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Effects of graphene nanoplatelets and carbon nanotubes on the mechanical–electrochemical performance and hydration evolution mechanisms of solid-waste-based cementitious structural electrodes

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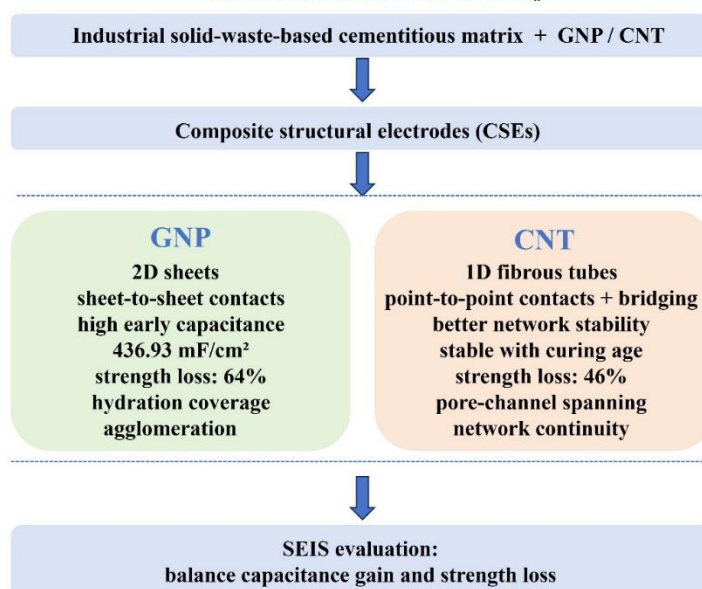
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Graphical abstract

《Effects of Graphene Nanoplatelets and Carbon Nanotubes on the Mechanical–Electrochemical Performance and Hydration Evolution Mechanisms of Solid-Waste-Based Cementitious Structural Electrodes》



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Abstract: Building-integrated distributed energy storage offers a promising route toward energy self-sufficiency in buildings. In this study, industrial solid-waste-based composite structural electrodes were fabricated by incorporating graphene nanoplatelets (GNP) or carbon nanotubes (CNT) into cementitious matrices, and their effects on mechanical performance, electrochemical behavior, and hydration-induced conductive-network evolution were systematically compared. Both carbon materials markedly enhanced energy-storage performance but reduced compressive strength. GNP increased the areal capacitance by up to 1040-fold to 436.93 mF/cm², exceeding the 499-fold enhancement achieved by CNT, but caused a greater strength loss (64% vs. 46%). The superior capacitance of GNP was associated with its two-dimensional lamellar morphology, which provides a large electrochemically accessible surface and facilitates sheet-based electron transport. However, continued hydration progressively covered GNP surfaces and promoted agglomeration/restacking, leading to capacitance attenuation. In contrast, the fibrous morphology of CNT helped maintain conductive-network continuity across pore channels and hydration products, resulting in better electrochemical stability with curing age. A Structure–Energy Integrated Score (SEIS) based on compressive strength, areal capacitance, and the Warburg coefficient further showed that dosage optimization should balance energy-storage gains against load-bearing losses. These findings provide guidance for designing solid-waste-based structural energy-storage electrodes.

Keywords: structural supercapacitor; industrial solid waste; conductive network; electrochemical performance; hydration evolution

1. Introduction

With the transition of the energy structure toward low-carbon and renewable systems, the limitations of centralized energy systems, such as high transmission losses and the fluctuating output of renewable energy, have become increasingly prominent. Building-integrated distributed energy storage has therefore emerged as an important pathway for achieving energy self-sufficiency and low-carbon operation in urban buildings ^[1,2]. Structural supercapacitors (SSCs) integrate energy-storage functionality into building materials, enabling structural components to simultaneously provide load-bearing capacity and electrical energy storage. Consequently, SSCs have attracted considerable attention in the field of green buildings ^[3].

Previous studies have shown that incorporating GNP, CNT, and other carbon-based conductive materials into cementitious/concrete-based energy-storage systems can endow cement-based SSCs with considerable energy-storage capacity ^[4,5], satisfactory mechanical strength, electrochemical stability ^[6], and safety ^[7]. The energy-storage mechanism of these SSCs is mainly governed by the principle of electric double-layer capacitance ^[8,9]. During charging, a layer of oppositely charged ions forms on the electrode surface, giving rise to an electric double-layer structure. During discharging,

the ions are released back into the electrolyte, the interfacial charge is neutralized, and the stored energy is delivered.

Existing studies in this field can be broadly classified into two categories. The first category employs graphene-based sheets as independent electrodes, while cementitious materials serve as separators or electrolyte carriers ^[10–12]. The second category incorporates carbon-based conductive materials, such as carbon black, activated carbon, CNT, graphene oxide (GO), or their hybrids, into cementitious matrices to form carbon–cement structural electrodes with both load-bearing and energy-storage capabilities ^[13,14]. In this type of system, the carbonaceous phase primarily contributes to electron transport and charge storage, while the inorganic cementitious matrix provides mechanical support, ionic transport pathways, and structural integration. For example, previous studies have proposed a functional division and synergistic enhancement between CNT and GO, in which CNT constructs a three-dimensional percolation network, whereas GO promotes the densification of C-S-H gels and the optimization of pore structures ^[15,16].

However, the differences in micro-morphology between GNP and CNT may lead to distinct effects on the mechanical and electrochemical performance of such systems. GNP is a two-dimensional sheet-like carbon nanomaterial with a high specific surface area and excellent electrical conductivity ^[17,18]. By contrast, CNT possesses a one-dimensional hollow fibrous structure, which is favorable for forming continuous conductive pathways ^[19] and enhancing the toughness and fracture resistance of composites ^[20]. Therefore, the contributions of these two carbon-based materials to the load-bearing capacity and energy-storage performance of structural electrodes are expected to differ. It is thus necessary to further clarify the role of carbon-based materials with different dimensional structures in cementitious systems.

In addition, inorganic cementitious materials are characterized by continuous hydration reactions, during which their microstructure, pore distribution, and hydration products evolve with curing age. For example, steel slag contains cementitious mineral phases such as dicalcium silicate (C_2S) and tricalcium silicate (C_3S), and its hydration process is comparable to that of Portland cement ^[21]. Ground granulated blast-furnace slag (GGBFS), as one of the commonly used alkali-activated materials (AAMs), exhibits satisfactory pozzolanic reactivity and can continuously generate hydration products in alkaline solutions, thereby contributing to high compressive strength ^[22]. These reactions are dynamic processes that drive the time-dependent evolution of cementitious systems at the microscopic scale (Ban et al., 2025; Fei et al., 2025). Such microstructural evolution may, to some extent, affect the stability of the conductive network. Therefore, after GNP or CNT is incorporated into a cementitious system, the resulting conductive network is not static, but is continuously influenced by the ongoing hydration process. Owing to the differences in dimensional structure, contact mode, and dispersion behavior between GNP and CNT, the hydration-age-dependent evolution of their electrochemical performance requires further clarification.

In this study, composite structural electrodes (CSEs) with both load-bearing capacity and energy-

storage functionality were developed by combining industrial solid wastes with carbon-based materials. The effects of GNP and CNT on the compressive strength and electrochemical performance of solid-waste-based cementitious binders were comparatively investigated. The differences in the spatial configuration of conductive networks were revealed, and the dynamic evolution mechanism of these networks during hydration was further elucidated, thereby clarifying their regulatory pathways for electrochemical performance. On this basis, a Structure–Energy Integrated Score (SEIS) evaluation framework was established. Through normalization and weighted modeling, compressive strength, areal capacitance, and the Warburg coefficient were comprehensively analyzed to compare and rank the performance of specimens prepared with different carbon-based materials and dosages.

2. Materials and methods

2.1. Materials

The ground granulated blast furnace slag (GGBFS) and phosphogypsum were sourced from Yichang City, Hubei Province, China, while the steel slag was obtained from Maanshan City, Anhui Province, China. Figure S1 presents the X-ray diffraction (XRD) patterns of GGBFS, steel slag, and phosphogypsum. The mineral composition of GGBFS consists primarily of amorphous silicate products, with no distinct peaks in the XRD spectrum except for minor quartz peaks. The steel slag comprises Ca_2SiO_4 and CaSiO_4 , along with some iron-containing compounds. The main component of phosphogypsum is $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$. Table S1 shows the elemental compositions of these three solid waste raw materials, where GGBFS is dominated by calcium (Ca), silicon (Si), and aluminum (Al); steel slag contains a high iron (Fe) content; and phosphogypsum is primarily composed of calcium (Ca) and sulfur (S). The elemental compositions were characterized using X-ray fluorescence (XRF) analysis.

The carbon-based conductive materials used in this study include GNP and carbon nanotubes (CNTs), both purchased from Changzhou Tanbo New Materials Technology Co., Ltd., China. These carbon-based materials were incorporated into the cementitious matrix in the form of dispersions with a mass fraction of 5 wt%. Figure S2 illustrates the scanning electron microscopy-energy dispersive X-ray spectroscopy (SEM-EDS) image (a) and elemental composition (b) for GNP. Figure S3 illustrates the scanning electron microscopy-energy dispersive X-ray spectroscopy (SEM-EDS) image (a) and elemental composition (b) for carbon nanotubes. In the SEM images, GNP exhibits a thin sheet-like or layered morphology with a sheet diameter of approximately 10 μm , while CNTs appear as elongated fibrous structures forming entangled networks. The EDS spectra of GNP and CNTs reveal prominent carbon (C) peaks at around 0.277 keV, accounting for 98.34% and 95.71% by mass, respectively, indicating that carbon is the primary constituent. Additionally, weak $\text{K}\alpha$ peaks for nitrogen

(N) at 0.392 keV, oxygen (O) at 0.525 keV, and sulfur (S) at 2.308 keV were detected, suggesting the presence of trace amounts of these elements. No significant signals from other elements were observed, confirming the high purity of the GNP and CNT samples.

Furthermore, the sodium sulfate reagent (analytical grade) used in this study was procured from Sinopharm Chemical Reagent Co., Ltd., China, serving as an alkali activator. The current collector employed was nickel foam produced by Tianjin Annohe New Energy Technology Co., Ltd., with a thickness of 1 mm, pore sizes ranging from 0.2-0.6 mm, and an areal density of 280-420 g/m², and the porosity is approximately 97.2%. All cementitious materials were prepared using deionized water.

2.2. Preparation of the solid waste composite structural electrodes

Composite structural electrodes (CSE) were prepared by directly adding the purchased carbon-based material dispersions (GNP and CNTs) into the solid waste powders, followed by the addition of a small amount of deionized water to facilitate processing. The mixture was then stirred in a mixer at a controlled speed of 800 rpm for 5 minutes. The dry mass ratios of carbon-based materials to solid waste raw materials were set at 1%, 3%, 5%, 7%, and 9%. The detailed mixing proportions are presented in Table S2. Specifically, OR denotes the reference specimens composed of pure solid waste cementitious materials without carbon-based additives. The G-series specimens refer to those incorporating GNP, while the CN-series specimens contain carbon nanotube (CNT).

After thorough mixing of the industrial solid waste with carbon-based materials, the resulting slurry was divided into two portions. The first portion was used for compressive strength testing, with specimen dimensions determined based on the studies by Yang et al. (2025) and Wang et al. (2025). The slurry was cast into cubic molds with dimensions of 20 mm × 20 mm × 20 mm to prepare cubic compressive strength specimens. The second portion was cast into rectangular molds with dimensions of 4 cm × 4 cm × 0.5 cm. While the slurry remained in a flowable state, a nickel foam sheet (3.5 cm × 5 cm) was inserted as a current collector to facilitate electrochemical testing.

Following casting, the specimens were cured in a constant-temperature curing box at 45°C for 48 hours before demolding. Fig. 1 illustrates the preparation process of solid waste-based composite electrodes and compressive strength specimen. Subsequently, the specimens were immersed in a 6 mol/L KOH solution for curing periods of 7, 14, and 28 days, after which compressive strength and electrochemical performance tests were conducted.

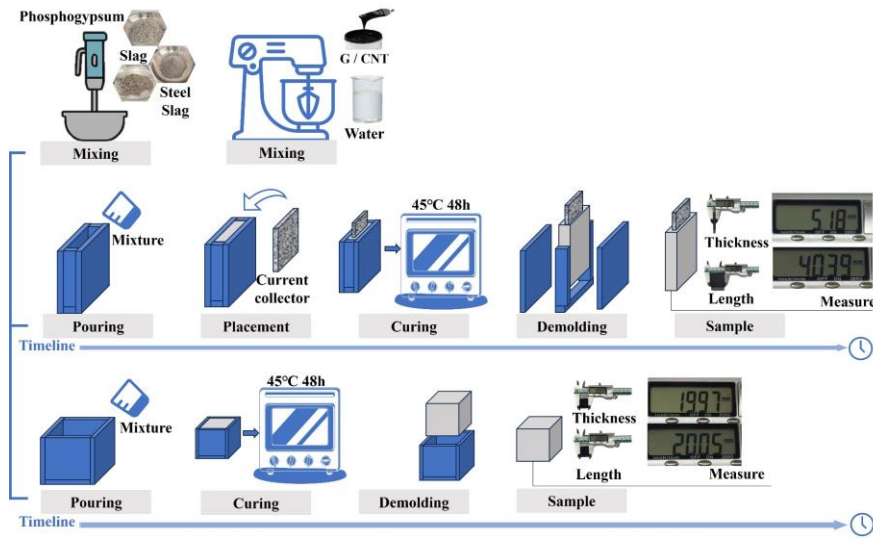


Fig. 1. Flowchart of solid waste-based electrode preparation process.

2.3. Test method

Compressive strength tests were conducted on cubic specimens using a YAW-2000D microcomputer-controlled electro-hydraulic servo pressure testing machine with a loading rate of 0.05 kN/s. Force-time curves were recorded during loading, and the peak force was taken as the ultimate load-bearing capacity of the specimen.

Electrochemical performance characterization was performed using a CS350M electrochemical workstation. Tests were conducted in a three-electrode system, with the composite structural electrode (CSE) as the working electrode, a platinum (Pt) electrode as the counter electrode, a mercury oxide (Hg/HgO) electrode as the reference electrode, and a 6 mol/L KOH solution as the electrolyte. Tests were conducted at 20°C with three replicates.

Cyclic voltammetry (CV) tests were carried out at scan rates ranging from 20 to 100 mV/s, with a voltage window of -0.8 V to -0.2 V. Galvanostatic charge-discharge (GCD) tests were performed at current densities ranging from 0.32 to 4 mA/cm². The areal capacitance was calculated from the discharge branch of the GCD curves according to Eq. (2):

$$C_{areal} = \frac{I\Delta t}{10 \times A\Delta V} \quad (1)$$

where C_{areal} is the areal capacitance (mF/cm²), I is the constant discharge current (A), Δt is the discharge time (s), ΔV is the effective discharge voltage window (V), and A is the exposed geometric surface area of the electrode in direct contact with the electrolyte (m²). The factor of 10 accounts for the unit conversion from F/m² to mF/cm².

Electrochemical impedance spectroscopy (EIS) tests were conducted over a frequency range of 10⁶ Hz to 10⁻² Hz to extract the Warburg coefficient (σ), which characterizes the ion diffusion resistance within the electrode material. The Warburg impedance analysis adopted a semi-infinite

diffusion model, assuming that the diffusion path in the vertical direction far exceeds the scale of the diffusing particles ^[27–29]. Under semi-infinite diffusion conditions, the Warburg impedance (Z_w) can be expressed as Equation (4) ^[30]. From Equation (5), the imaginary impedance (Z'') exhibits a linear relationship with the inverse square root of the angular frequency ($\omega^{-1/2}$). Linear fitting was performed to determine the Warburg coefficient, thereby characterizing the ion diffusion resistance in the electrode:

$$Z_w = \sigma \omega^{-\frac{1}{2}} - j \left(\sigma \omega^{-\frac{1}{2}} \right) \quad (2)$$

$$Z'' = -\frac{\sigma}{\sqrt{\omega}} \quad (3)$$

where σ is the Warburg coefficient, ω is the angular frequency, j is the imaginary unit, and Z_w is the Warburg impedance.

2.4. Characterizations

2.4.1. Transmission electron microscopy (TEM)

The morphology and local contact characteristics of GNP and CNT extracted from the hydrated composite specimens were examined using a JEM-2100 transmission electron microscope (JEOL Ltd., Japan) operated at an accelerating voltage of 200 kV. The cured composite specimens were ground into fine powder, dispersed in anhydrous ethanol, and ultrasonicated for 30 min. A drop of the resulting suspension was deposited onto a carbon-coated copper grid (200 mesh) and dried at room temperature. TEM images were acquired at different observation scales according to the microstructural features of interest, with scale bars ranging from 100 nm to 2 μ m.

2.4.2. Scanning electron microscopy (SEM)

The morphological features of hydration products and their spatial relationships with carbon-based materials were examined using a Sigma 360 field-emission scanning electron microscope (Carl Zeiss, Germany). Specimens cured for 7 and 28 days were fractured into pieces of approximately 2–5 mm, and hydration was terminated by immersion in anhydrous ethanol for 24 h. The samples were subsequently dried at 45 °C for 48 h, mounted on conductive carbon tape, and gold-coated. SEM observations were performed using an SE2 detector at an accelerating voltage of 3.0 kV and a working distance of approximately 8.2–8.4 mm. Images were acquired at magnifications of 30,000 $\times$ –50,000 $\times$.

2.4.3. Fourier transform infrared spectroscopy (FTIR)

The evolution of functional groups and hydration-related chemical bonds at different curing ages was characterized using a Nicolet 6700 Fourier transform infrared spectrometer (Thermo Fisher Scientific, USA). Hydration was terminated using anhydrous ethanol, followed by drying at 45 °C for 48 h. FTIR spectra were collected using the KBr pellet method over 400–4000 cm^{-1} , with a spectral resolution of 4 cm^{-1} . Background spectra were collected before each measurement.

2.4.4. Optical microscopy with camera probe

Surface macropores and crack-like defects were examined using the camera probe of an HC-F600 comprehensive crack tester (Beijing Haichuang Gaoke Technology Co., Ltd., China). The imaging system provides a resolution of 1280×720 pixels and a quantitative width measurement range of 0–10 mm with an accuracy of ± 0.01 mm. Surface images were acquired directly using the camera probe, and the widths of visible macropores and crack-like defects were determined using the built-in measurement function.

2.4.5. Mercury intrusion porosimetry (MIP)

The pore-size distribution was characterized using an AutoPore V 9600 mercury intrusion porosimeter (Micromeritics Instrument Corporation, USA). Specimens cured for 28 days were crushed into particles of approximately 1–5 mm, and hydration was terminated using anhydrous ethanol. The samples were subsequently vacuum-dried at 45°C for 48 h. Mercury intrusion measurements were performed over a pressure range of 0–60000 psia, corresponding to an accessible pore diameter range of approximately 3.5 nm–350 μm . Pore diameters were calculated according to the Washburn equation using a contact angle of 130° .

3. Results

3.1. Effect of GNP/CNT doping on compressive strength

Fig. 2 illustrates the 28-day compressive strength trends of GNP-doped and carbon nanotube (CNT)-doped specimens. Compressive strength values are presented as the mean of three specimens ($n = 3$). The results indicate that the baseline solid waste cementitious material, without carbon-based additives, achieved a 28-day compressive strength of 21.8 MPa. With increasing doping levels of GNP or CNTs, the compressive strength decreased to 7.8 MPa and 11.76 MPa, respectively, representing reductions of approximately 64.22% and 46.06% compared to the undoped reference specimen (OR).

Notably, at identical doping levels, the compressive strength of GNP-doped specimens (G-series) was lower than that of CNT-doped specimens (CN-series). For instance, at a 3% doping level, the G3 specimen exhibited a compressive strength of 12 MPa, while the CN3 specimen reached 13.36 MPa. This consistent strength disparity across all specimens suggests that GNP has a more pronounced negative impact on the mechanical properties of solid waste cementitious materials compared to CNTs.

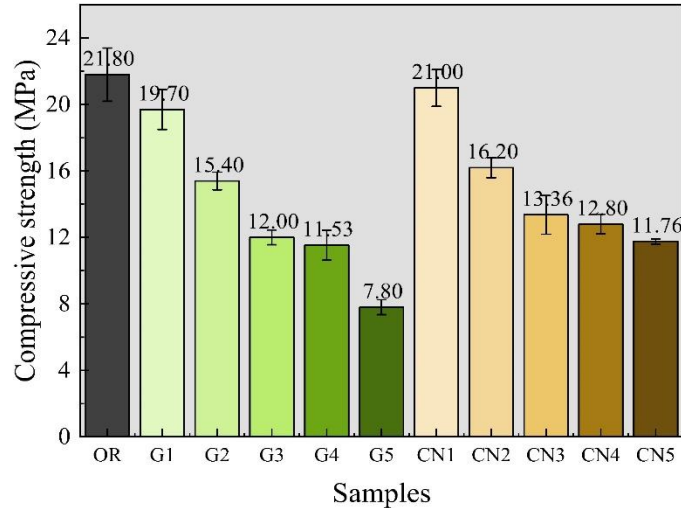


Fig. 2. Effect of GNP and carbon nanotube doping on the compressive strength of solid waste cementitious materials.

3.2. Effect of GNP/CNT doping on electrochemical performance

Fig. 3 displays the cyclic voltammetry (CV) curves of GNP-doped (a) and carbon nanotube (CNT)-doped specimens (b), respectively. The incorporation of GNP (G-series) or CNTs (CN-series) significantly increases the CV curve area and peak current. The baseline solid waste cementitious material exhibits poor conductivity and lacks effective charge storage pores, which is inherent to inorganic materials. The addition of carbon-based materials effectively enhances the electrode's conductivity and charge storage capacity. At equivalent doping levels, the CV curve area and peak current of GNP-doped specimens are higher than those of CNT-doped specimens, demonstrating that GNP provides a superior enhancement effect on the electrode's energy storage performance.

Fig. 3 present the CV curves of GNP-doped (c) and CNT-doped specimens (d) at varying scan rates, respectively. The CV curves exhibit excellent symmetry without redox peaks, indicating that the energy storage mechanism is primarily based on electric double-layer capacitance (EDLC) behavior, with good electrochemical reversibility and stability. The peak current increases progressively with higher scan rates. Specifically, at high scan rates, ions struggle to diffuse sufficiently to internal active sites within the electrode, resulting in charge accumulation primarily on the electrode surface; during voltage reversal, surface charges are released rapidly, leading to a stronger current response. This is reflected in the relationship between peak current and scan rate, as given by Equation (6):

$$I = C \cdot v \quad (6)$$

where I is the current (mA), C is the capacitance (F), and v is the scan rate (mV/s).

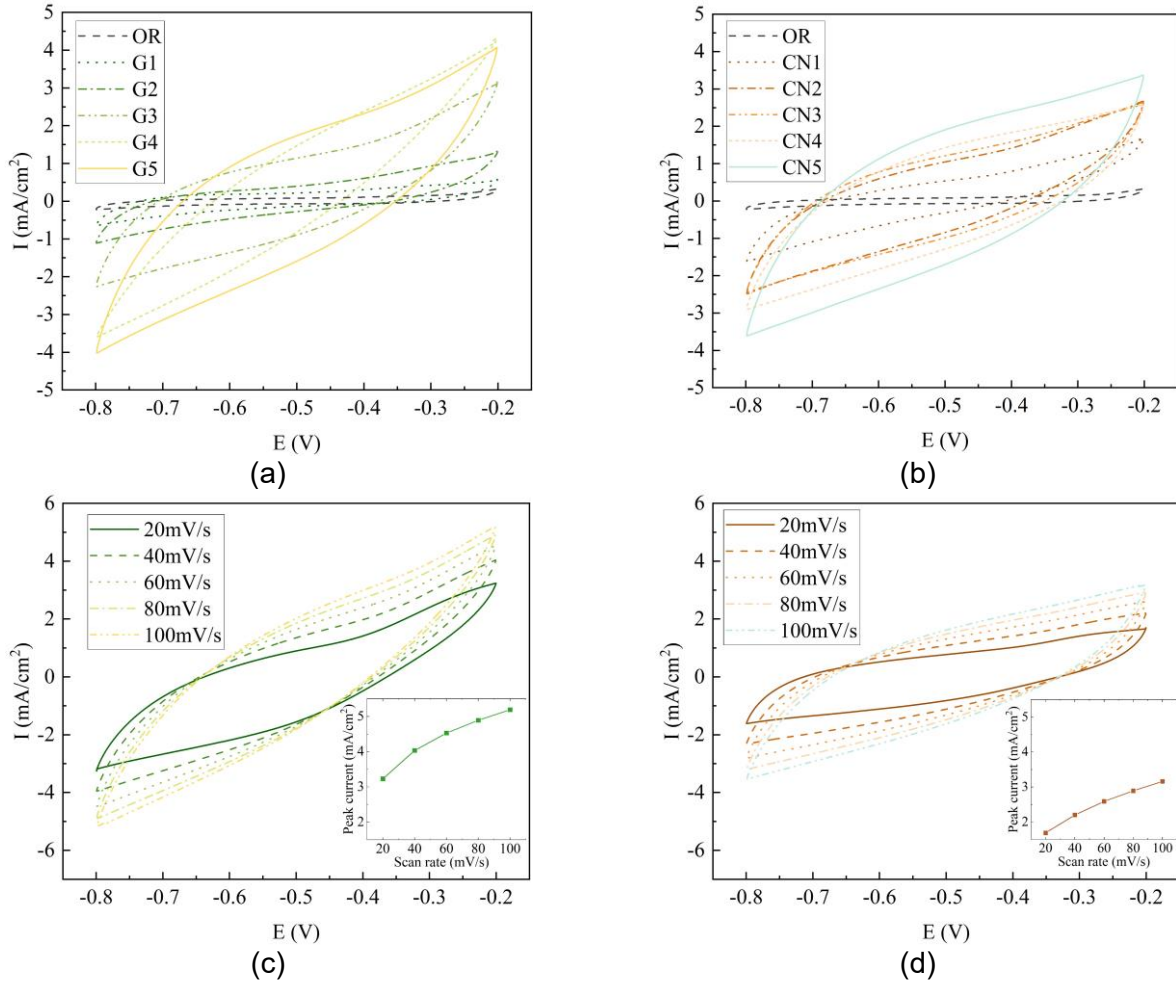


Fig. 3. Cyclic voltammetry (CV) curves of solid waste-based electrodes: (a) GNP electrodes with varying doping levels, (b) CNT electrodes with varying doping levels, (c) GNP electrode (G5) at different scan rates, and (d) CNT electrode (CN5) at different scan rates.

Fig. 4 presents the galvanostatic charge-discharge (GCD) curves of GNP-doped (a) and carbon nanotube (CNT)-doped specimens (b), respectively, along with the corresponding areal capacitance statistics in the insets. The charge-discharge duration of the specimens increases with higher GNP doping levels. As the GNP doping level rises from 0 % to 9 %, the areal capacitance of the solid waste-based electrode increases from 0.42 mF/cm² to 436.93 mF/cm² (at a current density of 0.32 mA/cm²), an increase of 1040 times. Notably, a significant rise occurs when the doping level changes from 5% to 7%, with the areal capacitance increasing from 50.9 mF/cm² to 335.09 mF/cm², representing an enhancement of 558.33%. For specimens doped with CNTs, the areal capacitance reaches a maximum value of 209.58 mF/cm² at a 5% doping level. Compared to GNP, this increase is only 499 times. Beyond 5%, the areal capacitance decreases, indicating a negative effect on capacitance at higher CNT doping levels.

In comparison, at a 9% doping level, the GNP-doped specimens achieve a percolation threshold in the conductive network, demonstrating superior enhancement in capacitance compared to CNT-

doped specimens. For instance, the G5 specimen exhibits an aerial capacitance of 436.93 mF/cm², while the CN5 specimen shows 108.89 mF/cm².

Fig. 4 display the GCD curves of GNP-doped (c) and CNT-doped specimens (d) at varying current densities, respectively, with the corresponding areal capacitance statistics in the insets. The charge-discharge duration decreases with increasing current density. At a low current density of 0.5 mA/cm², the charge-discharge process is slower, allowing sufficient time for charge storage and release within the solid waste electrode, resulting in areal capacitances of 332.08 mF/cm² for GNP-doped specimens and 82.39 mF/cm² for CNT-doped specimens. As the current density increases to 4 mA/cm², the areal capacitance decreases to 24 mF/cm² and 7.1 mF/cm², respectively.

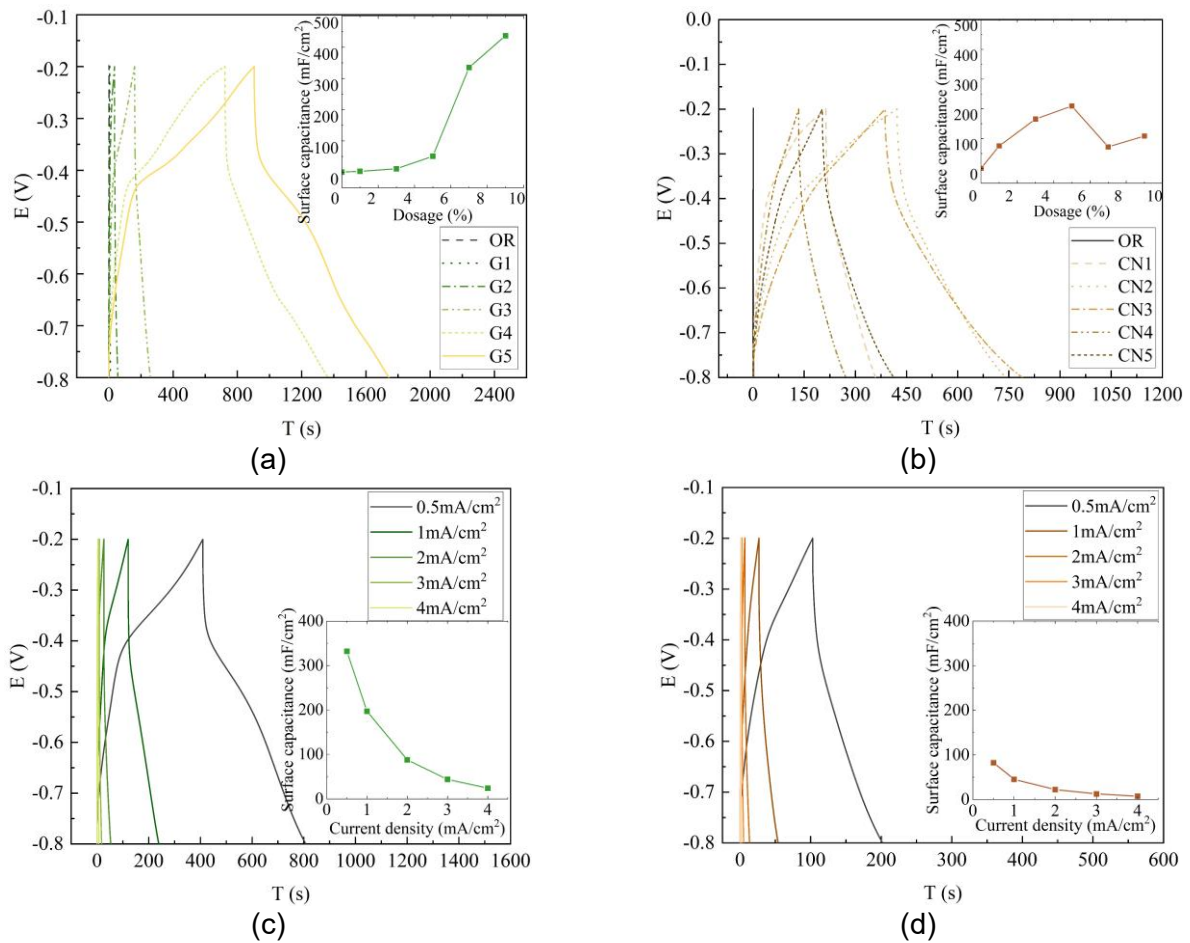


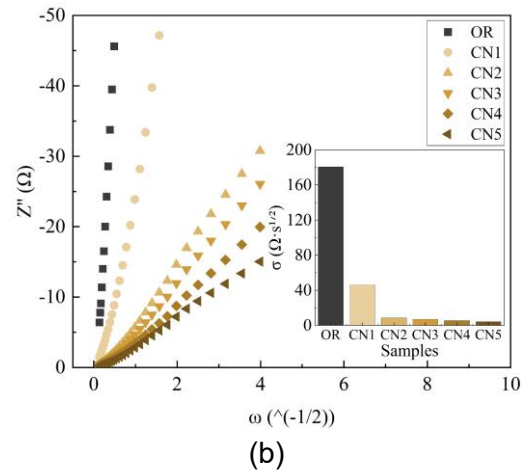
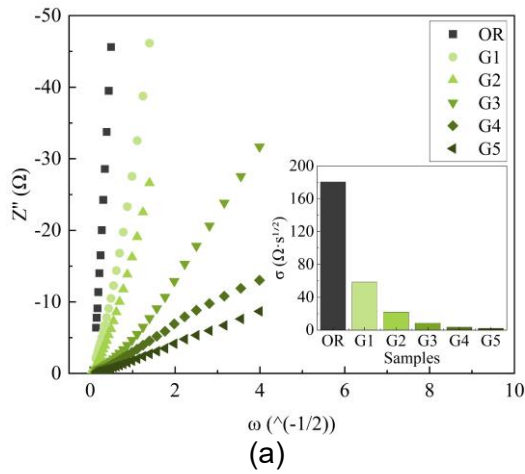
Fig. 4. Galvanostatic charge–discharge (GCD) curves of solid waste-based electrodes: (a) GNP electrodes with varying doping levels, (b) CNT electrodes with varying doping levels, (c) GNP electrode (G5) at different current densities, and (d) CNT electrode (CN5) at different current densities.

Fig. 5 present the Z'' vs $\omega^{-1/2}$ curves for GNP-doped (a) and carbon-nanotube-doped specimens (b), respectively, with the corresponding Warburg coefficients (σ) summarized in the insets. The Warburg coefficient reflects the diffusion resistance of electrolyte ions within the structural electrode [30,31], where a lower value indicates reduced transport resistance during ion migration. The results

show that the reference solid waste cementitious specimen (OR) exhibits a high σ value of $180.74 \Omega \cdot s^{1/2}$, suggesting significant hindrance to ion transport due to its internal structure. As the doping levels of GNP and CNTs increase from 0% to 9%, the σ values decrease to $2.27 \Omega \cdot s^{1/2}$ and $4.07 \Omega \cdot s^{1/2}$, respectively, representing reductions of 67.63% and 74.47%. This demonstrates that the incorporation of carbon-based materials improves the ion migration environment within the electrode, thereby lowering the transport resistance.

Fig. 5 display the Nyquist plots for GNP-doped (c) and CNT-doped specimens (d), respectively, with the solution resistance (R_s) values summarized in the insets. The intercept of the Nyquist curve with the real axis in the high-frequency region represents the ohmic component of EIS, or solution resistance. R_s varies due to factors such as the distance between the reference and working electrodes, the geometry and surface area of the working electrode [32]. The ohmic component provides information about electrolyte resistance, and its variations do not directly reflect the intrinsic electrochemical performance of the electrode material [33].

In summary, the addition of carbon-based materials provides porous channels for electrolyte ion diffusion within the specimens, enhancing the penetration depth of ions into the electrode interior. This allows the formation of electric double-layers not only on the shallow surface but also deeper within the structure, thereby increasing the effective energy storage interface. In contrast, the pure solid waste cementitious specimen (OR) has a denser structure, where most electric double-layer capacitance effects are confined to the shallow surface, resulting in low utilization of active sites. This will be further analyzed in the subsequent sections.



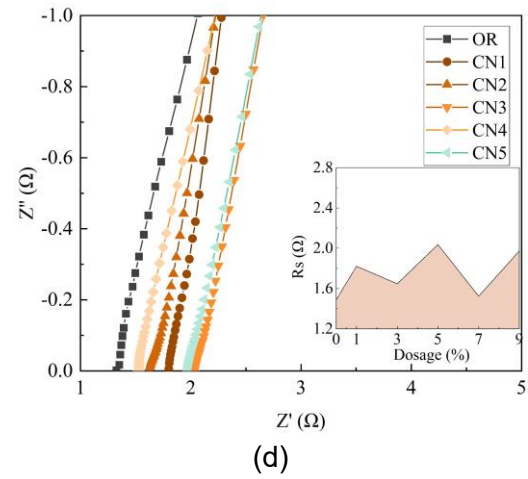
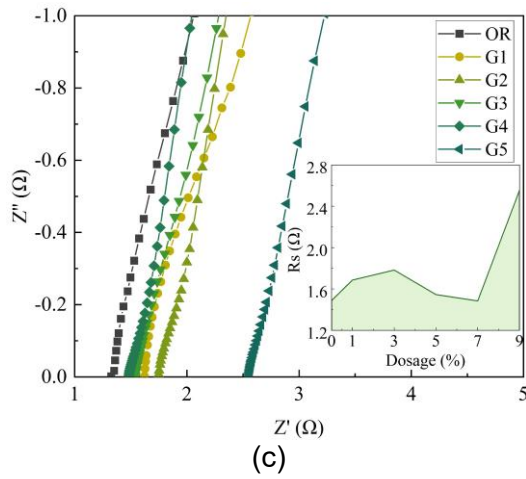
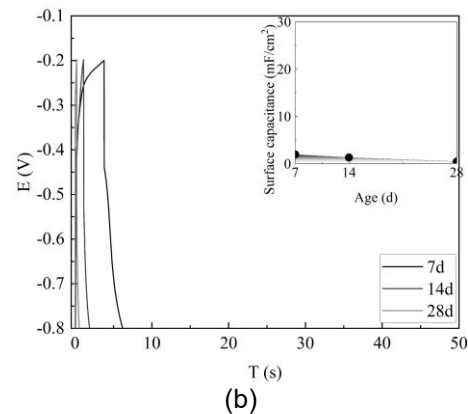
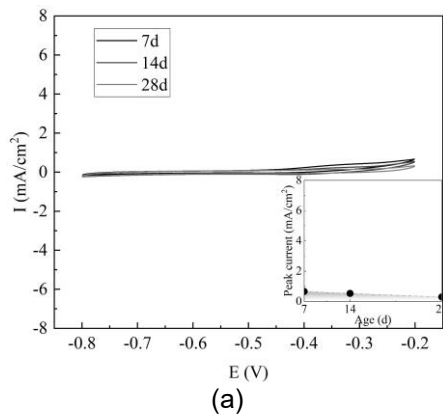


Fig. 5. Electrochemical impedance spectroscopy (EIS) results of solid waste-based electrodes: (a) Z'' vs $\omega^{-1/2}$ curves of GNP-doped specimens and (c) CNT-doped specimens, and (b) Nyquist plots of GNP-doped specimens and (d) CNT-doped specimens at varying doping levels.

3.3. Electrochemical performance trends of specimens with curing age

Fig. 6 illustrates the cyclic voltammetry (CV) and galvanostatic charge-discharge (GCD) curves for the OR (a, b), G5 (c, d), and CN5 (e, f) specimens at different curing ages (7, 14, and 28 days). GNP-doped solid waste cementitious specimens exhibit the highest electrochemical performance at 7 days of hydration, with a peak current density of 7.41 mA/cm^2 and an areal capacitance of 456.9 mF/cm^2 (at a current density of 0.32 mA/cm^2). However, as the curing age extends to 28 days, the areal capacitance decreases slightly to 436.93 mF/cm^2 , corresponding to a reduction of approximately 4.37%. In contrast, CNT-doped specimens show an improvement in electrochemical performance with increasing curing age. At an early stage (7 days), the peak current density is 2.57 mA/cm^2 , with an areal capacitance of 84.4 mF/cm^2 . By 28 days, the areal capacitance rises to 108.89 mF/cm^2 , representing an increase of 29.02%. The differences in areal capacitance trends with curing age will be analyzed further in subsequent sections. Notably, despite the decline in electrochemical performance over time, GNP-doped specimens consistently outperform CNT-doped specimens across all curing ages up to 28 days.



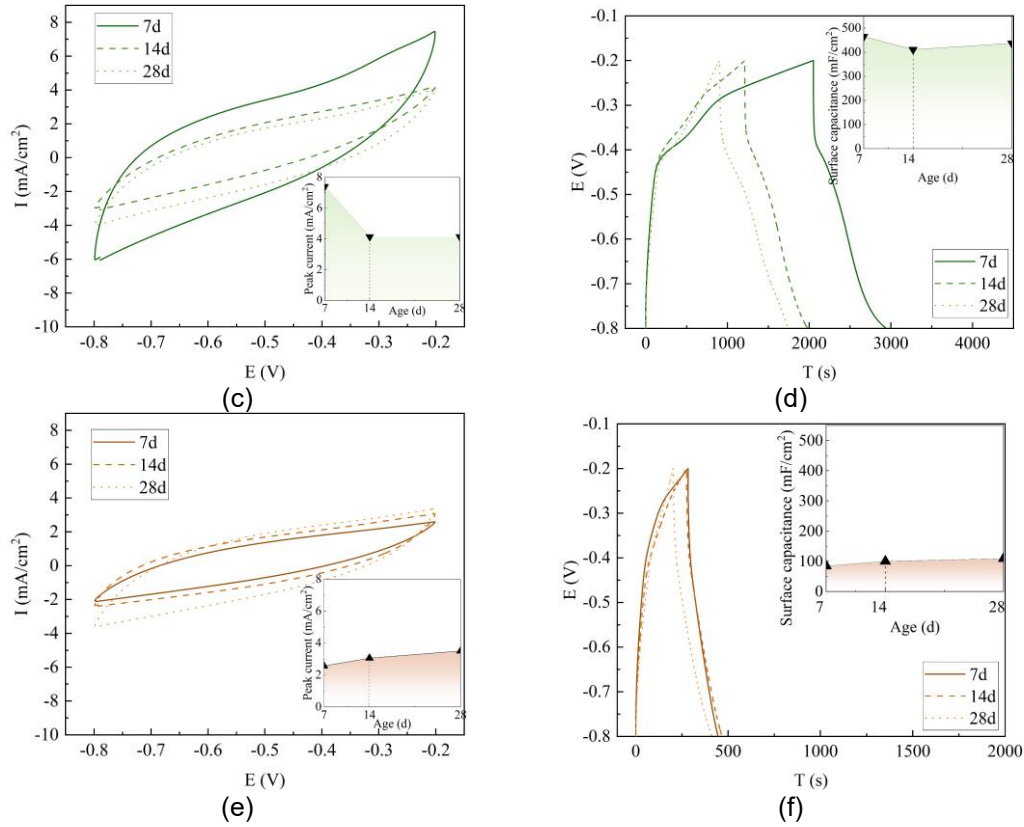


Fig. 6. Cyclic voltammetry (CV) and galvanostatic charge–discharge (GCD) curves of solid waste-based electrodes at different curing ages: (a) CV of the reference specimen (OR), (b) GCD of OR, (c) CV of GNP-doped specimen (G5), (d) GCD of G5, (e) CV of CNT-doped specimen (CN5), and (f) GCD of CN5.

4. Discussion

4.1. Microstructural characterization of conductive networks

To investigate the distribution characteristics of nanoscale structures within the matrix, transmission electron microscopy (TEM) analysis was conducted on specimens with 9% GNP doping and 9% carbon nanotube (CNT) doping using a JEM-2100 TEM. TEM enables the differentiation of nanostructure layer numbers through electron transmission, playing a critical role in studying the morphology and dispersion of GNP-based composites [34]. Fig. 7 presents the TEM images of the G5 specimen (a) and CN5 specimen (b). The good transparency of the GNP sheets (Fig. 7a) allows for the distinction of different layer numbers, indicating that GNP exists in a single-layer or few-layer stacked state within the inorganic cementitious matrix, suggesting excellent dispersion between sheets. The wrinkles in GNP are clearly visible in the transmission images, providing high-conductivity channels for the GNP sheets [35] and imparting superior conductivity to the solid waste electrode. This spatial overlapping behavior of two-dimensional nanosheets, such as inter-plane crosslinking, forms a continuous three-dimensional conductive network within the matrix.

On the other hand, Fig. 7(b) reveals that CNTs form an intertwined three-dimensional network structure in the solid waste cementitious matrix. Some CNTs are visible as individual tubes, while in certain regions, they exhibit rope-like knotted morphologies, resulting from their high aspect ratio, which leads to mutual entanglement and bundling.

In summary, both GNP and CNTs construct three-dimensional conductive networks in the matrix. Macroscopically, this manifests as an increase in areal capacitance from 0.42 mF/cm² to a maximum of 436.93 mF/cm² and 209.58 mF/cm² for solid waste cementitious materials doped with GNP and CNTs, respectively (as shown in Fig. 7).

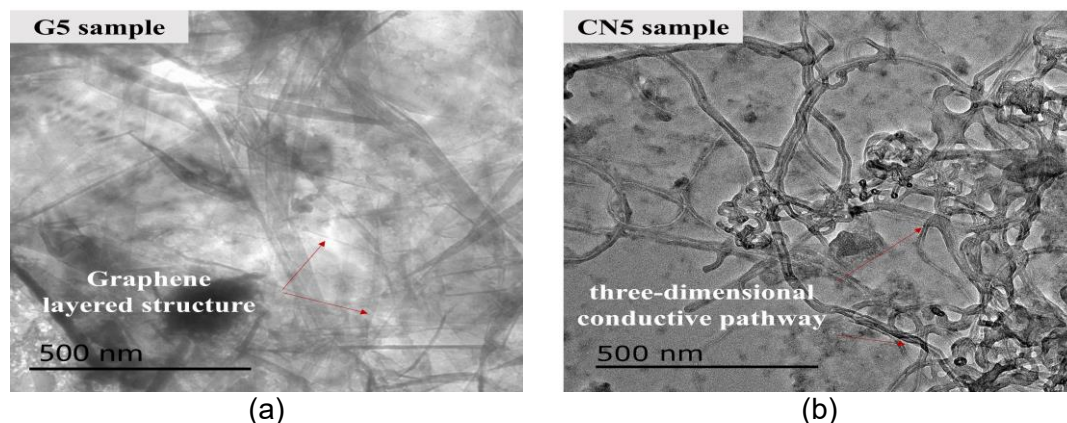


Fig. 7. Local morphologies and contact configurations of GNP and CNT observed by TEM.

GNP exhibited a greater enhancement in areal capacitance than CNT in the present system. This difference is closely associated with their distinct dimensional morphologies^[36,37]. As shown in Fig. 8, TEM observations revealed thin, wrinkled, and partially overlapping GNP sheets, whereas CNT exhibited elongated one-dimensional tubular morphologies with a high aspect ratio. These images represent local electron-transparent fragments.

The two-dimensional lamellar morphology of GNP provides a relatively large electrochemically accessible surface when sufficiently dispersed, while overlapping neighboring sheets can facilitate local electron transport. The wrinkles and folds observed in Fig. 8 may also help preserve exposed surface regions and limit complete restacking of adjacent sheets^[9]. By contrast, CNT forms conductive pathways primarily through the spatial percolation of high-aspect-ratio tubular fillers. The effectiveness of such networks strongly depends on CNT dispersion and connectivity within the cementitious matrix. Accordingly, the higher capacitance obtained with GNP is attributed to its large accessible surface and favorable sheet-based conductive pathways, whereas the fibrous morphology of CNT is more favorable for maintaining network continuity and providing mechanical bridging within the hydrated matrix. The evolution of these conductive networks during hydration is further evaluated by SEM observations in the following section.

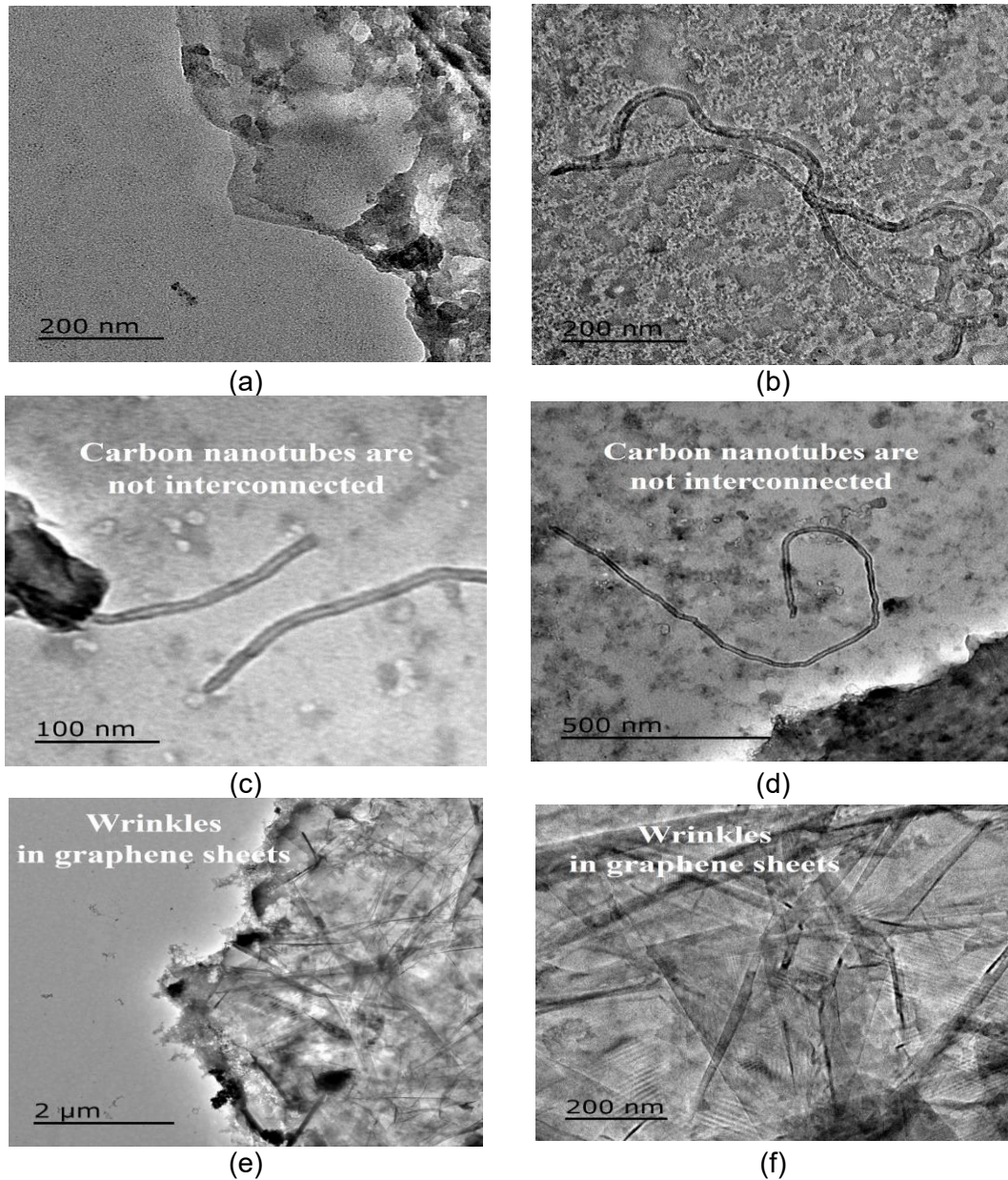


Fig. 8. Representative TEM images showing the local morphologies of GNP and CNT in electron-transparent fragments obtained from hydrated composite specimens: (a) overlapping GNP sheets; (b–d) representative CNT morphologies; and (e, f) wrinkled and folded GNP sheets.

4.2. Pore structure analysis of specimens at multiple scales

To comprehensively evaluate the pore distribution in the specimens, multi-scale observations of the pore structure were conducted. Fig. 9 shows the surface morphology of specimens with 0% (a) and 9% (b) GNP doping under the optical microscope probe. The observations reveal that the pure solid waste cementitious material (0% GNP) exhibits higher surface density, primarily attributed to the formation of compact hydration products during the hydration process (to be analyzed in detail later). In contrast, the specimen with 9% GNP doping displays numerous pores ranging from 0.1 mm to 0.3

mm on the surface, resulting from the generation of macropores induced by excessive GNP doping [38]. Such porosity evolution may also account for the sharp increase in areal capacitance observed between 5% and 7% GNP, as improved ion-accessible pathways enhance charge storage. These pores therefore facilitate easier transport of electrolyte ions into the electrode interior, enhancing the electrochemical performance of the specimens.

Fig. 10 display the logarithmic differential intrusion volume versus pore size (Log Differential Intrusion vs. Pore Size) (a) and cumulative intrusion volume versus pore size relationships (b), respectively. According to the pore classification by Lu et al., 2022; Sun et al., 2024, 2025; Wang et al., 2020^[39–42], the pore structure of cementitious materials is typically divided into four categories: thin mesopores (5-27 nm), coarse mesopores (27-50 nm), medium capillaries (50-100 nm), and supermicropores (0.1-5 μm), micropores (5-30 μm), mesopores (30-75 μm), and macropores (>75 μm). This classification is used to discuss the MIP results in this section. The OR specimen exhibits a predominance of thin and coarse mesopores, with curve peaks at approximately 11.5 nm and 43.8 nm. The G5 specimen (9% GNP doping) shows more medium capillaries and supermicropores, with peaks at about 61.2 nm and 276.3 nm. The CN5 specimen (9% CNT doping) is characterized by supermicropores and micropores, with a broader peak distribution in the 1 μm to 20 μm range. This is attributed to agglomeration caused by excessive CNT doping, which leads to the formation of larger pores [43]. From the pore size distribution curves, an optimal range for charge storage and transport is identified, with pores concentrated in 50-100 nm and 200-400 nm being particularly favorable for these processes.

Table 1 summarizes the pore structure statistics of the specimens. In addition to porosity and pore size distribution, mercury intrusion porosimetry (MIP) provides a range of structural parameters including total intrusion volume, porosity, total pore area, permeability, tortuosity. Representative data are summarized in the Supplementary Information (Table S3). The G5 specimen has a pore volume of 1.02 mL/g and a porosity of 67.80%, significantly higher than the CN5 specimen (0.50 mL/g, 54.60%) and the OR specimen (0.28 mL/g, 40.02%). This ranking aligns with the capacitance test results (G5 > CN5 > OR, with values of 436.93 mF/cm², 108.89 mF/cm², and 0.42 mF/cm², respectively), indicating that pore volume and porosity make substantial contributions to capacitance. Larger pore volume and porosity provide more storage space for electrolyte ions and expand charge transport pathways, thereby enhancing the capacitance of the G5 specimen.

Additionally, the OR specimen has the highest pore surface area of 76.91 m²/g, theoretically offering the greatest potential for charge adsorption. However, its smaller pore sizes (in the 10-50 nm range) and higher pore tortuosity factor (47.47) limit the ion transport rate, hindering timely ion entry into the pores, thereby reducing its electrochemical performance (e.g., areal capacitance of 0.42 mF/cm²). In comparison, the pore size distributions of the G5 and CN5 specimens are more conducive to ion transport, resulting in higher areal capacitances. Notably, the CN5 specimen exhibits the highest permeability (159.93 mdarcy) and the lowest tortuosity factor (13.77), indicating superior

interconnectivity in its internal pore network. This enhanced pore connectivity significantly improves charge transport efficiency. However, the substantial increase in pore size in the CN5 specimen leads to a reduction in specific surface area per unit volume, thereby decreasing the effective surface area available for charge adsorption. Consequently, despite the favorable connectivity, the capacitance of the CN5 specimen remains lower than that of the G5 specimen.

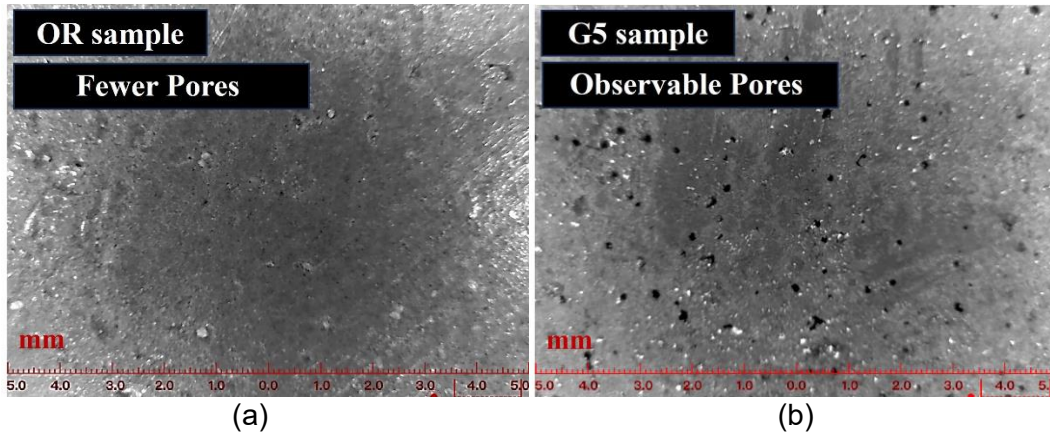


Fig. 9. Surface morphology of solid waste-based specimens under optical microscopy: (a) 0% GNP doping and (b) 9% GNP doping.

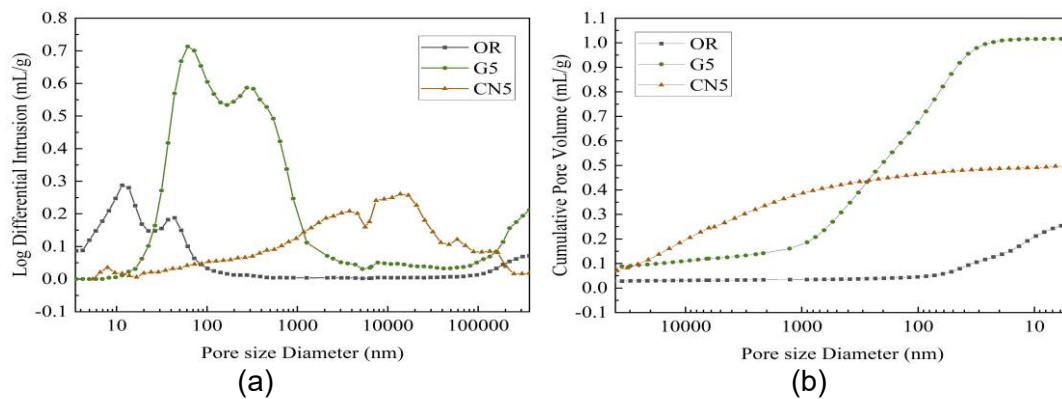


Fig. 10. Mercury intrusion porosimetry (MIP) results of solid waste-based specimens: (a) log differential intrusion volume versus pore size and (b) cumulative intrusion volume versus pore size for OR, G5, and CN5.

Table 1. Pore Structure Summary of OR, CN5 and G5 samples.

Pore Structure Summary	OR	CN5	G5
Total intrusion volume (mL/g)	0.28	0.50	1.02
Porosity (%)	40.02	54.60	67.80
Total pore area (m ² /g)	76.91	7.35	36.52
Permeability (mdarcy)	30.77	159.93	51.91
Tortuosity	47.47	13.77	42.27

4.3. Effect of curing age on the conductive network in specimens

The capacitance of GNP-doped specimens decreases with increasing hydration time, indicating that the continuity of the conductive network is affected during the hydration process of the solid waste cementitious material. Fig. 11 presents the FTIR spectra of GNP-doped specimens at 7 days and 28 days of hydration (a), along with magnified views at 800 cm^{-1} and 1300 cm^{-1} (b). The FTIR analysis reveals O-H stretching and bending vibration peaks at 1636 cm^{-1} and 3422 cm^{-1} , which are characteristic of weakly bound water within the structure, potentially occurring during the formation of hydration products such as C-S-H gel [44]. Additionally, the peak at 1636 cm^{-1} also corresponds to C=C stretching vibrations, overlapping with the O-H bending peak, and is associated with carbon groups in carbon-based materials. As the hydration time increases from 7 days to 28 days, the peak at 1636 cm^{-1} remains relatively stable, suggesting that the C=C structure of GNP does not undergo significant changes, thereby preserving conductivity [35]. The absorption peaks between 1420 and 1483 cm^{-1} represent carbonate bending vibrations, reflecting carbonate-related components due to carbonation reactions from exposure to air. The absorption peak at 619 cm^{-1} is attributed to tricalcium silicate in steel slag [45], resulting from incomplete reaction of tricalcium silicate during early hydration (7 days).

On the other hand, the absorption peak at approximately 1115 cm^{-1} is assigned to the vibration of SiQ^3 units, where Q^3 denotes $[\text{SiO}_4]$ tetrahedra connected to three adjacent Si atoms via oxygen bridges ($-\text{Si}-\text{O}-\text{Si}-$) [46]. After 28 days of hydration, this peak shifts to 1089 cm^{-1} , indicating a shift toward lower wavenumbers and a reduction in the strength of the bridging oxygen bonds ($-\text{Si}-\text{O}-\text{Si}-$) [47]. This may arise from interactions between oxygen-containing functional groups on the GNP surface (such as hydroxyl or carboxyl groups) and the silicate polymers. Due to the presence of minor oxygen functional groups on the GNP surface, these groups bind to the silicate network through coordination or hydrogen bonds, altering the vibrational frequency of Si-O-Si bonds and causing the peak shift. Furthermore, as hydration time extends to 28 days, the absorption peak broadens, reflecting increased disorder or heterogeneity in the silicate network structure. This is due to the gradual accumulation of hydration products (such as Aft and C-S-H) on the GNP surface with prolonged hydration, leading to interfacial interactions between GNP and calcium silicate gel [48], which enhance the heterogeneity of the silicate network structure. Simultaneously, the coverage by hydration products reduces the contact interface between GNP and electrolyte ions, resulting in a decrease in the areal capacitance of the specimen with 9% GNP from 456.9 mF/cm^2 to 437 mF/cm^2 . This phenomenon indicates that the encapsulation of GNP surfaces by hydration products not only affects the microstructure of silicates but also impacts the electrochemical performance of the composite material.

Additionally, in the FTIR spectrum at 7 days of hydration, the peak at 963 cm^{-1} is attributed to Si-O-T (T=Al/Si) stretching vibrations, serving as evidence for the formation of C-(A)-S-H gel hydration

products [49,50]. After 28 days of hydration, this absorption peak shifts to a higher wavenumber, moving to 1031 cm^{-1} , signifying an increase in the degree of polymerization of silicate chains. This represents the manifestation of a three-dimensional network in the synthesized silicate structure [51,52]. The further polymerization of C-S-H gel densifies the matrix, restricting electrolyte ion transport and leading to a decline in capacitance for the GNP-doped specimen after 28 days of hydration.

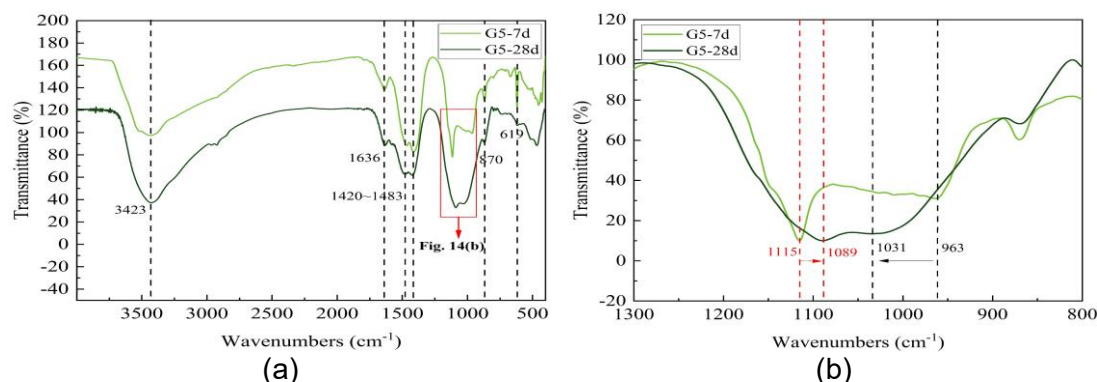


Fig. 11. (a) FTIR spectra of GNP-doped specimens at 7 and 28 days of hydration and (b) magnified FTIR spectra at 800 cm^{-1} and 1300 cm^{-1} .

Fig. 12. presents the SEM images of the G5 (a, b), and CN5 (c ~f), OR (g, h) specimens at 7 days and 28 days of hydration. At 7 days of hydration, the surface of the GNP-doped specimen appears rougher, indicating good dispersion [53], as shown in Fig. 12(a). This suggests that the structural electrode forms a relatively complete three-dimensional conductive network during the early stages of hydration. As the hydration reaction progresses, the internal pores gradually diminish and become denser. Under the encapsulation and compression by hydration products, the GNP sheets undergo a "compaction" phenomenon, resulting in reduced spacing between sheets and subsequent restacking and aggregation, as illustrated in Fig. 12(b). This leads to a decrease in the specimen's capacitance, consistent with the observed reduction in electrochemical performance of GNP-doped specimens with increasing hydration age, as depicted in Fig. 6(d).

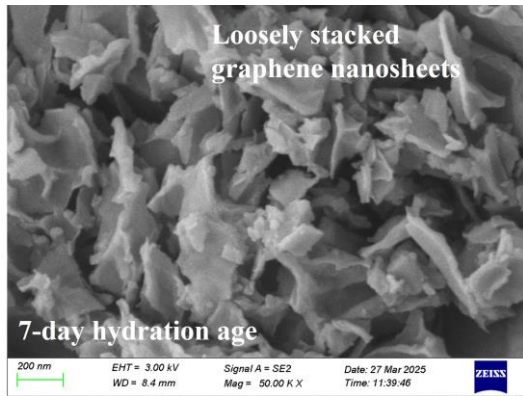
In contrast to GNP, the CNT-doped specimens at 7 and 28 days of hydration exhibit CNTs embedded within the pore channels among hydration products and interwoven with the developing silicate phases, thereby preserving the continuity of the conductive network. It should be noted that CNTs do not grow within the matrix; rather, it is the hydration products that grow around the CNTs. Due to their fibrous morphology and ability to bridge across pores, CNTs experience minimal disruption from the growth of hydration products, in contrast to GNP sheets that tend to be separated or aggregated during hydration.

Furthermore, as the hydration reaction continues and the system pores decrease, the contact points between CNTs increase, enhancing the connectivity and stability of the conductive network under certain conditions, as illustrated in Fig. 12(e) and (f). Such pore closure increases the overall

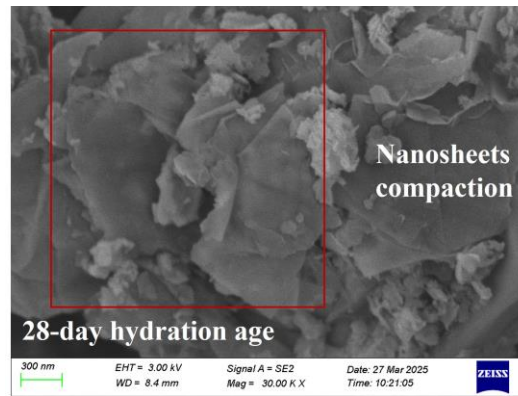
matrix density and reduces the ion-accessible surface area, which accounts for the gradual capacitance decay observed in GNP-doped specimens. In contrast, the fibrous CNTs are capable of threading through the finer pore network formed during hydration, thereby retaining ion accessibility while simultaneously increasing conductive contact points, which explains their long-term electrochemical stability, as shown in Fig. 12(c) and (d).

Additionally, owing to their one-dimensional fibrous morphology, CNTs can bridge adjacent hydration products [54,55], providing a fiber reinforcement effect. Compared to the two-dimensional structure of GNP, CNTs offer greater advantages in bridging microcracks and transferring loads, resulting in higher compressive strength for CNT-doped specimens (CN5, 11.76 MPa) than for GNP-doped specimens (G5, 7.8 MPa), as shown in Fig. 2. This strength reduction in GNP-doped specimens can be attributed to the extremely high surface area of GNP sheets, which, when added in excess, obstructs the growth of hydration products and disrupts the water balance, thereby compromising matrix compactness and compressive strength.

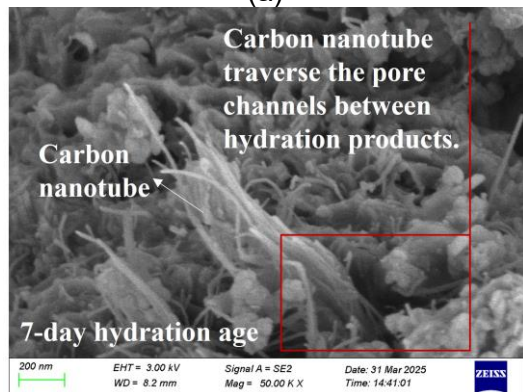
Furthermore, Fig. 12(g) and (h) display the microstructural morphology of the pure solid waste cementitious specimen (OR) at 28 days of hydration. The images reveal the formation of abundant calcium aluminosilicate hydrate (C-A-S-H) products. These hydration products exhibit various typical morphologies, including sheet-like, spherical aggregates, flaky structures, and gel-like forms [56,57]. Through mutual overlapping, filling, and bridging, these products construct a dense and well-connected gel framework within the pores, providing primary mechanical support for the cementitious system. Consequently, the OR specimen demonstrates the best load-bearing capability.



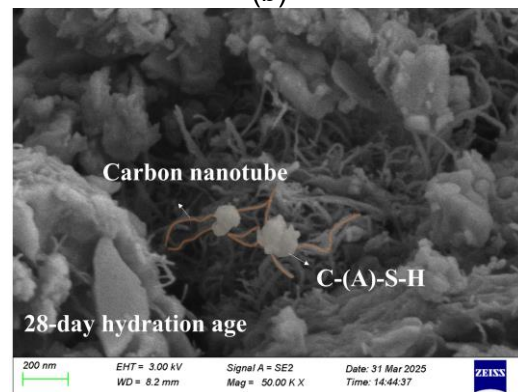
(a)



(b)



(c)



(d)

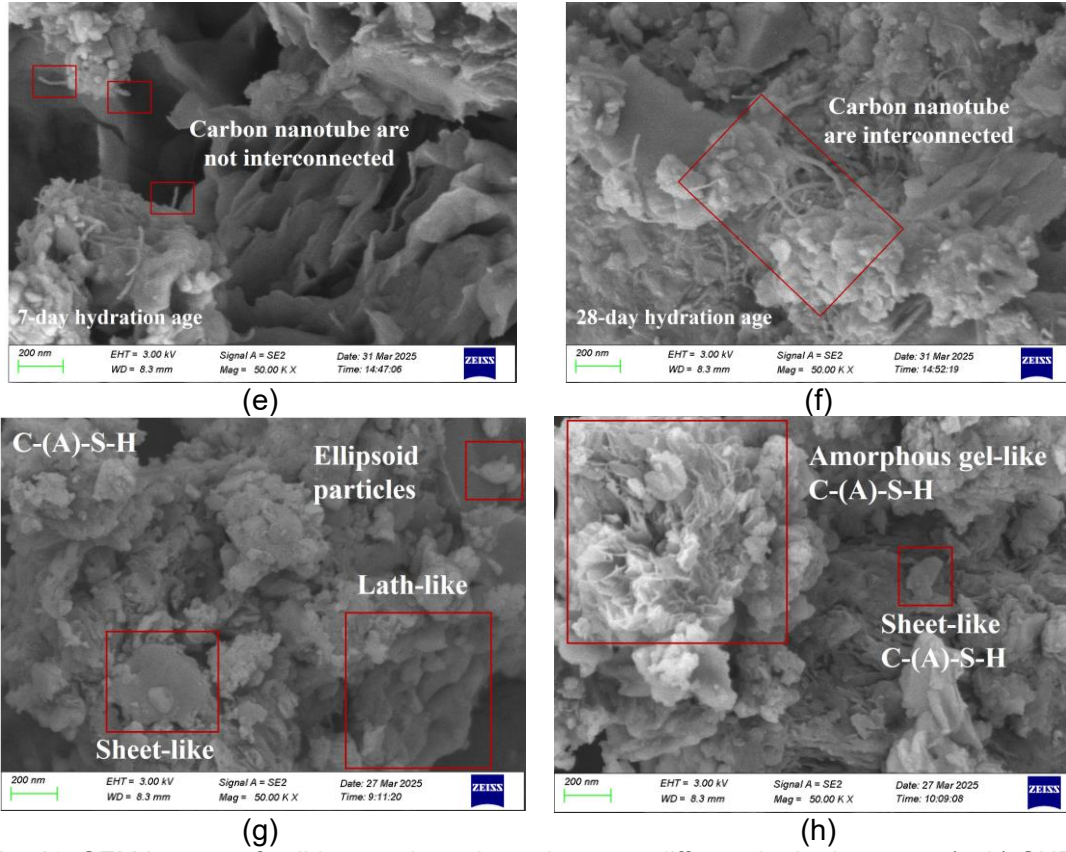


Fig. 12. SEM images of solid waste-based specimens at different hydration ages: (a, b) GNP-doped specimen G5 at 7 and 28 days, (c, d) CNT-doped specimen CN5 at 7 and 28 days (view 1), (e, f) CNT-doped specimen CN5 at 7 and 28 days (view 2), and (g, h) reference specimen OR at 28 days.

5. Comprehensive performance evaluation and assembly application of specimens

To effectively enhance capacitance, increasing the GNP doping level is beneficial; however, it leads to a sharp decline in specimen strength and may even prevent proper molding. In practical applications, structural supercapacitors must satisfy both load-bearing requirements and exhibit superior electrochemical energy storage capabilities. Therefore, to evaluate the comprehensive performance of structural supercapacitors in terms of load-bearing capacity and energy storage performance, a Structure-Energy Integrated Score (SEIS) model was developed. Fig. 13 illustrates the flowchart of the structure-energy integrated score (SEIS) evaluation system.

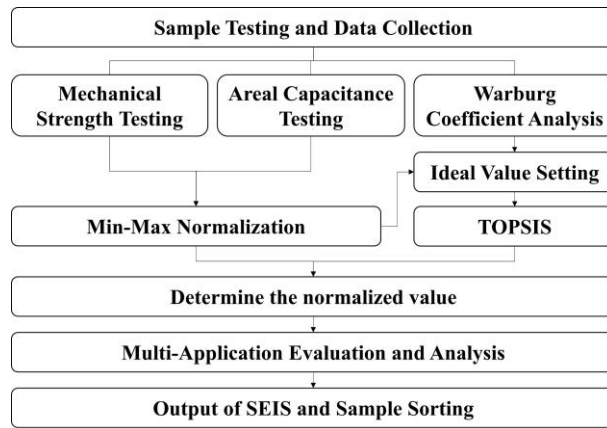


Fig. 13. Flowchart of the structure-energy integrated score (SEIS) evaluation system.

The SEIS framework integrates three key indicators: compressive strength, areal capacitance, and Warburg coefficient (σ). Compressive strength serves as an indicator of load-bearing capacity, areal capacitance represents electrochemical performance, and the Warburg coefficient reflects ion migration resistance—a higher σ value indicates greater resistance and poorer electrochemical performance. Meanwhile, the Warburg coefficient also acts as a coupled indicator reflecting both compressive strength and areal capacitance, as smaller pores lead to denser specimens with stronger load-bearing capacity. Table 2 lists the compressive strength, areal capacitance, and Warburg coefficient values obtained in this study.

Table 2. Compressive strength, areal capacitance, and Warburg coefficient indicators.

Specimens	Compressive strength (MPa)	Areal capacitance (mF/cm ²)	Warburg coefficient indicators ($\Omega \cdot s^{-1/2}$)
OR	21.8	0.41875	180.735
G1	19.7	2.447914	58.505
G2	15.4	10.81596	21.92375
G3	12	50.90281	8.5425
G4	11.53	335.0875	3.5525
G5	7.8	436.9091	2.27125
CN1	21	77.96875	46.135
CN2	16.2	156.8228	8.41
CN3	13.36	209.5833	7.030625
CN4	12.8	72.03128	5.33125
CN5	11.76	108.8888	4.06875

To eliminate challenges arising from different units and preserve the relative magnitudes of the three indicators, normalization was applied. Specifically:

Min-Max normalization (Equation 7) ^[58] was used for compressive strength and areal capacitance to obtain their normalized values (Norm values). Assigning equal weights to energy storage

performance and load-bearing capacity, the Weighted Sum Model (WSM) (Equation 8) was employed. The Warburg coefficient σ corresponding to the specimen with the highest weighted score of compressive strength and areal capacitance Norm values was designated as the "ideal value." In this study, the highest-scoring specimen was CN1, with an ideal Warburg coefficient value of 46.135. Target-based normalization (Equation 9) was then applied to obtain the normalized Warburg coefficient values. The normalized results for compressive strength, areal capacitance, and Warburg coefficient are presented in Table 3.

$$x_n = \frac{x_i - x_{min}}{x_{max} - x_{min}} \quad (4)$$

$$SEIS = \sum_{i=1}^n \omega_i x_i \quad (5)$$

$$x_n = 1 - \frac{|x_i - x_{target}|}{x_{max} - x_{min}} \quad (6)$$

where x_n is the normalized data; x_i is the original data; x_{max} and x_{min} are the maximum and minimum values in the dataset, respectively; x_{target} is the ideal value; SEIS is the comprehensive performance evaluation index; and ω_i is the weight coefficient.

Table 3. Normalized results for compressive strength, areal capacitance, and Warburg coefficient indicators.

Specimens	Norm- compressive strength	Norm-areal capacitance	Norm-Warburg coefficient indicator
OR	1	0	0.245785208
G1	0.85	0.004649	0.930686204
G2	0.542857	0.02382	0.864335194
G3	0.3	0.115659	0.789354981
G4	0.266429	0.766727	0.761394121
G5	0	1	0.754214792
CN1	0.942857	0.177667	1
CN2	0.6	0.358322	0.788612533
CN3	0.397143	0.479196	0.780883373
CN4	0.357143	0.164064	0.771361131
CN5	0.282857	0.248505	0.764286865

**'Norm' refers to normalized.

The Weighted Sum Model (WSM) was employed to conduct a multi-application-oriented evaluation of the normalized values for compressive strength, areal capacitance, and Warburg coefficient indicators. This study established three weight allocation strategies: "structure-biased," "balanced structure and energy storage," and "energy storage-biased," corresponding to typical scenarios prioritizing load-bearing capacity, balancing structural and energy storage performance, and prioritizing energy storage performance, respectively. The weights for the three indicators—compressive strength $\omega_{\text{compressive strength}}$, areal capacitance $\omega_{\text{areal capacitance}}$, and Warburg coefficient

$\omega_{\text{Warburg coefficient}}$ —are assigned as shown in Table 4.

Table 4. Weight coefficients for three indicators.

Scenario	$\omega_{\text{compressive strength}}$	$\omega_{\text{areal capacitance}}$	$\omega_{\text{Warburg coefficient}}$	Description
Structure-dominated	0.5	0.15	0.35	Prioritizes material density and structural safety
Balanced	0.35	0.35	0.3	Equally weights load-bearing and energy storage performance, balancing compressive strength and areal capacitance
Energy-dominated	0.15	0.6	0.25	Focuses on energy storage performance, emphasizing energy density and ion diffusion rate

Note: Weight coefficients can be adjusted based on specific application scenarios.

To validate the WSM model, the Technique for Order Preference by Similarity to Ideal Solution (TOPSIS) ^[59,60] was also applied for analysis. TOPSIS evaluates alternatives based on their geometric proximity to the ideal solution and distance from the worst-case scenario, providing a more nuanced perspective for multi-criteria decision-making. Fig. 14 illustrate the analytical steps of TOPSIS.

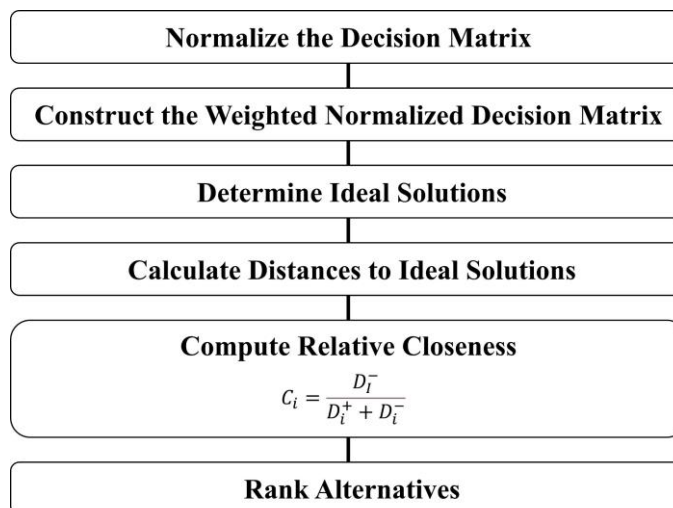


Fig. 14. Technical Roadmap: TOPSIS Method.

Fig. 15 summarizes the SEIS scores for the specimens under the three different scenarios using WSM model (a) and TOPSIS model (b). In the energy storage-biased scenario, GNP-doped specimens at high doping levels exhibit excellent energy storage capabilities, such as the G5 specimen achieving SEIS scores of 0.789 under WSM and 0.791 under TOPSIS. However, this performance gain comes at the expense of reduced load-bearing capacity. The CN-series specimens generally score lower, indicating that CNTs provide less enhancement to energy storage performance compared to GNP.

In the balanced structure and energy storage scenario, the GNP specimen at medium doping

level (G3) has the lowest comprehensive score. Analysis of the electrochemical and strength test results reveals that from G1 to G3, as GNP doping increases, areal capacitance rises while compressive strength decreases. The SEIS score declines because the gain in energy storage performance from increased GNP is insufficient to offset the loss in load-bearing capacity, resulting in the lowest comprehensive score. From G3 to G4, with further increase in GNP doping (when doping >5%), the SEIS score surges to 0.59 (WSM) and 0.537 (TOPSIS), attributed to a significant improvement in areal capacitance for G4 compared to G3, which aligns with reaching the percolation threshold for areal capacitance. At this point, the gain in energy storage performance from GNP addition compensates for the decline in load-bearing capacity. In other words, the key to optimizing carbon-based material doping lies in ensuring that the gain in energy storage performance exceeds the loss in load-bearing capacity.

In the structure-biased scenario, specimens with 1% carbon-based material doping (G1, CN1) score higher than the undoped specimen (OR), with CN1 achieving SEIS scores of 0.848 (WSM) and 0.810 (TOPSIS). This result reveals that low doping levels of carbon-based materials can enhance energy storage effects while maintaining load-bearing stability. This doping range provides a positive energy storage gain, forming an additional "net benefit."

The ranking of scores under WSM and TOPSIS models across all evaluation scenarios shows high consistency, indicating robust data structure and stable evaluation system that are not overturned by switching assessment methods. This validates the robustness and reliability of the evaluation framework. Therefore, regardless of the evaluation model used, the final recommended conclusions are dependable.

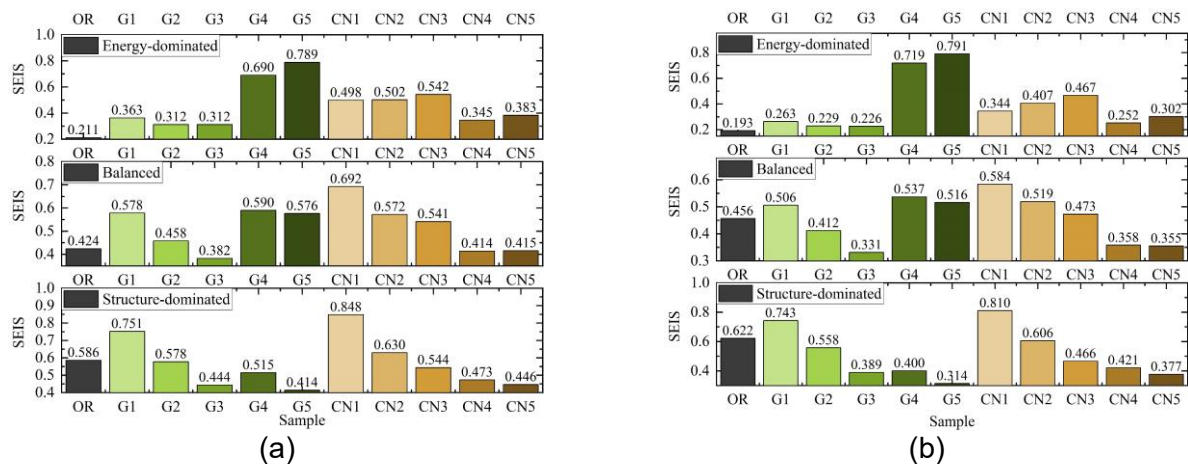


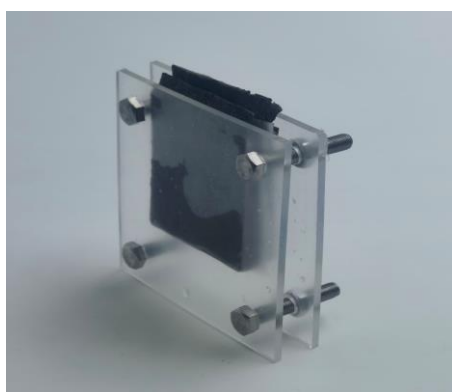
Fig. 15. SEIS scores of specimens under three scenarios using (a) WSM and (b) TOPSIS evaluation models.

To explore the performance of solid waste structural electrodes in practical applications, this study further assembled two structural electrodes (G5 specimens) into a symmetric structural supercapacitor, designated as SSC-G5. Two G5 specimens, each measuring 4 cm × 4 cm × 0.5 cm, were used as the positive and negative electrodes, with filter paper impregnated with 6 M KOH

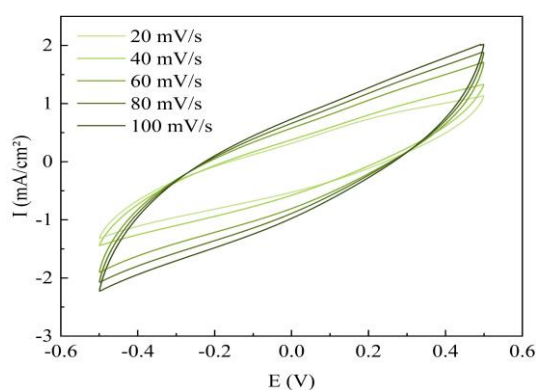
electrolyte serving as the separator. The entire capacitor assembly was fixed using acrylic plates under ambient pressure and temperature conditions, forming a compact symmetric system. Fig. 16 illustrates the assembly and electrochemical performance of the SSC-G5 structural supercapacitor with (a) assembly image, (b) cyclic voltammetry (CV) curves, (c) galvanostatic charge-discharge (GCD) curves, and (d) electrochemical impedance spectroscopy (EIS) plots (-0.5 to 0.5 V, 0.1 mA/cm²).

Electrochemical performance tests revealed that the symmetric structural supercapacitor exhibits favorable electrochemical characteristics under galvanostatic charge-discharge mode. The CV curves, GCD curves, and EIS spectra are presented in Fig. 16(b, c), and (d), respectively. Under a voltage window of -0.5 V to 0.5 V and a discharge current density of 0.1 mA/cm², the capacitor achieves an areal capacitance of approximately 339.06 mF/cm². The calculated energy density is about 0.4 Wh/m², and the power density is approximately 0.36 W/m². These performance metrics indicate that the solid waste structural electrodes, when assembled into a structural supercapacitor, can effectively balance mechanical strength and electrochemical energy storage requirements, making them suitable for distributed energy storage in buildings.

To verify its practical application potential, this study assembled three SSC-G5 capacitors in series to form a capacitor module, which was connected to a DC power supply to illuminate an LED signboard with a rated power of 0.15 W (voltage 3.5 V, current 0.043 A), achieving a Coulombic efficiency of approximately 71.7%. Fig. 17 illustrates the SSC-G5 structural supercapacitor module powering an LED signboard, with (a) showing the setup before illumination and (b) showing the illuminated state. This result demonstrates that the module can support the normal operation of electrical devices, validating the practicality of the solid waste structural electrodes and highlighting their potential in sustainable energy storage fields, such as self-powered systems in smart buildings or environmental monitoring equipment. In future research, energy losses should be considered, and studies could focus on optimizing electrode formulations or increasing carbon-based material doping levels to further enhance energy density and power output, enabling longer-lasting and higher-power supply for electrical devices.



(a)



(b)

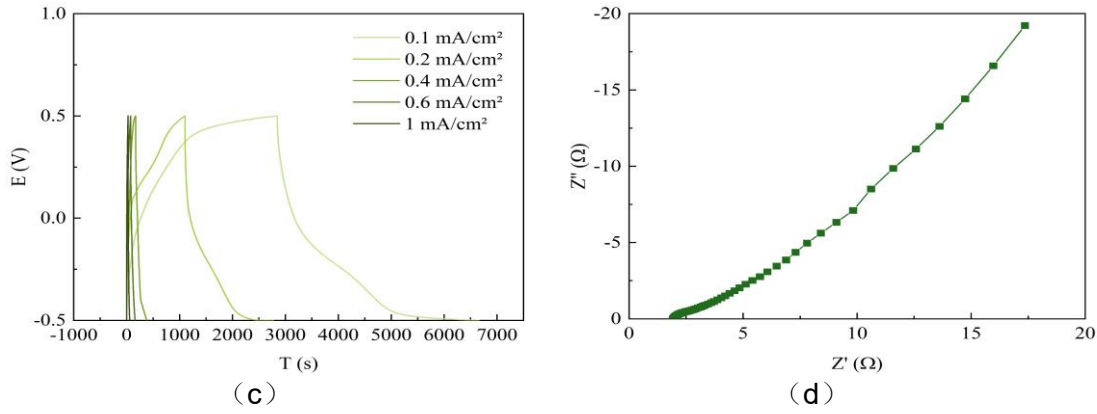


Fig. 16. Assembly and electrochemical performance of SSC-G5 structural supercapacitor: (a) assembly image, (b) cyclic voltammetry curves, (c) galvanostatic charge-discharge curves, and (d) Nyquist plots.

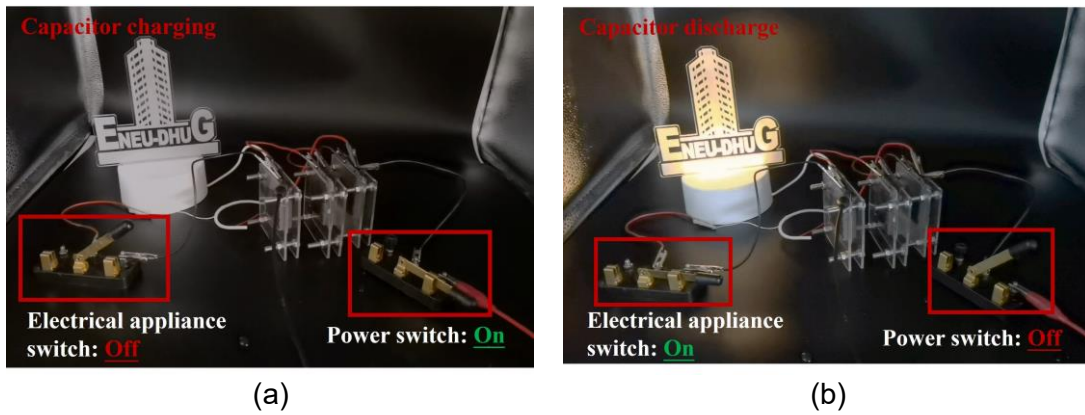


Fig. 17. SSC-G5 capacitor module powering an LED signboard.

6. Conclusion

In this study, composite structural electrodes (CSEs) were fabricated by incorporating graphene nanoplatelets (GNP) and carbon nanotubes (CNT) into an industrial solid-waste-based cementitious matrix. The effects of these two carbon-based materials with distinct dimensional structures on mechanical performance, electrochemical energy-storage behavior, and conductive-network stability during hydration-induced microstructural evolution were systematically compared. The main conclusions are as follows:

(1) Both GNP and CNT markedly enhanced the electrochemical performance of the solid-waste-based CSEs, as reflected by enlarged CV areas, increased areal capacitance, and decreased Warburg coefficients. However, these electrochemical improvements were accompanied by reductions in compressive strength, revealing an inherent trade-off between energy-storage performance and load-bearing capacity.

(2) GNP increased the areal capacitance by up to 1040-fold, reaching 436.93 mF/cm², whereas CNT achieved a maximum increase of 499-fold. The superior capacitance enhancement of GNP is associated with its two-dimensional lamellar morphology, which provides a large electrochemically

accessible surface and facilitates electron transport through overlapping sheet-based conductive pathways. Nevertheless, GNP caused a maximum compressive-strength reduction of 64%, compared with 46% for CNT. The smaller strength loss induced by CNT is attributed to its fibrous morphology, which can bridge hydration products and microcracks and thereby partially preserve the mechanical integrity of the matrix.

(3) Hydration-induced microstructural evolution exerted different effects on the conductive networks formed by GNP and CNT. With increasing curing age, hydration products progressively covered GNP surfaces and promoted sheet agglomeration/restacking, resulting in gradual capacitance attenuation. In contrast, CNT retained better connectivity by bridging pore channels and hydration products, leading to greater electrochemical stability during hydration.

(4) The Structure–Energy Integrated Score (SEIS), constructed from compressive strength, areal capacitance, and the Warburg coefficient, demonstrated that the optimal dosage of carbon-based materials should not be determined solely by maximizing capacitance. Instead, material optimization should balance the gain in energy-storage performance against the associated loss in load-bearing capacity. The proposed SEIS framework provides a quantitative basis for screening and designing multifunctional solid-waste-based structural energy-storage materials.

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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 available from the corresponding authors upon request.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Jun Yuan is employed by Northwest Electric Power Design Institute Co., Ltd. of China Power Engineering Consulting Group.

Author contribution statement

Qian-Tong Tang: Conceptualization, Methodology, Investigation, Data curation, Formal analysis,

Visualization, Writing – original draft. **Wen-Jing Sun**: Conceptualization, Methodology, Supervision, Funding acquisition, Writing – review & editing. **Yue-E Miao**: Supervision, Project administration, Writing – review & editing. **Jie Yao**: Methodology, Validation, Data interpretation, Writing – review & editing. **Yanming Su**: Investigation, Validation, Data analysis. **Peng Jin**: Methodology, Validation, Writing – review & editing. **Mehdi Mehrali**: Scientific discussion, Data interpretation, Writing – review & editing. **Jun Yuan**: Project administration, Resources, Writing – review & editing. **Hilary Rutto**: Investigation, Writing – review & editing. All the authors have approved the final manuscript.

Use of AI statement

None.

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