C stretching of  $\beta$ -glycosidic linkages, C–O stretching, and C–H deformation of  $\beta$ -glycosidic linkages, respectively. BCNF-25 showed multiple peaks in the region of  $3187$ – $3574\text{ cm}^{-1}$ , attributed to O–H stretching from different environments, including CNF, borax, CNF-borax, and bound water [37]. The appearance of doublet bands at  $1408$  and  $1353\text{ cm}^{-1}$  was assigned to the asymmetric stretching of trigonal  $\text{BO}_3$  units. A band centered at  $992\text{ cm}^{-1}$  was attributed to the overlapping asymmetric stretching of tetrahedral  $\text{BO}_4$  units and B–OH in-plane bending. Additionally, doublet peaks at  $832$  and  $808\text{ cm}^{-1}$  corresponded to B–O stretching associated with tetrahedral  $\text{B}(\text{OH})_4^-$  species. After hot pressing at  $100^\circ\text{C}$  for 30 min (BCNF-100), the multiple O–H stretching bands merged into a single broad peak at  $3347\text{ cm}^{-1}$ , indicating water loss, reduced hydrogen-bond heterogeneity, and structural reorganization. Notably, a new band appeared at  $1393\text{ cm}^{-1}$ , assigned to asymmetric stretching vibration of B–O–C, consistent with the formation of borate ester linkage between borate and CNF hydroxyl groups. Meanwhile, the original bands assigned to  $\text{BO}_3$ ,  $\text{BO}_4$ , and  $\text{B}(\text{OH})_4^-$  species disappeared, and new doublet bands at  $647$  and  $644\text{ cm}^{-1}$  assigned to bending vibration of B–O–B emerged.

Hot pressing–induced borate crosslinking including borate esterification (Figure 2b) led to distinct changes in the crystallinity index (CrI) and crystal structure of BCNF-100 compared with BCNF-25 (Figure 2c). The XRD patterns of all CNF-based films exhibited characteristic peaks at approximately  $14.9^\circ$ ,  $16.5^\circ$ ,  $22.6^\circ$ , and  $34.5^\circ$ , corresponding to  $(1\ -1\ 0)$ ,  $(1\ 1\ 0)$ ,  $(2\ 0\ 0)$ , and  $(0\ 0\ 4)$  lattice planes of cellulose I, respectively [38]. These results indicate that the CNF crystalline framework was preserved across BCNF-25, BCNF-100, and BWCNF. BCNF-25 showed a relatively high CrI of 82.80%, possibly reflecting the contributions from ordered borax hydrate domains. Upon hot pressing, however, the CrI of BCNF-100 dropped to 77.15%, a change attributable to dehydration and structural reorganization of borax domains accompanied by the formation of B–O–C and B–O–B crosslinks. Following water activation, BWCNF showed a further decrease in CrI to 72.73%, suggesting partial disruption of the borate–cellulose network. TGA analysis further confirmed the stabilizing role of borax crosslinking. Compared with pure CNF, BCNF-100 displayed a higher initial decomposition temperature and a substantially larger char yield at  $800^\circ\text{C}$  (10.76%) (Figure S4), indicating altered thermal degradation and enhanced carbonization. These effects are consistent with the formation of thermally stable B–O–C ester bonds and B–O–B bridges, which promote the development of a carbon-rich, crosslinked residue. Together, these results explain why BCNF-100 shows higher strength but less extensibility.

After water immersion and redrying (BWCNF), the characteristic peaks at  $647$  and  $644\text{ cm}^{-1}$  associated with B–O–B bending vibrations diminished (Figure 2d, Table S1), consistent with the lower CrI measured by XRD. More importantly, during



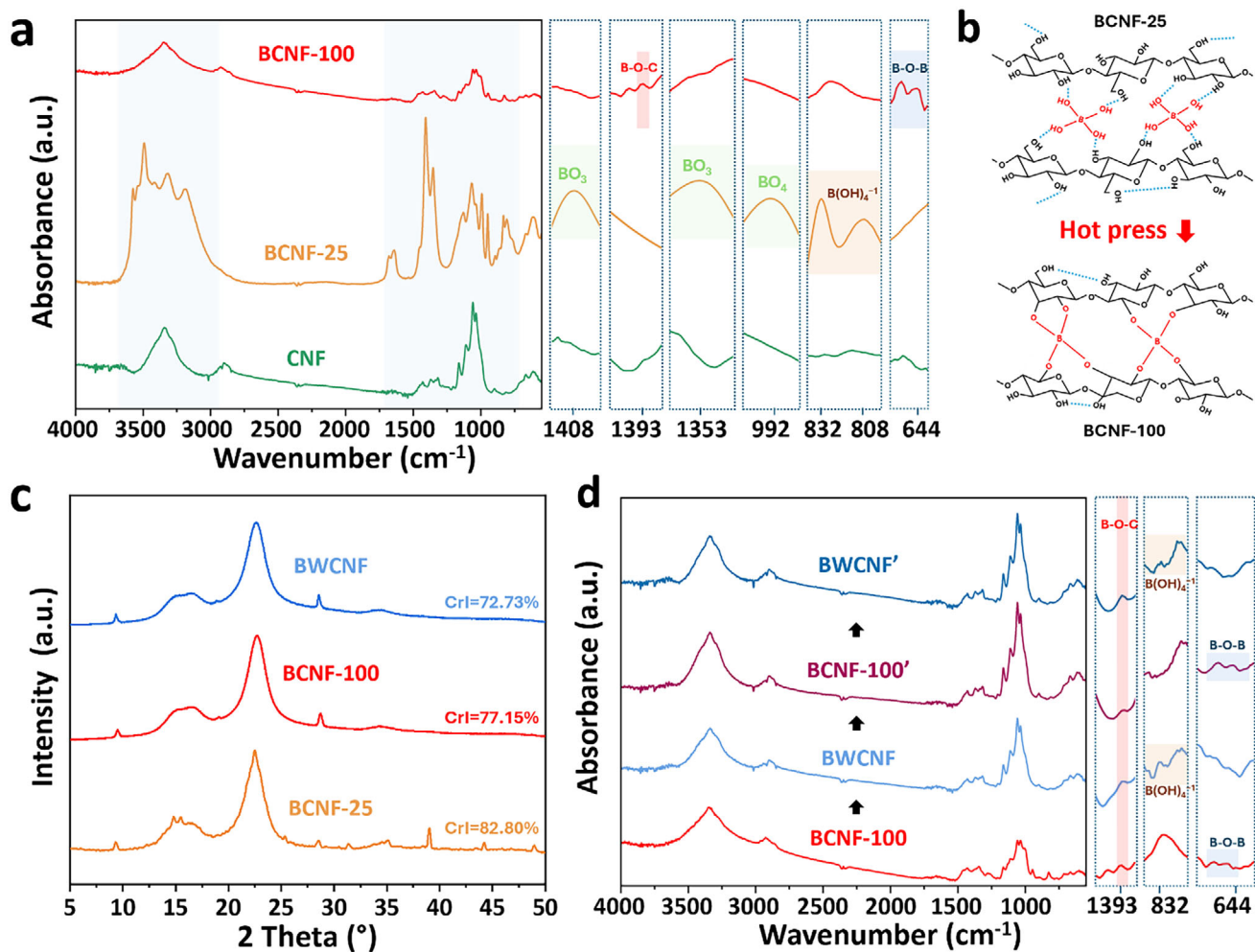
**FIGURE 1** | Mechanical properties of BWCNF. (a) Stress–strain curves of CNF, BCNF-100, and BWCNF. (b) Schematic of the molding process and representative 3D BWCNF structures. (c) Load-bearing and manual-compression demonstrations of 3D BWCNF structures. (d) Force response over 1000 stretch–release cycles at 40% strain; insets show the deformation states and representative cycles near the beginning and end of the test.

thermal–aqueous cycling (Figure 2d), the CNF/borax composite underwent reversible transformations between BCNF and BWCNF states (denoted as BCNF-100' and BWCNF', respectively, in the second cycle) through alternating water activation and hot pressing (Table S1). In the FTIR spectra (Figure 2d), the hot-pressed samples (BCNF-100 and BCNF-100') showed the disappearance of the doublet peaks at 832 and 808  $\text{cm}^{-1}$  corresponding to the symmetric B–O stretching of tetrahedral  $\text{B}(\text{OH})_4^-$  and the enhancement of the B–O–B bending doublet at approximately 647 and 644  $\text{cm}^{-1}$ , consistent with dehydration-induced borate condensation. Upon rehydration and redrying (BWCNF and BWCNF'), the  $\text{B}(\text{OH})_4^-$  peaks reappeared while the B–O–B peaks weakened, consistent with hydrolysis of condensed B–O–B bridges and regeneration of hydrated  $\text{B}(\text{OH})_4^-$  species. Meanwhile, the B–O–C asymmetric stretching peak at 1393  $\text{cm}^{-1}$  persisted throughout the entire cycle throughout the entire cycle, reflecting the high structural stability of the covalent B–O–C ester linkages. Collectively, these results show that BCNF-100 derives high strength from rigid B–O–B bridges and B–O–C linkages formed during hot pressing, but this locked network limits extensibility and moldability. Water activation reintroduces dynamic  $\text{B}(\text{OH})_4^-$  coordination into the covalently anchored framework, allowing these junctions to rearrange more readily under strain, enabling molded 3D structures to preserve strength and structural retention. This cooperative mechanism gives BWCNF a balance of robustness and adaptability for 3D electronics. The fibrous network provides structural integrity and load-bearing capacity, while the molded geometry accommodates deformation through structural reconfiguration. In contrast to conventional flexible substrates that are either intrinsically soft with limited structural support or relatively rigid with restricted deformability [39–41], BWCNF integrates these attributes into a

thin, moldable, and sustainable platform suitable for robust 3D electronics (Table S3).

### 3.2 | LIG Characterization

CNF could not support LIG formation because it burned under laser irradiation. In contrast, BWCNF exhibited flame-retardant behavior that suppressed combustion and stabilized the substrate, allowing LIG to form. The custom FDLW platform is shown in Figures 3a and S5. Laser powers of 0.6–2.1 W were applied to BWCNF. The optical and SEM images of the laser-treated samples are shown in Figure 3b. At 0.6 W, the energy was below the carbonization threshold and did not produce LIG. At 2.1 W, excessive energy caused thermal damage to the substrate. A usable power window of 0.9–1.8 W produced LIG without compromising BWCNF integrity. As the laser power increased from 0.9 to 1.8 W, the porosity of LIG gradually increased, with pore sizes expanding from approximately 5 to 50  $\mu\text{m}$  (Figure S6). This pore evolution arises from the photothermal and photochemical decomposition of lignocellulosic components: initial dehydration and decarboxylation, subsequent cleavage of C–O, C–H, and C–C bonds, and final rearrangement of  $\text{sp}^3$  into  $\text{sp}^2$  networks [42]. Higher laser energy promoted more extensive decomposition and gas evolution, producing larger pores in LIG [43]. The Raman spectra of all LIG samples (Figure S7) showed three characteristic bands: the D band ( $\sim 1360 \text{ cm}^{-1}$ ), associated with defects in  $\text{sp}^2$  carbon; the G band ( $\sim 1580 \text{ cm}^{-1}$ ), associated with in-plane  $\text{sp}^2$  carbon vibrations; and the 2D band ( $\sim 2670 \text{ cm}^{-1}$ ), arising from a second-order Raman process [44]. The lowest conductivity ( $R_s = 54 \Omega \text{ sq}^{-1}$ ) and lowest D-to-G band intensity ratio (1.5) were obtained at 1.8 W (Figure 3c,d). The 2D band was well fitted by



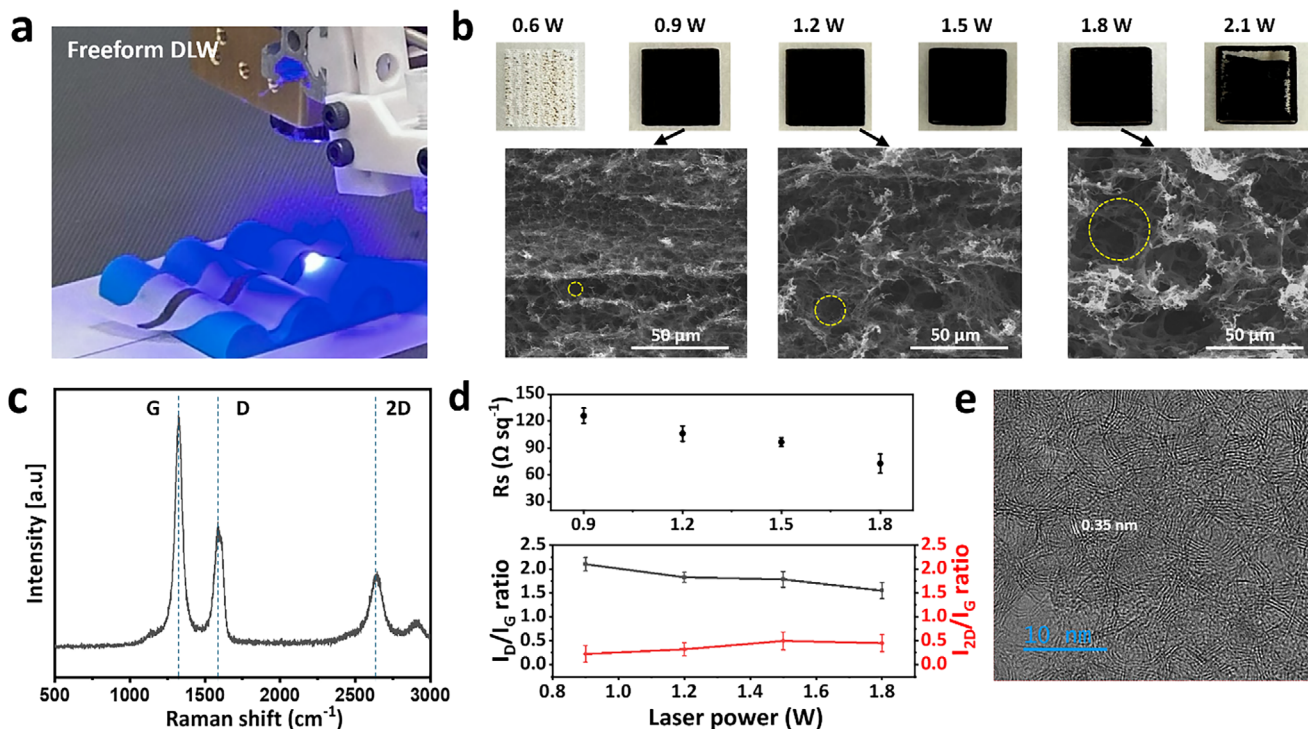
**FIGURE 2** | Characterization of CNF-based substrates. (a) FTIR spectra of CNF, BCNF-25, and BCNF-100. (b) Schematic of borate esterification induced by hot pressing. (c) XRD patterns of BCNF-25, BCNF-100, and BWCNF. (d) FTIR spectra showing reversible B—O—B/B(OH)<sub>4</sub><sup>-</sup> dynamics during hot-pressing/water-activation cycles while B—O—C linkages remain preserved.

a single symmetric peak ( $R^2 = 0.985$ ; Figure S9), consistent with graphene-like graphitic domains. HRTEM revealed lattice fringes with a spacing of approximately 0.35 nm (Figure 3e), further supporting the formation of graphitic domains. Energy-dispersive X-ray spectroscopy coupled with SEM (EDS-SEM) showed that the LIG was composed primarily of carbon, with smaller oxygen, boron, and sodium signals (Figure S9). The relatively uniform sodium distribution was consistent with dispersed borax-derived species in BWCNF (Figure S9) [45].

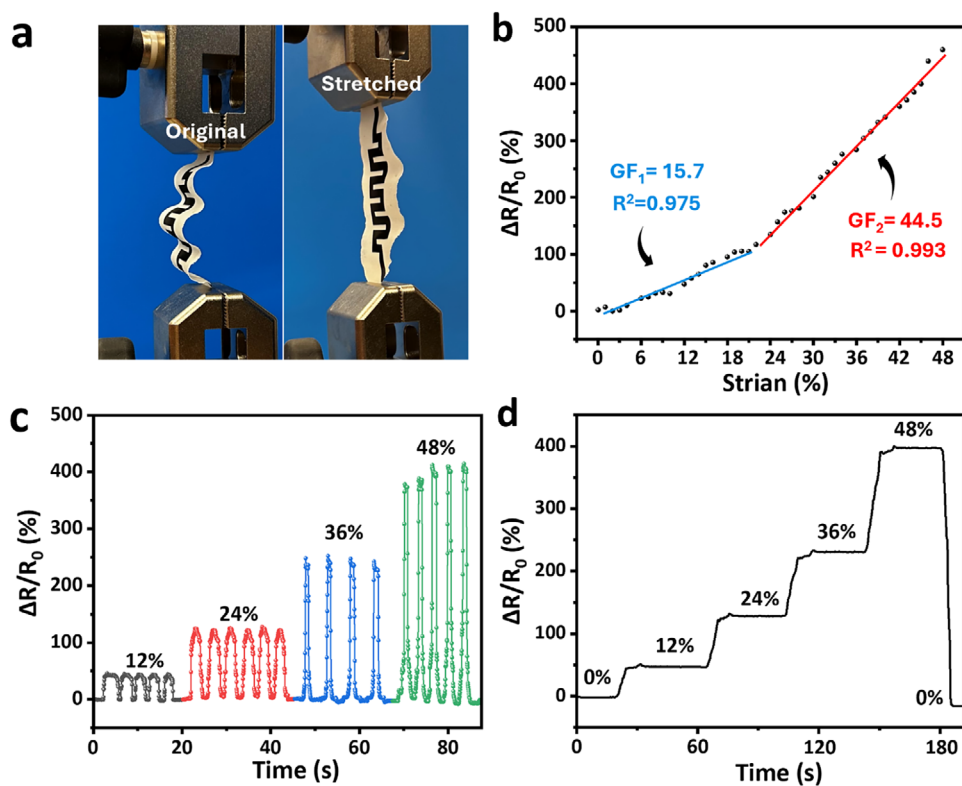
### 3.3 | 3D LIG Electronics

Conventional paper-based strain sensors are typically restricted to planar designs and are therefore limited to basic bending detection, particularly within low strain ranges [46–49]. In contrast, a 3D architecture accommodates strain through structural deformation and stress redistribution, while the accessible strain range can be tuned by geometry parameters such as curvature, amplitude, and structural height [13]. Here, we demonstrate a 3D wave-structured fully paper-based strain sensor capable of detecting strains up to 48%, significantly outperforming 2D paper-based sensors (Figure 4a and Table S4). The sensor exhibited two

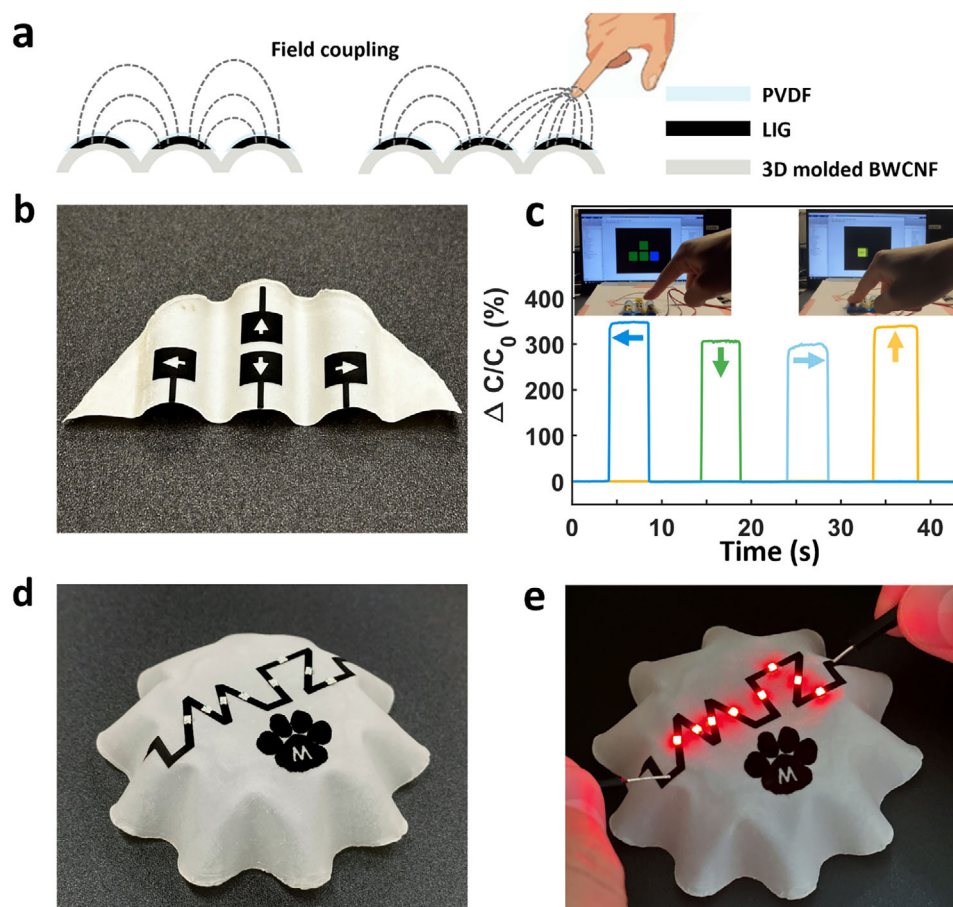
distinct operational regimes (Figure 4b). At strains below 20%, the sensor exhibited a gauge factor (GF) of 15.7, whereas in the higher strain range of 20%–48%, the GF increased to 44.5, indicating higher sensitivity. This transition is likely attributed to increased microcrack formation at higher strains, which amplifies resistance changes [50–52]. Both regions maintained strong linearity ( $R^2 > 0.97$ ), confirming reliable signal output. Dynamic performance was further validated through rapid cycling at multiple strain levels (12%, 24%, 36%, and 48%), where the sensor exhibited fast, highly repeatable responses with low hysteresis (6%–9%), confirming its reliable operation under dynamic deformation (Figures 4c and S10). In a stepwise hold test, the sensor signal remained stable at each strain level for approximately 30–50 s (Figure 4d). All strain-detection experiments were conducted on the same sensor, which consistently exhibited high sensitivity, durability, and repeatable performance under dynamic loading without requiring additional adhesion layers, underscoring the intrinsic robustness of the LIG-BWCNF integration. This behavior is consistent with previous biomass-derived LIG systems [53], in which strong LIG-substrate integration likely arises from amorphous carbon in the interfacial region (Figure S11a), facilitating mechanical integration with the fibrous network [51]. Collectively, these results underscore that BWCNF



**FIGURE 3** | Three-dimensional freeform direct laser-writing platform and LIG characterization. (a) Photograph of the custom platform. (b) Optical images of samples prepared at different laser powers and representative SEM images of LIG-0.9, LIG-1.2, and LIG-1.8. (c) Raman spectrum of LIG-1.8. (d) Sheet resistance and D-to-G and 2D-to-G Raman band intensity ratios for the LIG samples. Error bars represent the standard deviation from five independent measurements. (e) HRTEM image of LIG-1.8 showing a lattice spacing of approximately 0.35 nm.



**FIGURE 4** | Performance of the 3D BWCNF strain sensor. (a) Photographs of the original and stretched sensor states. (b) Relative resistance change as a function of strain, showing two linear regions with different gauge factors. (c) Relative resistance responses during repeated cycles at 12%, 24%, 36%, and 48% strain. (d) Stepwise relative resistance response, with each strain level maintained for approximately 30–50 s.



**FIGURE 5** | Three-dimensional BWCNF devices. (a) Finger–LIG electric-field coupling with PVDF as the dielectric layer. (b) Wavy touch sensor. (c) Relative capacitance responses of four channels; insets show directional control. (d,e) LIG circuit with attached LEDs before and during illumination.

provides a mechanically robust and shape-retaining paper platform where LIG is directly integrated without detachment even under dynamic use. Freeform laser writing enables efficient in situ LIG patterning on 3D BWCNF models, yielding high-quality, ready-to-use LIG-paper composites. To mitigate moisture-induced degradation in paper electronics, introducing a hydrophobic coating can effectively preserve the LIG–BWCNF integrity under wet conditions (Figure S11b). This integrated approach provides a practical route to reliable and scalable 3D paper-based electronics.

A tactile sensor was readily fabricated by coating a PVDF dielectric layer on top of the LIG electrodes patterned on the 3D BWCNF model. The sensor operates by detecting relative capacitance changes between a finger and the LIG electrodes (Figure 5a). Upon finger contact, the capacitance rapidly increased and produced an approximately 300% relative change for each  $0.8 \times 0.8$  cm electrode (Figure 5b,c), which was readily detected by the signal-processing module for transmission and control. The signal was sustained during continuous touch and vanished immediately upon release, confirming a fast, sensitive, and stable response. As a proof-of-concept, a paper-based touch sensor was integrated with MATLAB to achieve directional control of a virtual object: individual touches activated corresponding commands (Video S3), while continuous operation guided a virtual square through multiple directions and back to its origin (Video S4). Unlike planar touch sensors, this 3D configuration enables

contact detection on curved or shape-defined surfaces commonly found in wearable interfaces, soft robotics, and human–machine interaction systems, where conformal contact and geometry-dependent responses are often required. Although demonstrated here using simple finger touch, this result highlights the potential of geometry-enabled tactile sensing.

To demonstrate integration of the conductive LIG pathways with external electronic components, seven LEDs were attached to a conductive pattern spelling “MIZ” (Figure 5d,e). The LEDs illuminated under an applied voltage of 10 V, demonstrating electrical continuity across the patterned LIG pathway. This proof-of-concept further illustrates the potential of molded BWCNF structures for integrated paper-based electronics.

## 4 | Conclusion

Thermal treatment followed by borax–water activation transformed CNF into BWCNF with high mechanical strength, moldability, and structural resilience, establishing a robust 3D paper platform for LIG-based electronics. Strain sensing, tactile interfacing, and LED integration demonstrated the multifunctionality and dynamic reliability of LIG–BWCNF composites. In situ LIG formation on molded BWCNF also provided strong interfacial adhesion and stable conductivity, which are important for integrating electronic components into complex spatial

architectures. This work presents a potentially scalable and sustainable strategy for 3D paper-based electronics in flexible devices, human–machine interfaces, and IoT systems.

## Author Contributions

The manuscript was written through contributions from all authors. The authors have given approval to the final version of the manuscript.

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## Conflicts of Interest

The authors declare no conflicts of interest.

## Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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## Supporting Information

Additional supporting information can be found online in the Supporting Information section.

**Supporting File 1:** admt71269-sup-0001-SuppMat.pdf.

**Supporting File 2:** admt71269-sup-0002-S1.mp4.

**Supporting File 3:** admt71269-sup-0003-S2.mp4.

**Supporting File 4:** admt71269-sup-0004-S3.mp4.

**Supporting File 5:** admt71269-sup-0005-S4.mp4.