

Coordination polymers enhanced double-spiral zigzag-origami triboelectric nanogenerators for self-powered electrochemical bromination of alkylarenes

Congying Zhao, Ying-Ying Zhang , Linlin Cui, Yinghao Li, Dandan Wang, Lin Zhang, and Chao Huang 

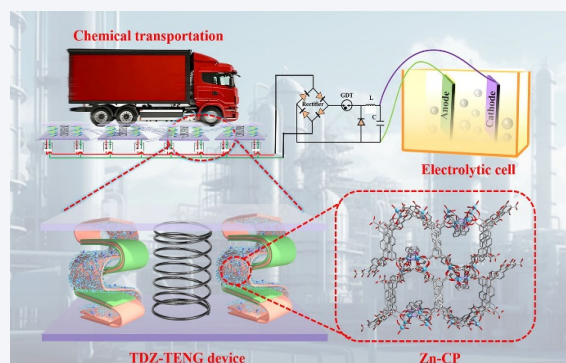
School of Materials Electronics and Energy Storage, and Center for Advanced Materials Research, Zhongyuan University of Technology, Zhengzhou 450007, China



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ABSTRACT: Electrochemical synthesis is a promising green strategy for organic synthesis, but its sustainability is compromised by its reliance on external power sources. Herein, we designed a double-spiral zigzag-origami triboelectric nanogenerator (DZ-TENG) based on zinc coordination polymer@cellulose acetate (Zn-CP@CA) composite films that can harvest mechanical energy from a vehicle-road pavement interface to drive an electrochemical system. A Zn-CP was synthesized and incorporated into CA at various mass ratios to fabricate a series of Zn-CP@CA composite films. Systematic characterization demonstrated that the 5% Zn-CP@CA composite film exhibited the highest triboelectric performance. Subsequently, a DZ-TENG utilizing the 5% Zn-CP@CA composite film was constructed and its output was optimized by adjusting the contact area and the number of folding layers. The resulting six-layer device DZ-6 delivered the optimal output under human mechanical energy. When mounted on a simulated vehicle setup, the TDZ-TENG device composed of two DZ-6 units harvested mechanical energy and converted it into electricity to drive an electrolytic cell for the site-selective bromination of alkylarenes, affording the corresponding benzyl bromide products in high yields and with excellent regioselectivity. This study presents a feasible method for designing vehicle-road pavement energy harvesters and expands the application of TENGs to sustainable and autonomous electrochemical synthesis.

KEYWORDS: coordination polymers, composite film, electrochemical synthesis, triboelectric nanogenerators, double-spiral zigzag-origami structure



1 Introduction

Driven by the global pursuit of carbon neutrality and the growing demand for sustainable manufacturing, electrochemical synthesis has emerged as a pivotal green technique for molecular activation, which is essential for organic synthesis, pharmaceutical development, and advanced materials fabrication [1–3]. Compared with conventional synthetic protocols, it employs electrons as the core reactive species instead of stoichiometric oxidants or reductants to drive chemical transformations, thus offering high

atom economy and low environmental risk, fully aligning with modern green chemistry principles [4–7]. Its mild reaction conditions not only accommodate sensitive substrates and eliminate side reactions and safety hazards associated with harsh conditions, but also significantly reduce energy consumption [8–12]. Precise control over redox processes can be readily realized by modulating the applied potential or current density, further expanding its applicability to sensitive substrates [13–17]. However, electrochemical synthesis is critically constrained by its inherent dependence on external power sources, which elevates the operational costs, electricity usage, and environmental burden, thereby creating a paradox with its green credentials [18, 19]. Therefore, it is an urgent need to develop self-sustained and eco-friendly energy supply systems for electrochemical reactions.

Triboelectric nanogenerators (TENGs) have garnered substantial attention as a promising energy harvesting technology, which can efficiently convert ubiquitous mechanical energy into electricity,

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✉ Address correspondence to Ying-Ying Zhang, 6477@zut.edu.cn; Chao Huang, huangchao@zut.edu.cn

featuring broad material versatility, low manufacturing costs, and high energy conversion efficiency [20–28]. Triboelectric materials have a certain impact on the output performance of TENGs. Commonly used materials include triboelectric dielectric layer materials (e.g., polytetrafluoroethylene (PTFE), polyvinylidene fluoride (PVDF), polydimethylsiloxane (PDMS), Kapton, and nylon) [29–34]. Although these materials have good triboelectric properties, their structures cannot be precisely controlled, which limits the improvement of the performance of TENGs [35–38]. Coordination polymers (CPs), possessing designable metal active sites, high porosity, and large specific surface areas, can markedly enhance charge-storage capacity by optimizing their metal-node types, organic-ligand structures, and coordination modes [39–44]. By strategically integrating CPs into polymeric matrices, it is possible to engineer composite materials with improved triboelectric polarities, higher charge densities, and tailored mechanical properties, thereby boosting the overall output and adaptability of TENGs [45–50]. Furthermore, the inherent flexibility of the composite materials in architectural design allows CP-based triboelectric layers to be configured into multilayer or stacked arrays, effectively breaking through the traditional bottlenecks of insufficient charge density and limited energy conversion efficiency [51–53]. Therefore, the development of CP-based TENGs with enhanced charge density and structural adaptability is essential to enable their practical deployment in self-powered electrochemical synthesis systems.

Herein, we report a double-spiral zigzag-origami triboelectric nanogenerator (DZ-TENG) based on a zinc CP incorporated into a cellulose acetate matrix (Zn-CP@CA) for harvesting vehicle-road pavement energy to drive electrochemical bromination of alkylarenes. Zn-CP was synthesized and CA was identified as the optimal candidate through a systematic screening of the polarity of various polymers. Zn-CP was then dispersed into CA at varying mass ratios to fabricate a series of Zn-CP@CA composite films, among which the 5% Zn-CP@CA composite exhibited the highest triboelectric performance. Leveraging this optimized material, the DZ-TENG was fabricated, and its output was further improved by optimizing the contact area and the number of folding layers. Two optimized six-layer DZ-6 devices were integrated to form a TDZ-TENG, which was then coupled with an electrochemical cell through a capacitor. The generated electricity harvested from simulated vehicle-road pavement energy successfully drove the regioselective bromination of alkyl aromatic compounds.

2 Experimental

2.1 Materials

4,4''-Carbonylbis((1,1'-biphenyl)-3,5-dicarboxylic acid) (H_4 CBBBD) was synthesized according to improved methods in Ref. [54]. The other reagents and solvents were commercially available without further purification.

2.2 Synthesis of $[Zn_2(CBBBD)(2,2'\text{-bpy})]\cdot H_2O$ (Zn-CP)

$Zn(NO_3)_2$ (0.015 g, 0.05 mmol), H_4 CBBBD (0.0255 g, 0.05 mmol), 2,2'-bipyridine (2,2'-bpy, 0.0078 g, 0.05 mmol), MeCN (5 mL), and H_2O (5 mL) were mixed in a 25 mL Teflon-lined stainless steel container for heating at 160 °C for 3 days, and colorless crystals of Zn-CP were obtained (yield, 68%, depended on Zn). Elemental analysis calcd (%) for $C_{39}H_{24}N_2O_{10}Zn_2$: C, 57.73; H, 2.98; N, 3.45; Found: C, 57.75; H, 2.97; N, 3.47. IR (KBr, cm^{-1}): 3430 (s), 1720

(vw), 1660 (vw), 1600(vw), 1570 (vw), 1450 (m), 1360 (vw), 1270 (m), 1020 (s), 933 (s), 861 (s), 764 (s), 717 (s).

2.3 Preparation of the PVDF frictional electrode

3.75 g of PVDF powders were dissolved in a mixture of 8.5 g of N,N'-dimethylformamide (DMF) and 12.75 g of acetone and stirred at 60 °C for 30 min. The PVDF solution was applied to the Kapton film by rotating it at 2000 rpm for 90 s. The copper tape was then glued to the back of the Kapton film and the copper wire was fixed to the copper foil with conductive silver epoxy resin.

2.4 Assembly of Zn-CP-TENG, 2,2'-bpy-TENG, and H_4 CBBBD-TENG

Firstly, powder samples of Zn-CP, 2,2'-bpy, and H_4 CBBBD were individually coated as positive friction layer onto the adhesive side of the Cu tape. Then, using traditional contact separation mode, the PVDF film (as negative friction layer) and the positive friction layer were assembled face to face to form Zn-CP-TENG, 2,2'-bpy-TENG, and H_4 CBBBD-TENG, respectively. The effective contact area between them was 5 cm × 5 cm.

2.5 Assemble of Zn-CP@CA composite films

The CA (0.4 g) powders were dissolved in DMF (6 mL) and magnetically stirred at 100 °C for 30 min to obtain a homogeneous EC solution. In a separate centrifuge tube, Zn-CP was dispersed in 6 mL DMF at mass fractions of 1%, 3%, 5%, 7%, and 9%, and the mixture was subjected to ultrasonication for 30 min. The two solutions were then mixed and continuously stirred for 4 h to obtain a blended solution. The solution was poured onto a clean glass plate and dried in an oven at 80 °C for 30 min to form a film. After peeling off the film, it was cut into 5 cm × 5 cm squares, and each square was adhered to the adhesive side of copper tape. Copper wires were then bonded to the backside of the copper tape using conductive silver epoxy to obtain Zn-CP@CA composite film electrodes containing different mass fractions of Zn-CP.

2.6 Assemble of Zn-CP@CA-TENGs

The PVDF films and Zn-CP@CA composite films with different ratios were used as negative and positive friction layer, respectively. They were assembled together to fabricate Zn-CP@CA-TENG devices operating in vertical contact-separation mode with an effective contact area of 5 cm × 5 cm.

2.7 Preparation of DZ-2 device with different contact areas

Two PET films were cut into rectangles of the same size. Two positive and two negative electrode plates were attached to the upper and lower sides of each PET rectangle, respectively. The two PET films were then placed perpendicular to each other, bonded together, folded alternately, and finally sealed to complete the DZ-2 device. By adjusting the contact areas, DZ-2 devices with different contact areas were obtained (1, 4, 9, 16, and 25 cm²).

2.8 Preparation of DZ-4, DZ-6, DZ-8, and DZ-10

With a fixed area of 25 cm², DZ-4, DZ-6, DZ-8, and DZ-10 devices with different numbers of folding layers were obtained by adjusting the lengths of the PET substrate and the positive/negative composite films.

3 Results and discussion

We aimed to design a DZ-TENG based on Zn-CP@CA composite films to harvest mechanical energy from the vehicle–road interface and convert it into electricity to drive an electrochemical synthesis reaction (Fig. 1(a)). To that end, Zn-CP was first synthesized by reacting $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ with the main ligand H_4CBBP and the auxiliary ligand 2,2'-bpy in a mixed solvent of H_2O and MeCN, yielding colorless and transparent crystals. Single-crystal X-ray diffraction (SCXRD) revealed that Zn-CP crystallizes in the monoclinic space group $P2_1/m$ and features a three-dimensional (3D) network. The asymmetric unit contains two crystallographically independent Zn^{II} ions, one fully deprotonated CBBP^{4−} ligand, one coordinated 2,2'-bpy molecule and one coordinated H_2O molecule (Fig. 1(b)). The two Zn^{II} centers exhibit distinct coordination environments: Zn1 is tetrahedrally coordinated by four carboxylate oxygen atoms from four separate CBBP^{4−} ligands, while Zn2 is in a square-pyramidal coordination geometry, binding with two oxygen atoms from two CBBP^{4−} ligands, one oxygen atom from one H_2O molecule, and two nitrogen atoms from one 2,2'-bpy molecule. Each carboxylate group in the CBBP^{4−} ligand bridges two adjacent Zn^{II} ions, and the 2,2'-bpy chelates to Zn2, forming a helical substructure (Fig. 1(c)). The helical substructures are further linked via the terminal oxygen atoms of the CBBP^{4−} ligands to Zn^{II} ions in neighboring units, generating a 3D framework (Fig. 1(d)).

The powder X-ray diffraction (PXRD) pattern of the as-synthesized Zn-CP closely matched with the simulated one generated from the single-crystal structure, indicating its single-phase purity and excellent crystallinity (Fig. S1 in the Electronic Supplementary Material (ESM)). Solvent stability tests of Zn-CP demonstrated that its crystalline framework remained intact after immersion in water, ethanol and acetonitrile for 1 and 3 days, respectively, as evidenced by the identical PXRD patterns of all treated samples relative to the as-synthesized samples. In the Fourier transform infrared (FT-IR) spectroscopy of Zn-CP, the absorption bands at 1600 and 1360 cm^{-1} were assigned to the stretching vibration of carboxylate groups (COO^-), and the peak at

1450 cm^{-1} corresponded to the C=N stretching vibration of coordinated 2,2'-bpy (Fig. S2 in the ESM) [55, 56]. X-ray photoelectron spectroscopy (XPS) was employed to determine the chemical state of Zn, in which the corresponding Zn 2p_{3/2} and Zn 2p_{1/2} peaks at 1022.37 and 1045.39 eV, respectively, confirmed the presence of Zn^{II} (Fig. S3 in the ESM). The molecular formula of Zn-CP was verified by SCXRD, together with thermogravimetric analysis (TGA, Fig. S4 in the ESM), elemental analysis (EA), and charge balance analysis.

Following the structural characterization, we evaluated the triboelectric performance of the as-synthesized Zn-CP and determined the contribution of its constituent moieties to the triboelectric response. Zn-CP, the H_4CBBP ligand, and 2,2'-bpy were individually ground into powders to serve as positive triboelectric layers and paired with PVDF as the negative counterpart to fabricate the classical vertical contact-separation-mode devices (Fig. S5 in the ESM), designated Zn-CP-TENG, 2,2'-bpy-TENG, and H_4CBBP -TENG, respectively. The short-circuit current (I_{sc}) and open-circuit voltage (V_{oc}) were acquired at a fixed frequency of 5 Hz using a linear motor. As shown in Fig. S6 in the ESM, the Zn-CP-TENG exhibited the prominent output among all these three devices. The superior output is attributed to the abundant Zn^{II} metal-active sites and the porous architecture of Zn-CP, which accelerate charge accumulation and facilitate electron transport, thereby boosting the output of TENG [57, 58].

To improve the flexibility, integrability, and output performance of CP-based TENGs, Zn-CP powders were dispersed into organic polymers by pouring method to form composite films (Fig. 2(a)). Five readily processable polymers, including CA, polyacrylonitrile (PAN), ethyl cellulose (EC), methyl cellulose (MC), and PVDF, were first selected to determine the polarity. Cross-sectional scanning electron microscopy (SEM) images revealed that all prepared samples possessed comparable thicknesses, indicating that the influence of thickness could be excluded in subsequent performance comparisons (Fig. S7 in the ESM). Each polymer film was used as a positive triboelectric layer with PVDF to assemble TENGs (denoted as CA-TENG, PAN-TENG, EC-TENG, MC-

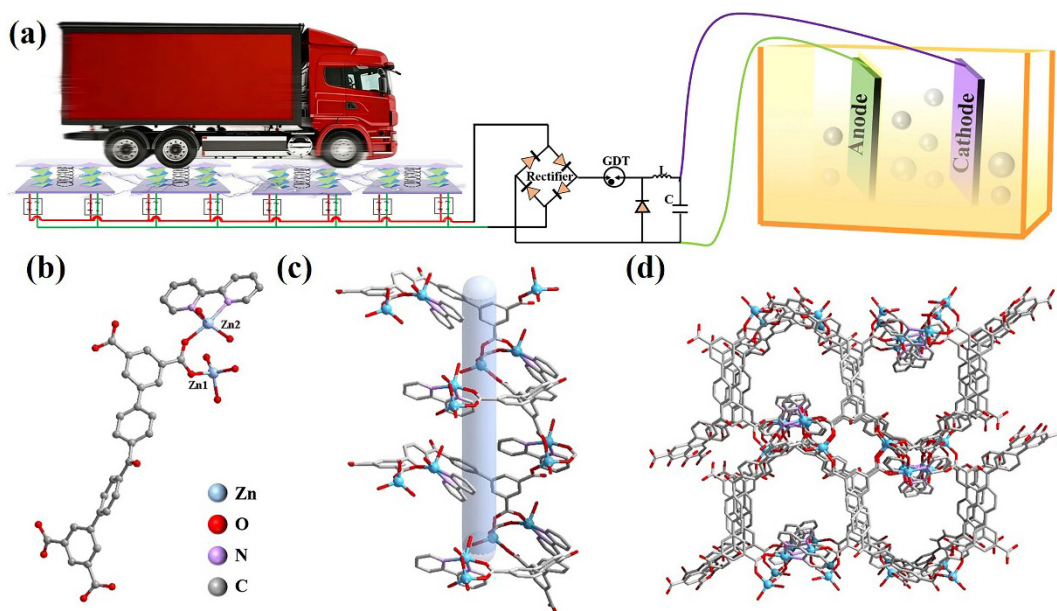


Figure 1 (a) Schematic illustration of the CP-based TENG for electrochemical synthesis. (b) The asymmetric unit of Zn-CP. (c) The helical substructure of Zn-CP. (d) The 3D framework of Zn-CP.

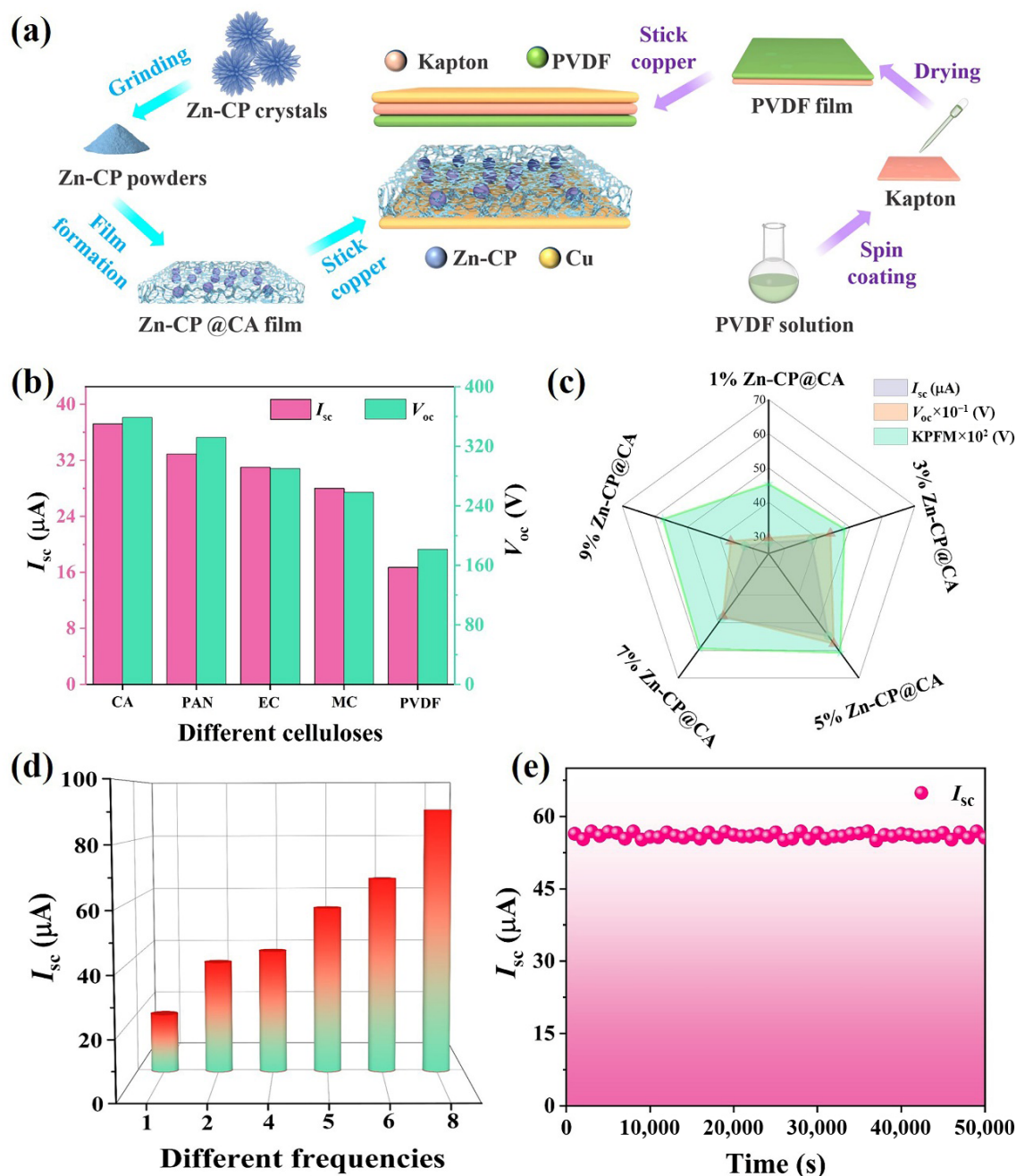


Figure 2 (a) Assembly process of Zn-CP@CA-TENG. (b) I_{sc} and V_{oc} of TENGs based on different organic polymers (CA, PAN, EC, MC, and PVDF). (c) I_{sc} , V_{oc} , and KPFM values for the different doping ratios of Zn-CP in Zn-CP@CA-TENG (1%, 3%, 5%, 7%, and 9%), respectively. (d) I_{sc} of 5% Zn-CP@CA-TENG under the frequencies from 1 to 8 Hz. (e) I_{sc} of 5% Zn-CP@CA-TENG after working 50,000 cycles.

TENG and PVDF-TENG), and the triboelectric outputs were measured under the identical conditions. The results revealed that the CA-TENG delivered the highest output, confirming its strongest positive triboelectric polarity and the optimal matrix material for fabricating the positive triboelectric layer of our TENG device (Fig. 2(b)). Subsequently, Zn-CP was incorporated into the CA polymer films, leveraging the synergistic effects of both Zn-CP and CA to modulate the overall performance. Composite films with different Zn-CP mass fractions (1 wt.%, 3 wt.%, 5 wt.%, 7 wt.%, and 9 wt.%) were synthesized. The SEM surface images clearly revealed the uniform distribution of Zn-CP within the Zn-CP@CA film matrix (Fig. S8 in the ESM), and the cross-sectional SEM images

revealed a similar thickness across all Zn-CP@CA composites (Fig. S9 in the ESM). In addition, the surface roughness (R_a) was found to be almost the same for all the composite films (Fig. S10 in the ESM). These observations indicated that the thickness and roughness of the Zn-CP@CA composite films exert negligible influence on the triboelectric output performance. The electrical outputs of the TENGs based on Zn-CP@CA with different doping ratios exhibited a distinct dependence on the Zn-CP loading. As shown in Fig. 2(c) and Fig. S11 in the ESM, both I_{sc} and V_{oc} of the Zn-CP@CA-TENGs rose significantly as the Zn-CP content increased from 1% to 5%, reaching 55 μA and 575 V, respectively. The corresponding transferred charge and charge density of 5% Zn-

CP@CA-TENG were 0.18 nC and 72.47 $\mu\text{C}\cdot\text{m}^{-2}$ (Fig. S12 in the ESM). This phenomenon was due to the enhanced positive triboelectric polarity of the composite film and the accompanying interfacial polarization induced by the intimate coupling between Zn-CP and CA. However, further increasing the Zn-CP content from 5 % to 9 % led to a decline in the device output performance, which destroyed the homogeneity of the composite film and thus diminished the triboelectric effect.

Kelvin probe force microscopy (KPFM) was employed to measure the surface potential of the Zn-CP@CA composite films. The surface potential values of the composite films with doping ratios of 1%, 3%, 5%, 7% and 9% were 0.453, 0.48, 0.608, 0.593, and 0.575 V, respectively, which were consistent with the electrical output trend of the Zn-CP@CA-TENG TENGs (Fig. S13 in the ESM). These results demonstrate that the 5% Zn-CP@CA composite film is the optimal positive triboelectric layer. The practical applicability and stability of the optimized 5% Zn-CP@CA-TENG were tested. As shown in Fig. 2(d) and Fig. S14 in the ESM, both I_{sc} and V_{oc} increased with the rising frequency (1–8 Hz) and reached their maximum values at 8 Hz. A durability test conducted at 5 Hz showed that I_{sc} and V_{oc} outputs remained virtually unchanged after 50,000 continuous working cycles, confirming their excellent stability (Fig. 2(e) and Fig. S15 in the ESM). Using the formula $P = FR$, the instantaneous power density was calculated and the 5% Zn-CP@CA-TENG achieved a maximum power density of 0.46 $\text{mW}\cdot\text{cm}^{-2}$ under an external load of 20 M Ω (Fig. S16 in the ESM), which was much higher than that of most cellulose-based or CP-based TENGs (Table S3 in the ESM). Furthermore, 5% Zn-CP was incorporated into other screened polymers (PAN, EC, MC, and PVDF), and the 5% Zn-CP@CA-TENG still exhibited the best output performance, further confirming the conclusions of the previous polymer triboelectric polarity screening (Fig. S17 in the ESM).

Based on the optimized 5% Zn-CP@CA composite film as the positive triboelectric layer and PVDF as the negative triboelectric layer, we designed a DZ-TENG with high elasticity and compactness using an alternating folding approach. The device significantly enlarged the contact area during each folding–unfolding cycle, thereby enabling efficient energy harvesting. As shown in Fig. 3(a), two polyethylene terephthalate (PET) strips were used as the substrate frameworks for the DZ-TENG: One strip was coated with Cu and 5% Zn-CP@CA on both sides, whereas the other was sequentially stuck with Cu, Kapton and PVDF on both surfaces. These two functional strips were joined together and folded alternately to construct the integrated DZ-TENG device. To further improve the integration and output performance of the DZ-TENG, the contact area and the number of folding layers were systematically optimized under hand-pressing. Taking the two folding layers device DZ-2 as the model, the output signals exhibited a continuous increasing trend with the enlargement of contact area ranging from 1 to 25 cm^2 , reaching their maximum values at a contact area of 25 cm^2 (Figs. 3(b) and 3(c) and Fig. S18 in the ESM). The phenomenon is attributed to the fact that a larger contact area allows more charges to participate in triboelectrification simultaneously, instantly raising the surface-charge density and thus the output performance. Subsequently, with the contact area fixed at 25 cm^2 , a series of DZ-TENGs with different numbers of folding layers (2, 4, 6, 8, and 10), named as DZ-2, DZ-4, DZ-6, DZ-8, and DZ-10, respectively, were assembled to investigate the effect of folding layers number on TENG output

performance (Fig. S19 in the ESM). Obviously, both I_{sc} and V_{oc} exhibited an initial increase followed by a decrease, with the maximum output performance (101 μA and 267.66 V) achieved at 6 folding layers. The output of DZ-6 was superior to that of most reported TENGs (Table S4 in the ESM). For the devices with 2 to 6 layers, the enlarged contact area dominated the improved electrical output (Figs. 3(d) and 3(e)). In contrast, when the layers exceed 6, excessive zigzag folding results in the increase of internal contact stress, which causes insufficient contact and thus decline the output [59].

On the basis of these findings, to explore the practical applications of the DZ-6 device for harvesting vehicle-road pavement energy, two DZ-6 units were assembled on opposite sides of a spring-supported base, noted as TDZ-TENG, and the wheel was attached to a linear reciprocator rolled over them to mimic vehicular motion, making the TDZ-TENG device to harvest energy through repeated contact and separation beneath the moving wheel (Fig. S20 in the ESM). The following tests and experiments were all conducted under these conditions. As shown in Figs. 4(a) and 4(b), the TDZ-TENG devices generated a stable output of 47 μA and 278.82 V at a fixed frequency of 2 Hz. To further assess its practical value, the charging capability of the TDZ-TENG device was examined and a self-powered system circuit consisting of a rectifier, a capacitor, and a load was constructed (Fig. 4(c)). Capacitors with different capacitances (33, 47, 100, 220, and 470 μF) were charged by TDZ-TENG device. The results showed that the charging voltage decreased as the capacitance increases, which was ascribed to the rise in the equivalent resistance of the circuit caused by the increase in capacitance, which further led to voltage loss during the charging process. The charging rate of the capacitor with the TDZ-TENG circuit was approximately eight times faster than that without the circuit (Fig. 4(d)). The TDZ-TENG device was connected to a 33 μF capacitor for energy storage, which could rapidly provide power to the thermohygrometer for environment monitoring (Fig. 4(e), Fig. S21 and Video S1 in the ESM). Moreover, the electrical energy generated by the TDZ-TENG device can also power light-emitting diodes (LEDs) and support the common operation of a calculator (Fig. 4(f), Fig. S21 and Videos S2 and S3 in the ESM). These results not only intuitively confirm the feasibility of the TDZ-TENG device for powering small electronic devices, but also laid a solid practical foundation for its targeted application in driving electrochemical systems.

It is known that alkyl aromatic compounds were essential chemical raw materials in organic synthesis and fine chemical manufacturing, and their production accompanied by frequent in-plant logistics transportation [60, 61]. By utilizing the TDZ-TENG device to harvest the kinetic energy generated from friction between transport vehicles and the ground and convert it into electrical power to drive product synthesis, synergistic integration of energy harvesting and green production would be realized with dual practical benefits. To evaluate the adaptability of the TDZ-TENG to real road situations, we investigated the output signals of TDZ-TENG under mechanical excitation with varying frequencies (Fig. S22 in the ESM). It was found that the electrical output of the device increased with the elevation of wheel moving frequency. Combined with its long-term operational stability validated by continuous movement at 2 Hz for over 3 h (Fig. S23 in the ESM), these results confirmed that the TDZ-TENG enabled robust and reliable vehicle-road energy harvesting under practical conditions.

Selective bromination of alkyl aromatic side chains is a key step

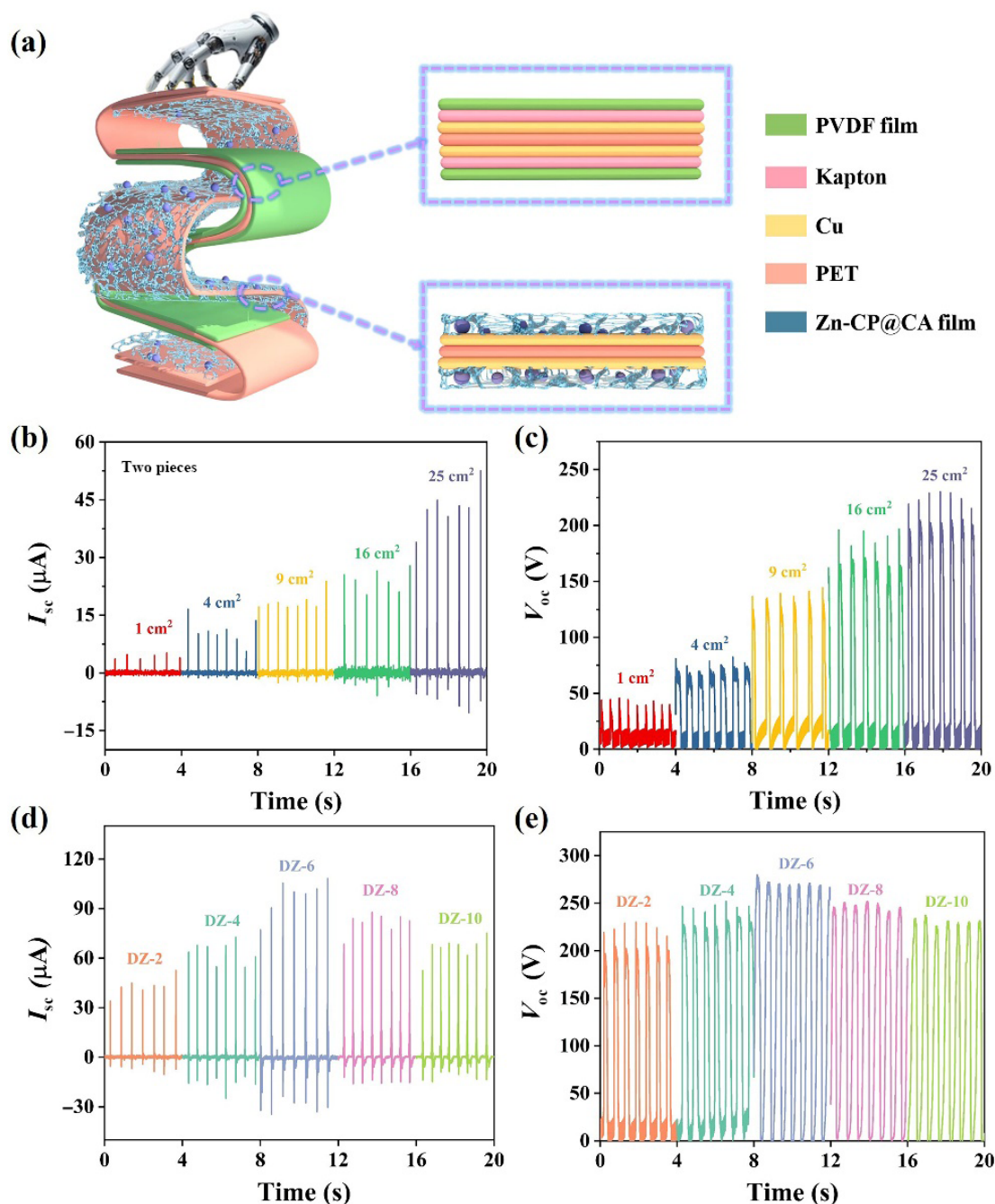


Figure 3 (a) Schematic diagram of DZ-6 device. (b) and (c) I_{sc} and V_{oc} of DZ-2 with the contact area of 1, 4, 9, 16, and 25 cm^2 , respectively. (d) and (e) I_{sc} and V_{oc} of DZ-2, DZ-4, DZ-6, DZ-8, and DZ-10 at a contact area of 25 cm^2 .

in the synthesis of benzyl bromide and subsequent preparation of pharmaceuticals and fine chemicals. Traditional methods predominantly rely on reagents such as Br_2 , NBS, or $CuBr_2$ along with radical initiators, which suffer from high toxicity, numerous byproducts, and poor regioselectivity [62, 63]. In contrast, electrochemistry offered high energy efficiency, environmental compatibility, and precise control over reactions under mild conditions, enabling sustainable chemical synthesis and energy conversion. Therefore, to avoid the need for an external power source, we integrated the TDZ-TENG with an electrochemical cell to construct a self-powered system for the site-selective bromination of alkylarenes. The TDZ-TENG device provided stable

direct current to the electrolytic cell through a capacitor, thereby converting the alkyl aromatic substrate into the corresponding benzyl bromide (Fig. 5(a)). A simple alkyl aromatic compound, toluene (2a), was selected as the model substrate to systematically optimize the reaction conditions for this self-powered bromination system (Table S5 in the ESM). Initially, the yield of the corresponding product 3a significantly increased with the elevation of NaBr concentration until saturation (Table S5 in the ESM, entries 1–5). After that, the yield decreased because of the extensive precipitation of NaBr crystals in the reaction system that influenced the homogeneity of the electrolysis environment (Table S5 in the ESM, entry 6). Notably, no bromination reaction occurred in the

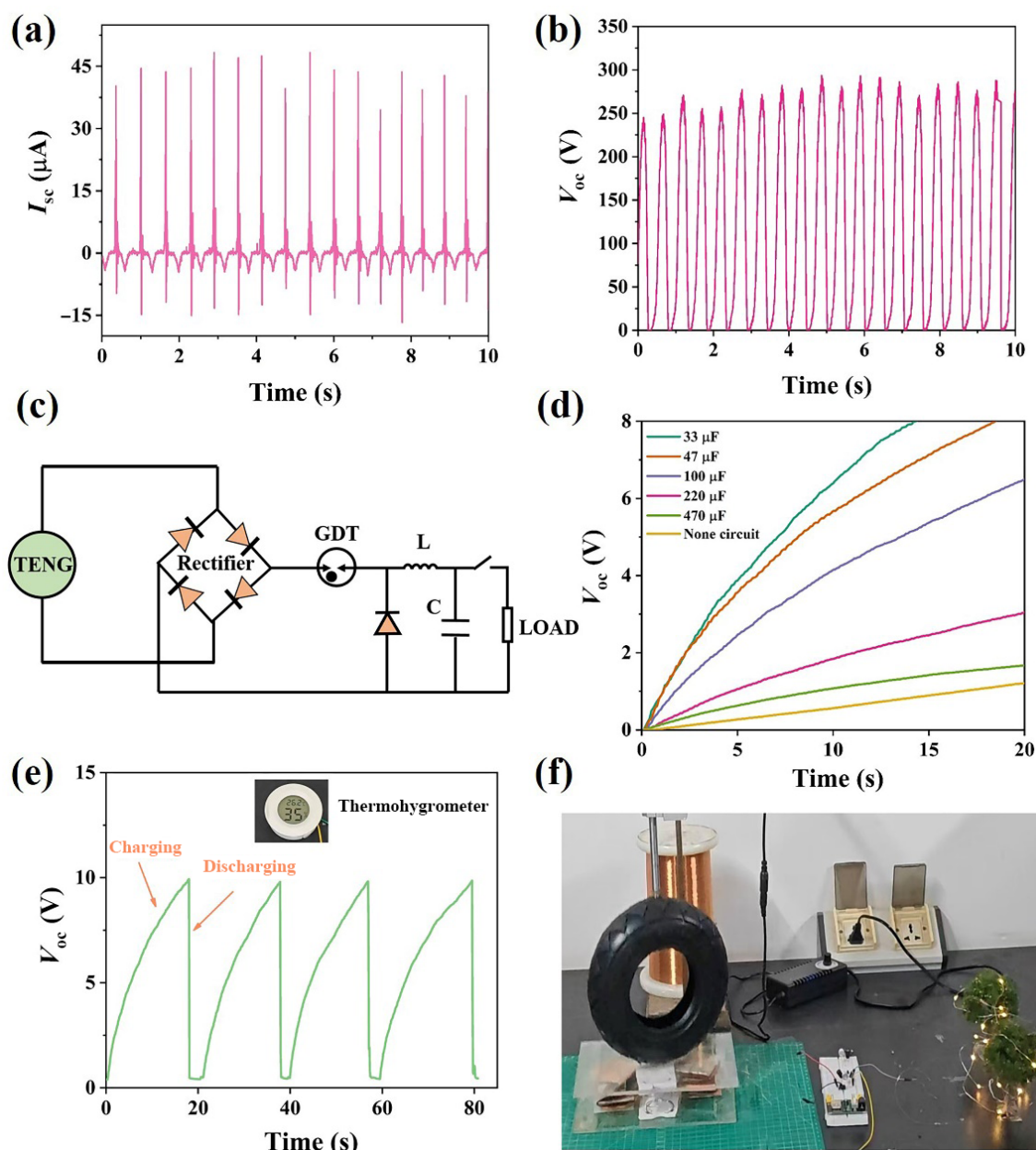


Figure 4 (a) and (b) I_{sc} and V_{oc} of TDZ-TENG device under the wheel moving. (c) Circuit diagram of the self-powered system. (d) Charging curves of capacitors with different capacities by TDZ-TENG device under the wheel moving. (e) The charging and discharging curve of the thermohygrometer driven by TDZ-TENG device under the wheel moving. (f) The digital photographs of LEDs powered by TDZ-TENG device under the wheel moving.

absence of electricity, verifying the inherent electrochemical nature of the reaction (Table S5 in the ESM, entry 7). Therefore, saturated solutions of NaBr, HBr, and CHCl_3 with Pt electrodes were employed as the standard reaction conditions to establish an electrochemical system powered by TDZ-TENG device for the selective bromination of alkyl aromatic compounds.

Upon the electron injection provided by the TDZ-TENG device, the reaction solution gradually turned from colorless to yellow, indicating the generation of brominating species. After powering the electrolytic cell by TDZ-TENG for a reaction time of approximately 4 h, complete conversion of substrate **2a** to product **3a** was observed by the process monitoring of ^1H NMR spectroscopy. As shown in Fig. 5(b), the peak of $\delta = 2.4$ ppm was assigned to the methyl protons ($-\text{CH}_3$), and the gradual attenuation

of this peak intensity with reaction time reflected the continuous consumption of the substrate **2a**. Meanwhile, a new singlet peak emerged at $\delta = 4.56$ ppm and its steady enhancement was attributed to the methylene protons ($-\text{CH}_2\text{Br}$) of the product **3a**.

To verify the generality of the method, a series of structurally diverse alkyl aromatic compounds were brominated under the optimized electrolysis conditions, and the results were displayed in Table 1. The self-powered two-phase electrolysis method afforded the corresponding α -brominated alkyl aromatic products with isolated yields of no less than 90%, along with excellent site-selectivity at the benzylic position. The reaction system exhibited sufficient tolerance toward various functional groups on the aromatic ring. Both electron-donating methoxy groups (**2b**) and electron-withdrawing halogen atoms (**2c**, **2d**, **2e** and **2f**) were well

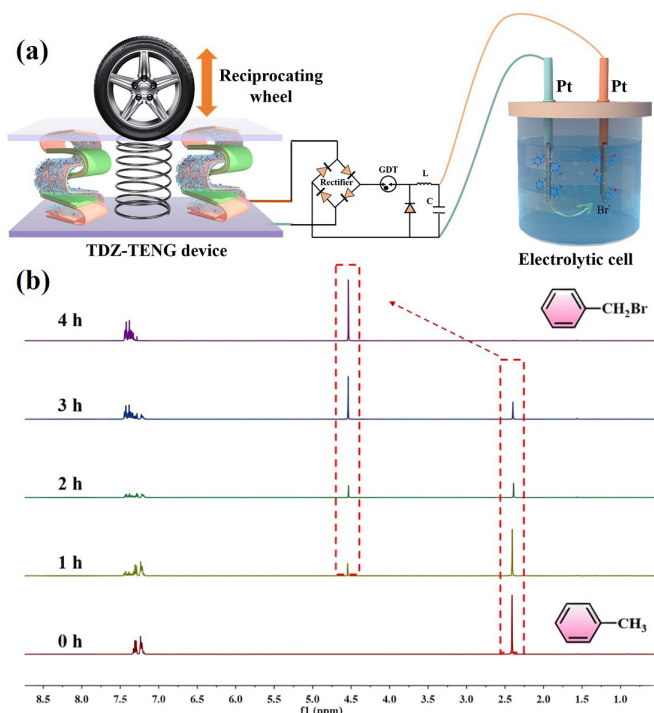


Figure 5 (a) The electrochemical synthesis system driven by TDZ-TENG device for the alkyl aromatic compounds. (b) ^1H NMR spectra of **2a** to **3a** driven by TDZ-TENG device for 0, 1, 2, 3, and 4 h, respectively.

tolerated, affording the desired products in excellent yields (Table 1, entries 2–4). Furthermore, different halogen substitution positions and numbers on the aromatic ring did not compromise the reaction efficiency (Table 1, entries 5 and 6).

Based on the above experimental results and the fundamental principles of electrochemical halogenation reactions, a hypothesis mechanism for the TDZ-TENG-driven bromination reaction was proposed (Scheme S1 in the ESM). Initially, bromide anions (Br^-) in the electrolytic cell undergo one-electron oxidation at the Pt anode to give bromine radicals ($\text{Br}^\cdot$), two of which rapidly dimerize to afford a Br_2 molecule. Subsequently, the electrochemically generated Br_2 reacts with H_2O to yield hypobromous acid (HOBr) and HBr . HOBr undergoes dehydration and condensation to form bromine monoxide (Br_2O). Homolytic cleavage of Br_2O produces the highly reactive $\cdot\text{OBr}$ radical that preferentially attacks a benzylic hydrogen from toluene, generating a benzyl radical. Finally, the benzyl radicals coupled with bromine radicals, generating the α -brominated product with high regioselectivity [64].

4 Conclusions

In summary, we successfully developed a self-powered electrochemical bromination system by integrating the TDZ-TENG into an electrolytic cell. A 3D porous Zn-CP with a well-defined crystal structure was synthesized, and five polymers with different triboelectric polarities were systematically screened to identify CA as the optimal matrix material for the positive triboelectric layer. By regulating the doping ratio of Zn-CP in the CA matrix, a series of Zn-CP@CA composite films were prepared and 5% Zn-CP@CA composite film was confirmed to exhibit the optimal triboelectric performance. Structural optimization of the DZ-TENG based on the superior 5% Zn-CP@CA composite films via regulating the contact area (25 cm^2) and the number of folding layers (6 layers)

Table 1 The self-powered electrochemical system of the site-selective bromination of alkylarenes^a

2a: $\text{R}_1=\text{R}_2=\text{H}$ 2d: $\text{R}_1=\text{H}, \text{R}_2=\text{Cl}$ The self-driven electrochemical system of the site-selective bromination of alkylarenes reaction 2b: $\text{R}_1=\text{H}, \text{R}_2=\text{OCH}_3$ 2e: $\text{R}_1=\text{Cl}, \text{R}_2=\text{H}$ 2c: $\text{R}_1=\text{H}, \text{R}_2=\text{Br}$ 2f: $\text{R}_1=\text{R}_2=\text{Cl}$		
Entry	Products (3a–3f)	Yield (%) ^b
1		98
2		98
3		97
4		95
5		93
6		90

^aReaction conditions: (+)Pt/Pt(−) cathode, undivided cell, anisole (**2a**, 1.3 mmol), saturated solution of NaBr (10 mL), HBr solution (0.5 mL), CHCl_3 (5 mL), $10\text{--}15^\circ\text{C}$, 4.0 h. ^bIsolated yield.

yielded the DZ-6 device that was assembled into TDZ-TENG, which could efficiently convert vehicle-road pavement mechanical energy into electrical energy. The TDZ-TENG device with an electrolytic cell enabled self-powered site-selective benzylic bromination of alkylarenes under mild conditions, affording the desired products in high yields ($\geq 90\%$) with excellent regioselectivity and good tolerance. This work not only provides a feasible strategy for the design of high-performance CP-based TENG devices and the efficient harvesting and utilization of vehicle-road pavement energy, but also further demonstrates the effectiveness of the TENG in driving electrochemical organic synthesis, breaking the reliance of electrochemical synthesis on external power sources.

Electronic Supplementary Material: Supplementary material (general methods, crystal data, and additional characterizations for Zn-CP and Zn-CP-TENG, etc.) is available in the online version of this article at <https://doi.org/10.26599/NR.2026.94909065>.

Data availability

All data needed to support the conclusions in the paper are

presented in the manuscript and 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

C. Y. Z.: Methodology, data curation, writing – original draft. Y.-Y. Z.: Conceptualization, formal analysis, supervision. L. L. C.: Software, visualization. Y. H. L.: Validation. D. D. W.: Project administration. L. Z.: Resources. C. H.: Supervision, funding acquisition, writing – review & editing. All the authors have approved the final manuscript.

Use of AI statement

None.

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