

RESEARCH ARTICLE

Lightweight, Strong, and Resilient 3D Graphene Metamaterial via a Multi-flow Assembly

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ABSTRACT

Materials aim to integrate excellent properties, including high strength, stiffness, significant elastic deformation, specifically at low density. However, synthetic materials usually involve trade-offs among these characteristics, resulting in distinct categories, such as hard and soft carbon materials, despite sharing identical elemental composition. Here, we demonstrate a lightweight graphene metamaterial fabricated via multi-flow assembly that integrates the mechanical robustness of low-density hard carbons with the elastic deformability of soft carbons. The representative graphene metamaterial features a cuttlebone-inspired lamella-wall architecture. This architecture reasonably strengthens and stiffens the graphene metamaterial, akin to the house-of-cards carbon layer arrangement in hard carbons. The intrinsic superelasticity under huge deformation (90%) is also retained in these graphene metamaterials. Our multi-flow assembly method is facile to prepare varied metamaterials by directly manipulating the arranged texture of individual graphene sheets, paving the way for exploring the unique properties of metamaterials in the macroscopic world and their applications.

1 | Introduction

The quest for lightweight materials with excellent mechanical efficiency has been a central theme in materials science, driven by the demands of modern technologies in aerospace, electronics, and structural engineering applications [1–8]. Achieving a balance among low density, high mechanical robustness, and elastic deformation in carbon materials is particularly challenging, especially under dynamic loading conditions that require materials to maintain structural integrity [1, 3, 9, 10]. Soft carbon materials

mainly consist of layer-stacked graphene sheets with light weight and huge elastic deformability. However, weak and random van der Waals attractions among graphene sheets result in notably inferior compressive strength in soft carbon materials [11–16]. For example, graphene aerogels exhibit superelasticity but limited compressive strength (~5 kPa) at a strain of 50%, constraining their utility in structural applications and wider-range stress sensors [17]. By contrast, hard carbon materials typically exhibit high mechanical robustness for load-bearing applications due to their rigidity from disordered turbostratic carbon layers [18, 19].

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For instance, a high-strength hard carbon nanofiber aerogel was reported with a strength reaching 50 kPa at 70% strain [18]. The synergy of improved compressive strength and excellent superelasticity of carbon aerogels promises a piezoresistive sensor with an enlarged detection range and mechanical robustness, holding potential applications in aerospace cushioning and industrial pressure monitoring [20]. To date, achieving the synergistic integration of high strength and superelasticity in carbon materials remains an unresolved challenge.

Mechanical metamaterials have been shown to combine nanoscale size effects with lightweight lattice structures to achieve optimized strength, stiffness, deformability, and recoverability [9, 21–29]. These microlattices are composed of nanoscale 1D solid rods or hollow tubes, usually produced by a two-photon lithography additive micromanufacturing [24]. Graphene-based metamaterials have recently emerged, mainly fabricated via macroscopic three-dimensional (3D) printing techniques that enable scalable production [30–33]. For example, Zhu et al. reported a 3D-printed metamaterial with exceptional compressibility and high modulus, and Lee et al. achieved a multiscale-architecture composite consisting of 3D-printed metamaterials with octet-truss cells and graphene aerogels [29, 31]. Recently, a laser-engraving technique was also used to fabricate metamaterials with high reversible elongation [14]. The reported graphene-based meta-structures are primarily on a macroscopic scale with basic units made of graphene aerogel fibers, which results in significantly lower compressive performance compared to those mechanically efficient microlattices and even low-density hard carbon materials. Assuming the monolayer graphene sheets as 2D building blocks, similar to the 1D solid rods or hollow tubes employed in mechanically efficient microlattices, directly manipulating the 3D spatial arrangement of the 2D building blocks could prepare versatile metamaterials with tailored architectures, promising for novel mechanical properties.

Here, we propose a multi-flow assembly approach to prepare graphene metamaterials by directing the arrangement of individual graphene sheets. We adjust the multi-flow field to prepare diverse graphene metamaterials. Specifically, a lamella-wall graphene metamaterial (LWGM) engineered via a layered multi-flow pattern exhibits structural homology with cuttlebone architecture. Such a unique lamella-wall architecture endows graphene metamaterials with a near-zero Poisson's ratio and layer-progressive deformation mechanics similar to cuttlebone, both of which enhance the mechanical stability and robustness. The vertically arranged graphene walls collectively bear stress under compression, ensuring efficient load transfer and uniform load distribution. Therefore, the LWGM with a low density of $\sim 10 \text{ mg cm}^{-3}$ displays 2.46-, 6.58-, and 2.24-fold enhancements in compressive strength, modulus, and energy absorption efficiency, respectively, compared with those of porous graphene materials. Moreover, the LWGM inherits intrinsic elasticity from its monolithic graphene bulk, maintaining structural integrity over 5×10^3 deformation cycles. This multi-flow assembly for facile preparation of metamaterials should facilitate the exploration of their mechanical properties and potential applications.

2 | Results

2.1 | Multi-Flow Assembly to Prepare Graphene Metamaterials

Previously reported graphene metamaterials were prepared by 3D printing or laser-engraving with basic units of graphene aerogel filaments [14, 30, 33]. We directly control the arranged texture of single-layer graphene oxide (GO) sheets to prepare graphene metamaterials by a multi-flow assembly approach. Our methodology comprises three sequential stages (Figure S1): (1) multi-flow wet-extruding the GO liquid crystals (GO average size of $30.9 \mu\text{m}$, Figure S2) into a Ca^{2+} -containing coagulation bath to prepare a meta-structured GO hydrogel with hierarchical lamella-wall architectures, (2) chemical reduction using sodium ascorbate solution to selectively remove oxygen functional groups while preserving structural integrity [34], (3) vacuum drying or ambient drying and thermal annealing at 1000°C to obtain bulk graphene metamaterials (Figure S3). Shear-flow assisted assembly has been well developed as an efficient method to align graphene sheets and fabricate high-performance graphene fibers, films, and aerogels [15, 35–37]. The present multi-flow assembly using designable shear-flow patterns has advances in achieving the 3D spatial arrangement of graphene sheets with programmable anisotropic alignment, compared with the previous single-flow assisted axial alignment method.

The multi-flow wet-extrusion of the meta-structured GO hydrogel is illustrated in Figure 1a. In this process, GO liquid crystals are forced to flow through specifically patterned holes (Figure S4). Under the patterned multi-flow shear field, GO sheets are compelled to be aligned by the connected wires, thus forming the corresponding patterned skeleton due to the dynamically stable behavior of GO liquid crystals [38, 39]. Within each individual fractal hole, GO liquid crystals undergo a sudden expansion, leading to the vertical velocity component (z -direction) deviating from the axial direction (x -direction). Such a vertical velocity gradient generates the shear rate dv_z/dz , which compels GO sheets to align along the z -direction and forms the vertical walls with a parabolic arrangement (Figure 1b and Figure S5). The synergy between the predefined multi-flow patterns and the vertically parabolic arrangement of GO sheets in each patterned hole enables control over the hierarchical architectures. The sample prepared via a layered multi-flow approach shows periodic bright-dark layers in polarized optical microscopy (POM) images. This confirms the meta-structure's dual features: a layered pattern with defined spacing (h) and vertically arranged GO sheets in each layer. These components form orthogonally oriented lamella-wall architectures, as visualized in the reconstructed 3D model from micro-computed tomography (μ -CT, Figure 1c; Figure S6, and Movie S1) and scanning electron microscope (SEM, Figure 1d). The two pairs of anisotropic streaks in small-angle X-ray scattering (SAXS) patterns obtained by x-ray exposure of the lamella-wall region also demonstrate the orthogonally oriented architectures (Figure S7). The orientation order of the vertical walls reaches 0.8. The resultant graphene metamaterial also keeps the lightweight nature ($\sim 10 \text{ mg cm}^{-3}$) while maintaining the structural integrity (Figure 1e).

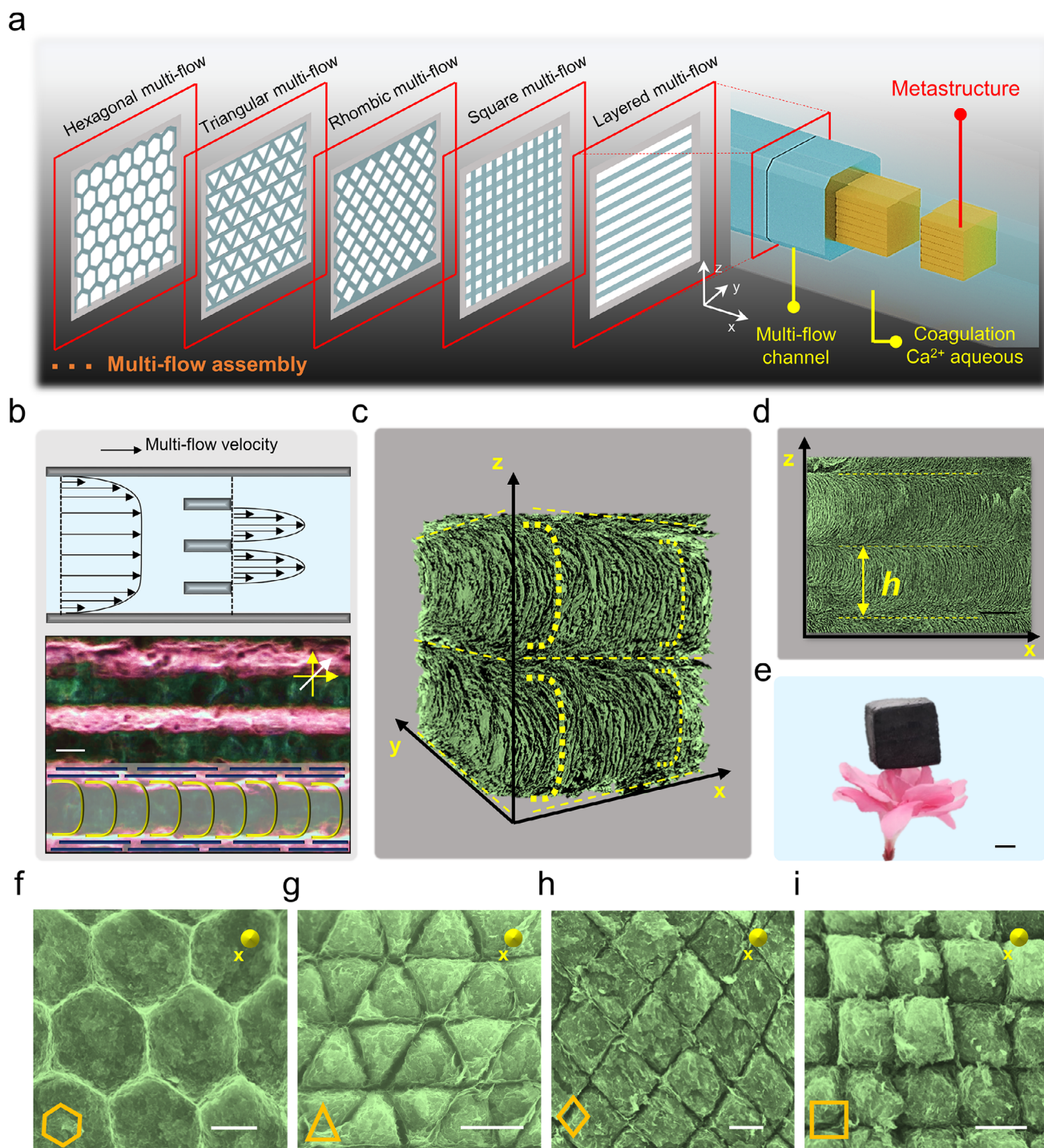


FIGURE 1 | Fabrication of graphene metamaterials via multi-flow wet-assembly. (a) Schematic illustration of the facile fabrication process for graphene metamaterials, involving the wet-extrusion of GO liquid crystals through a customized multi-flow channel followed by coagulation. (b) Schematic of velocity distribution within individual holes of the multi-flow patterns, where the center regions have the highest velocity but lowest shear rate, while the frictional forces in the boundary layer result in much lower fluid velocity at edges compared to the center, yielding a GO liquid crystal texture with alternating light and dark bands. (c) 3D reconstructed structure of the LWGM-1000. (d) SEM image of LWGM in the x - z plane, where the layer distance is denoted as h . (e) Digital photo of the fabricated 10 mg cm^{-3} LWGM standing on a flower, indicating its ultralow density. (f-i) Cross-sectional SEM images (y - z plane) of PGMs fabricated through varied multi-flow channels with designed patterns: hexagonal multi-flow (f), triangular multi-flow (g), rhombic multi-flow (h), and square multi-flow (i). Scale bar, $50 \text{ }\mu\text{m}$ (b), $500 \text{ }\mu\text{m}$ (d), 1 cm (e), $300 \text{ }\mu\text{m}$ (f-i).

Graphene aerogels have been effectively prepared by directional freezing, emulsion templating, and foaming [15, 37]. Despite the cellular structure by emulsion templating and foaming, lamellar structures with aligned graphene sheets were achieved by directional freezing, especially with a precise freezing temperature gradient. Our multi-flow assembly directly fabricates graphene metamaterials with cuttlebone-inspired lamella-wall architecture by designing the shear-flow patterns, not relying on a precise temperature gradient (Figure S8). Therefore, the multi-flow assembly is facile to scale up the preparation of large-size LWGMs as shown in the bulk sample (30 mm × 50 mm × 100 mm) and the strip sample (3 mm × 50 mm × 180 mm), both of which exhibit uniform and long-ranged lamella-wall architectures (Figure S9). Designing the multi-flow patterns also enables fabrication of programmable graphene metamaterials (PGM), including hexagonal, triangular, rhombic, and square architectures with embedded vertical graphene walls (Figure 1f–i and Figure S10).

2.2 | Layer-By-Layer Compressive Deformation of LWGM

Cuttlefish survive in the deep-sea environment under strong hydrostatic pressure due to their natural biomineralized porous bone, which has exceptional strength, stiffness, and energy-absorption capability [40, 41]. This structure features vertically aligned aragonite walls separated by chitinous lamellae, representing a lightweight yet mechanically robust 2D-layer metamaterial (Figure 2a–c). Under compressive deformation, the structure undergoes progressive layer-by-layer collapse, which effectively prevents catastrophic failure (Figure S11). Similar to the mechanically efficient cuttlebone, our LWGM has a layered structure combining the horizontal graphene lamellae with the vertical graphene walls (Figure S12), as confirmed by the reconstructed 3D structure in μ -CT and digital photograph (Figure 1c; Figure S13 and Movie S1). Such a cuttlebone-inspired lamella-wall structure stabilizes and reinforces the lightweight graphene metamaterials, which are distinct from conventional fragile porous graphene materials.

The compressive performance of our representative LWGM along the vertical walls (z -axis) is illustrated in Figure 2d. From the compressive stress–strain curve, the LWGM exhibits a high Young's modulus of 388 kPa under compressive loading at an initial strain of 10%. Real-time optical observation (Movie S2, Insets 1–5 of Figure 2d) documents that the sequential lamellae collapse initiates from the surface layers and propagates throughout the whole bulk via the graphene wall buckling. At 30% strain, it is evident that the bottom layers are compressed, yet the middle layers remain undeformed, demonstrating the layer-by-layer compressive deformation. This layer-by-layer deformation results in a relatively flat plateau region at a stress of 40–80 kPa occurring from 10% to 60% strain, which corresponds to the progressive deformation of the vertical graphene walls. The layer-by-layer deformation not only enhances the metamaterial's fracture resistance but also contributes to a significant energy absorption capacity. As the deformation increases further, the structure undergoes densification, leading to a sharp rise in modulus. The metamaterial shows no obvious change in size in the x – y plane even under 90% compression, exhibiting a near-zero Poisson's ratio ($-\epsilon_x/\epsilon_z \approx 0$) as demonstrated in Movie S2.

Upon unloading the compressive stress, the structure of the graphene metamaterial almost fully recovers, indicating that despite the presence of vertical graphene walls, the metamaterial retains the excellent rebound elasticity characteristic of graphene sheets. The LWGM is mechanically anisotropic with significantly different compressive properties in the three orthogonal directions. Compression along the vertical walls exhibits the optimal values for modulus, strength, and energy absorption (Figure S14). These vertically aligned graphene walls with identical height effectively and uniformly bear external loads and significantly dissipate energy during compression, resulting in their high compressive strength. The deforming graphene sheets store elastic energy without obvious cracks during compression, enabling the superelasticity of LWGMs (Figure S15a,e). In contrast, traditional cellular structure containing randomly oriented walls usually suffers from irregular multi-directional loads, potentially leading to early stress concentration and premature failure of walls and their joints (Figure S15b,f); previous lamellar structure without supporting walls generally exhibits weak compressive strength (Figure S15c,g).

To correlate the mechanical behavior of the metamaterials with their microscale architectures, in situ SEM was also conducted under cyclic compressive strain up to 70% (Figure 2e). The microscale deformation is in accordance with the layer-by-layer compression shown in Figure 2d. During compression, no obvious damage to the graphene lamellae was observed, indicating strong structural integrity. Between each pair of adjacent lamella layers, the collapse of graphene walls results from the sheet buckling under compression. The vertical graphene walls do not exhibit obvious breakage like the walls in cuttlebone, thus almost recovering after strain release (Figure 2e and Movie S3). As a result, the LWGM not only possesses excellent mechanical robustness, strength, and energy absorption capacity but also achieves a significant elastic recovery advantage that natural cuttlebone lacks. We systematically compared the compressive behavior and deformation of the LWGMs with lamellar aerogel (horizontally aligned graphene sheets) and wall aerogel (vertically aligned graphene sheets, Figure S16). Lamellar aerogel always exhibits an inferior compressive modulus and strength because of the large space without supports between adjacent lamellae (Movie S4). Wall aerogels with only vertical graphene walls usually show obvious negative stiffness, which indicates the structural instability by local and huge buckling, leading to reduced compressive strength and unrecoverable deformation. In situ SEM tracking of the wall aerogels intuitively illustrates the large-area local buckling of vertical graphene walls (Movie S5). The neighboring vertical graphene walls without fixing could undergo severe splitting and delamination after compression (Figure S15d,h). These unrecoverable premature cracks contribute to the reduced compressive strength and elasticity in wall aerogels. Our LWGM-100 has pre-curved vertical walls along the flow direction and connected joints between vertical walls and horizontal lamellae, guaranteeing the structural integrity.

2.3 | Tunable Mechanical Performance

The LWGM features with layers of vertical graphene walls separated by horizontal graphene lamellae. We fabricated

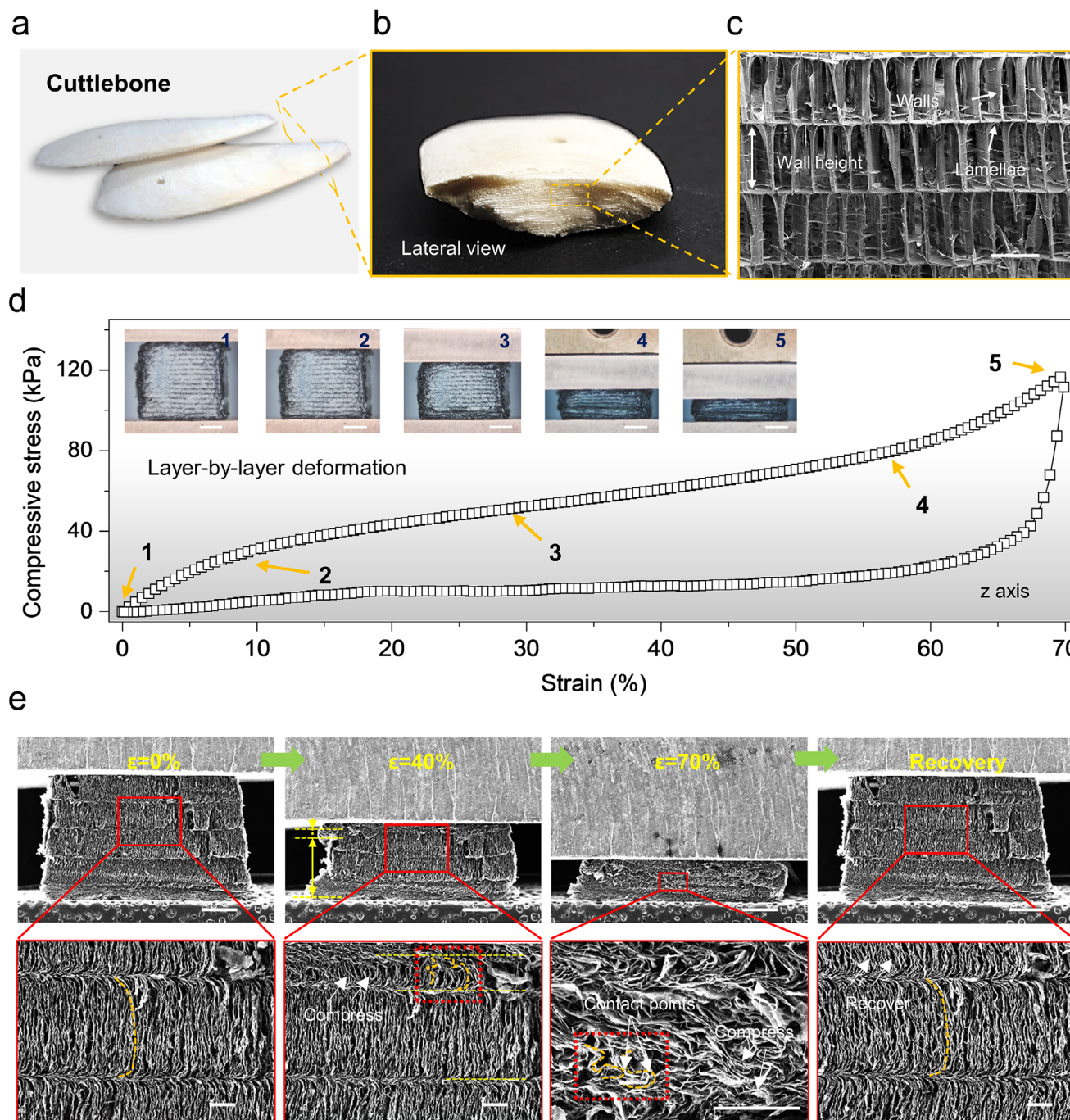


FIGURE 2 | Compression mechanism of the cuttlebone-inspired LWGM. (a,b) Optical images of a cuttlebone and its lateral section, respectively. (c) SEM image showing the hierarchical structure with parallel septa separated by uniformly distributed vertical walls. (d) Typical compressive stress-strain curves along the z -axis for LWGM-1000. The inset snapshots illustrate the layer-by-layer deformation. (e) SEM images of LWGM-1000 during the uniaxial compressive test. Magnified SEM images show the intact lamellae and sheet buckling in the vertical walls in compression (strain = 70%), and recovered vertical graphene walls upon releasing. Scale bar, 200 μm (c), 2 mm (insets of d), 1 mm (top of e), 200 μm (bottom of e).

LWGMs with varied layer distance h between adjacent horizontal graphene lamellae, also denoted as the length (L_v) of vertical graphene walls, by tuning the thickness of the layered multi-flow pattern, including $h = 100, 200, 400, 600$, and $1000 \mu\text{m}$. The mechanical properties of the graphene metamaterials were easily tuned based on the varied layer distance h , which is labeled as LWGM- h for clarity. LWGMs display higher compressive strength and modulus than graphene

aerogels with random sheet arrangement at the same density ($\sim 10 \text{ mg cm}^{-3}$), owing to the uniform and collective load-bearing in their vertically aligned graphene walls under compression (Figure 3a,b). Conversely, randomly distributed graphene sheets in the control sample of traditional graphene aerogels undergo uneven bending with stress concentration and premature cracks under compressive strain, leading to inferior compressive strength.

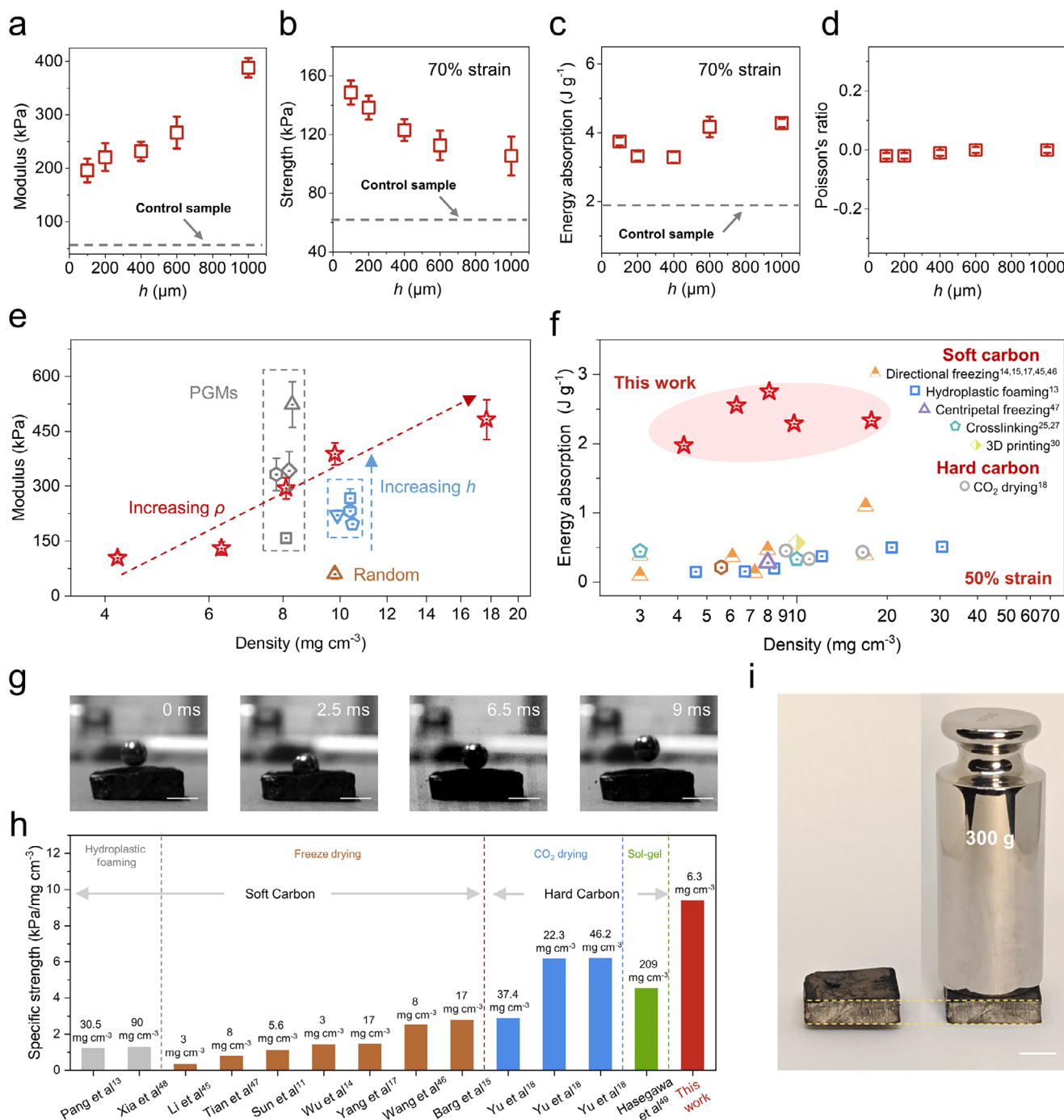


FIGURE 3 | Mechanical properties of LWGMs and PGMs. (a–d) Mechanical properties of LWGMs at varied layered heights: modulus (a), strength (b), energy absorption (c), and Poisson's ratio (d). (e) Tunable modulus versus density, including LWGMs-h, PGMs, and control samples with random graphene arrangement. (f) Comparison of energy absorption versus density between LWGMs-1000 (50% strain) and other carbon-based materials [13–15, 17, 18, 25, 27, 30, 42–44]. (g) High-speed camera snapshots of a small ball rebounding on LWGM-1000. (h) Comparison of specific strength at a strain of 50% with other soft carbon materials and hard carbon materials [11, 13–15, 17, 18, 42–46]. (i) Photographs show that the LWGM-1000 supports ≈ 4000 times of its weight without obvious deformation. Scale bar, 1 cm (g and i). Each test was conducted using three independent samples ($n = 3$).

We systematically investigated the h -dependent compressive behavior across LWGM variants ($h = 100$ – $1000 \mu\text{m}$). A distinct h -modulus-strength correlation emerges: modulus increases 1.9 times (196 to 388 kPa) while strength decreases by 29% (148.7 to 105.4 kPa) as h increases from 100 to 1000 μm . We attributed these performance differences to the different morphologies of vertical graphene walls in graphene metamaterials. As shown in

Figure S12, the vertical graphene walls between each adjacent graphene lamellae in LWGM-100 have a parabolic morphology, whereas those in LWGM-1000 are straighter. In the compressive deformation of LWGM-1000, the straight vertical graphene walls have to bend left or right exclusively, resulting in abundant steric hindrance preventing further deformation and the intuitively enhanced compressive modulus. On the contrary, graphene walls

in the LWGM-100 sample have a curved parabolic morphology, which makes them more susceptible to bending along the curvature direction and consequently reduces the compressive modulus. For further verification, we also prepared alternating LWGM with alternative 200 and 400 μm (a-200-400) and gradient LWGM with ordered 200, 300, and 400 μm (g-200-300-400) by designing the layered multi-flow pattern (Figure S17). The local deformation of graphene walls with larger h always falls behind the overall deformation as observed from in situ compressive SEM tracking, confirming their higher compressive modulus (Movies S6 and S7). LWGMs with lower layer distance h show improved compressive strength, which arises from the more uniform stress distribution in the curved parabolic graphene walls, as also shown in the finite element analysis (FEA) results (Figure S18). By contrast, the conventional cellular structures exhibit more pronounced stress concentration during compression, indicating less efficient load transfer and a higher susceptibility to fracture (Figures S15b and S19).

Given the unique layer-by-layer deformation that prevents catastrophic failure in a manner similar to cuttlebone, our LWGMs also show a high energy absorption capacity ranging from 3.29 to 4.28 J g^{-1} at 70% strain, which is 72% to 124% higher than that (1.91 J g^{-1}) of the control sample with random graphene sheets (Figure 3c). Both the specific strength and the energy absorption capacity of our LWGMs are comparable to those of natural cuttlebone (Figure S20). Moreover, unlike cuttlebone, whose vertical walls undergo irreversible collapse upon loading, the graphene sheets in the LWGM store elastic energy during compression and rapidly recover upon unloading, enabling the superelasticity. In addition, the LWGMs exhibit a near-zero Poisson's ratio under the unique layer-by-layer deformation (Figure 3d). The compressive modulus of the fabricated graphene metamaterials was plotted versus apparent density (Figure 3e). The increased density results in simultaneously improved modulus and strength (Figure S21). As the density increased to 17.7 mg cm^{-3} , the stress of LWGM-1000 at compressive strain of 50% reaches 140.2 kPa, and the modulus is 482 kPa. The LWGM exhibits a 115% improvement in energy absorption compared to control graphene aerogel [15], highlighting its potential for application in highly efficient energy-dissipating materials (Figure 3f and Figure S21). Compared with previously reported hard carbon aerogels, the energy absorption of LWGM-1000 is improved by 418% [18]. For example, the LWGM-1000 can rebound a steel ball (5.2 g, falling distance of 0.7 m) with a recovery speed of 938 mm s^{-1} , higher than other low-density (<10 mg cm^{-3}) carbon aerogels (Figure 3g) [18, 47]. Diverse programmable graphene metamaterials were also prepared by designing the multi-flow pattern, which shows tunable mechanical properties (Figure 1f–i and Figures S22,S23). Specifically, PGMs with a triangular skeleton together with internal vertical graphene walls have unique structural stability with ultrahigh compressive modulus of 451–532 kPa along both x - and z -axis at a low density of 8.3 mg cm^{-3} (Figure S22c). But the tendency for sliding and permanent deformation of the polygon components causes inferior elasticity in the PGMs compared with LWGMs.

The hierarchical meta-structure in LWGMs significantly enhances mechanical properties, exhibiting a specific strength higher than that of both low-density hard carbons and graphene aerogels (Figure 3h; Figure S24, and Tables S1–S4). Due to the

uniform stress distribution in the lamella-wall architecture, our graphene metamaterials show higher mechanical efficiency. LWGM-1000 with a low density of 9.8 mg cm^{-3} has a high compressive strength of 79.1 kPa and high modulus of 388 kPa, 140% and 220% improved compared with previous graphene aerogel [37], which has never been claimed in both soft and hard carbon bulks at the same low density (Figure S21d). As demonstrated in Figure 3i, this ultralight material maintained structural stability under compressive loads equivalent to 4000 $\times$ its own weight with minimal deformation.

2.4 | Strong and Elastic LWGMs for High Range Detection

To evaluate the fatigue resilience of LWGMs, cyclic loading–unloading tests were performed at compressive strains of 50% and 70% (Figure 4a and Figure S25). After 5×10^3 cycles at 50% strain, LWGM-100 retained over 92% of its initial maximum stress (Figure S26). Under more rigorous conditions (5×10^3 cycles at 70% strain), LWGM-100 still maintained 86% of its peak stress without significant height reduction, higher than the stress retention of the control sample with random sheet orientation (38%, Figure 4a,b). Stress decay occurred primarily in early cycles, with the structure stabilizing in subsequent cycles. We calculated the energy dissipation ratio during compression (Figure S26). The first cycle yielded a high energy loss coefficient of 0.65, which quickly dropped and stabilized at 0.61 in the following cycles. Therefore, our LWGM-100 possesses both high strength and high strength retention, as well as high resilience, surpassing previously reported graphene-based porous materials (Figure 4c and Table S5). This exceptional recovery from repeated compressive loading highlights its durability, making it suitable for applications demanding high resilience, such as detection, cushioning, and impact-absorbing materials.

The high compressive resilience and mechanical strength of our LWGMs provide the stability and dependability required for stress sensors with high detection ranges based on the piezoresistive principle [48]. The piezoresistive response originates from the progressive densification of the lamella-wall network. As compression proceeds, adjacent graphene walls contact closer and are layer-by-layer densified, increasing the number of contact points between graphene sheets, thereby reducing the effective resistance of the connected network (similar to the series resistance with in turn reduced resistance, Figure S27). The pressure sensitivity S of LWGMs is given by the formula $S = (\Delta I/I_{\text{off}})/\Delta P$, where ΔI is the current change, I_{off} is the initial current, and ΔP is the pressure variation [18, 49]. As depicted in Figure 4d, the sensitivity graph highlights two phases as pressure increases: the first phase at $P < 20$ kPa, with a sensitivity of $S_1 = 0.0047 \text{ kPa}^{-1}$, and the second at $20 \text{ kPa} < P < 140$ kPa, with a higher sensitivity of $S_2 = 0.017 \text{ kPa}^{-1}$. This suggests that LWGMs enable real-time electrical signal detection at elevated pressures. The current–voltage (I – V) behavior measured from -1 to 1 V, demonstrates a linear relationship, with the slope steepening as pressure increases, indicating continuous resistivity decline (Figure 4e).

We calculated the resistance change ratio ($\Delta R/R_0 = (R_0 - R_p)/R_0$) as a function of pressure, where R_0 is the initial resistance, and R_p is the resistance under applied pressure. Over the

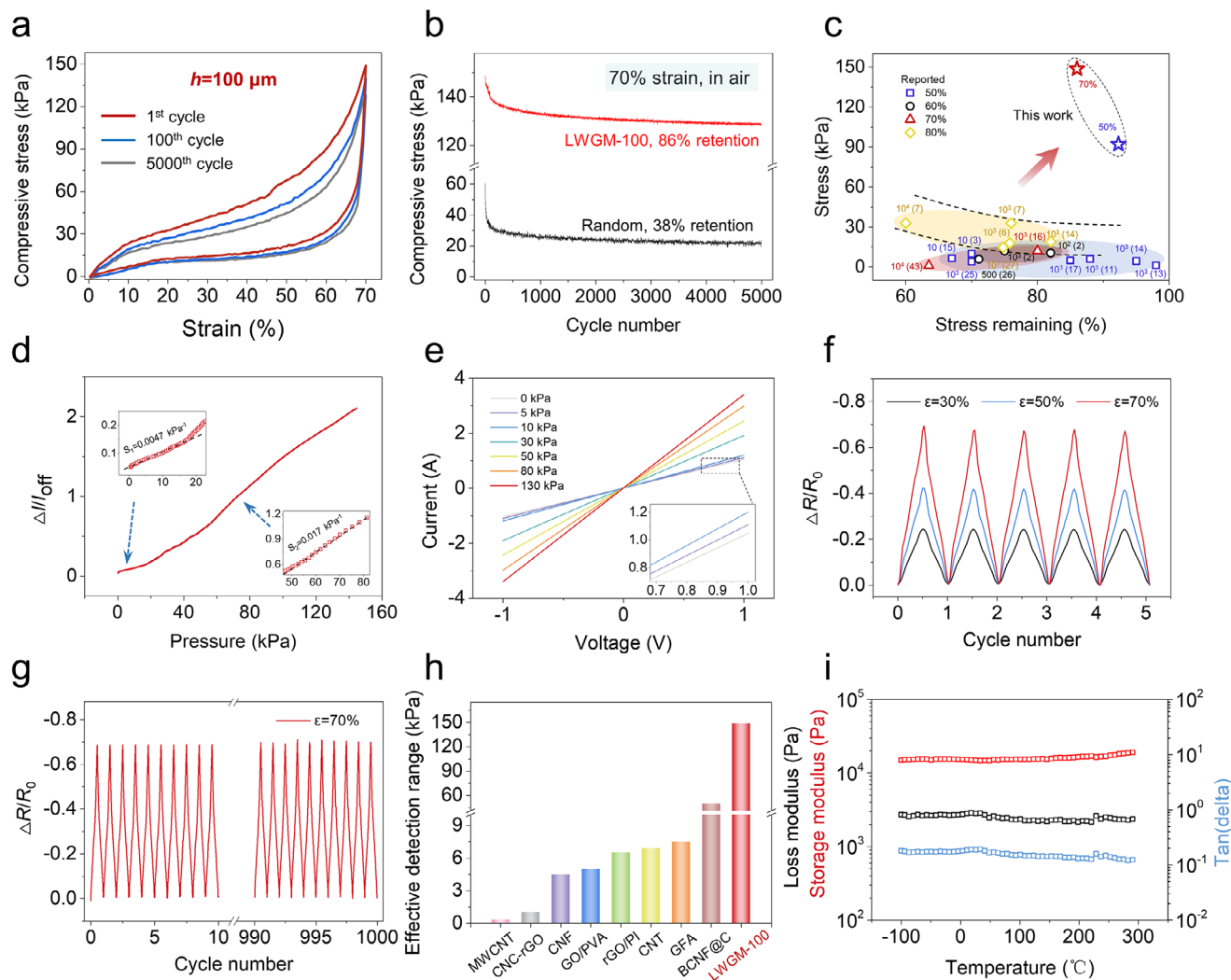


FIGURE 4 | Cyclic resilient property and the electromechanical performances of LWGM-100. (a) Stress–strain curves of LWGM-100 at 70% strain for 5×10^3 cycles. (b) Compressive stress change of LWGM-100 and control samples after 5×10^3 cycles at 70% strain, demonstrating improved mechanical stability. (c) Comparison of stress remaining versus stress during different compression cycles for LWGM-100 and previously reported graphene aerogels [2, 3, 6, 7, 11, 13–17, 25–27, 48]. (d) Current change of LWGM-100 under applied pressures. The insets highlight two distinct linear regions with different sensitivities. (e) I – V curves of LWGM-100 under various pressures. (f) Five-cycle tests of change in resistance under repeated loading and unloading pressure at different strains. (g) Cycling stability test of LWGM-100 under applied strain of 70% for 1000 cycles. (h) Comparison of effective detection ranges among reported sensors. (i) Temperature dependence of storage modulus, loss modulus, and $\tan \delta$ of LWGM-100.

first five cycles, the ratio of resistance change keeps at 24%, 42%, 69% for compressive strain of 30%, 50%, 70%, respectively (Figure 4f). Long-term cycling tests (10^3 cycles) confirmed the sensor's stability, showing no significant signal decay in the last 10 cycles and demonstrating the structural robustness and durability with a baseline drift $<2.5\%$ (Figure 4g and Figure S28). The sensing performance in the low-pressure regime below 1 kPa was also measured. The current change varies approximately linearly with pressure, with a sensitivity of $S = 0.0046 \text{ kPa}^{-1}$, and remains stable over the first 100 cycles (Figure S29). Despite the relatively lower sensitivity at the low-pressure regime due to the high modulus, our LWGMs should be a mechanically robust piezoresistive sensor for high-range pressure detection by taking full advantage of the record-high compressive strength and superelasticity. The effective detection range is defined as the pressure range over which resistance changes dynamically. Although carbon-based materials previously exhibited higher

pressure sensitivity, the effective detection ranges were limited due to their lower compressive strength. LWGM-100 achieves a detection range of 148 kPa at 70% strain, outperforming that of other porous hard materials and graphene aerogels (Figure 4h).

We further evaluated the dynamic sensing behavior of LWGM-100, including stepwise loading–unloading tests, frequency-dependent electrical response, and electrical hysteresis analysis under cyclic loading. As shown in Figure S30, the sensor exhibits rapid and repeatable resistance changes under stepwise pressures of 1, 2, 5, and 10 kPa, with response and recovery times of 0.2 s and 0.18 s at 1 kPa, and 0.25 s and 0.21 s at 10 kPa, respectively. In addition, the electrical output can reliably follow periodic compression over the frequency range of 0.1–2 Hz, showing stable and repeatable $\Delta R/R_0$ signals (Figure S31a). The hysteresis behavior under cyclic loading was analyzed using electrical loading–unloading loops. The calculated hysteresis error (see

Supporting Information) decreases from 17.3% in the first cycle to 12.8% after 100 cycles, confirming the superior signal reversibility and cyclic stability (Figure S31b). These results demonstrate that, unlike the damping materials with viscoelastic dissipation, LWGM provides a reliable dynamic electrical readout under repeated loading.

Figure 4i shows the results of dynamic thermomechanical analysis (DMA) for LWGM-100, tested over a temperature range from -100°C to 300°C at 1 Hz. The storage modulus G' and loss modulus G'' of LWGM remain constant with changes in temperature and frequency, indicating negligible variation in mechanical properties with temperature (Figure S32). Even in ultralow temperature (liquid N_2 , -196°C), the LWGM-100 retains stable resilience (Figure S33 and Movie S8). Traditional sensors, such as mature conductive polymer composites, have clear advantages in low-cost manufacturing and industrial scalability, which LWGMs have not yet achieved. However, LWGMs exhibit a high detection range and broad operating tolerance, endowing them with significant potential for deployment in extreme environments where conventional sensors fail to function reliably.

3 | Conclusions

In summary, we successfully fabricated diverse graphene metamaterials via a multi-flow wet-assembly approach, including the cuttlebone-inspired lamella-wall metamaterial and other programmable architectures with varied shaping, alternating, and gradient geometries. Similar to natural cuttlebone, LWGMs exhibit a unique layer-by-layer collapse deformation mechanism. The vertical walls enable cooperative load-bearing and nearly complete recovery from compression, creating a bio-inspired elastic cuttlebone material. This distinctive architecture endows LWGMs with superior compressive strength, modulus, and structural stability compared to conventional low-density carbon materials. Compressive strength increases with the reduction of layer distance h . It's suggested that further reducing the layer distance would result in more uniform stress distribution and higher compressive strength, providing new insights for developing lightweight yet mechanically robust carbon materials. The piezoresistive behavior under high pressures and long cycles suggests promising use in pressure sensors that demand both mechanical stability and wide detection range. Incorporating magnetic or polymeric components into such graphene metamaterials could also create more unique performances and multifunctionality for broader applications.

4 | Experimental Section

Methods and any associated references are available in the **Supporting Information**.

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Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

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

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Supporting Information

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

Supporting File 1: adma73453-sup-0001-SuppMat.doc.

Supporting File 2: adma73453-sup-0002-MovieS1-S8.zip.