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Highlight

Graphene meta-aerogels: When sculpture aesthetic meets 1D/2D composite materials

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ABSTRACT

The engraving technique has grown in parallel with our human civilization, along with the targeted materials evolving from stone and metals to wood. Benefiting from the blossom of nanotechnology, the bulky nicking tools have downsized themselves to a micro-/nanoscale, such as laser beams, and the materials have been extended from traditional hard ones to soft functional nanomaterials. When ancient sculpture art meets modern advanced micro-/nano fabrication techniques and low-dimensional materials, impossible materials are born, which will redefine the functional scope of well-developed materials. Recently, a team from Tsinghua University reported such fascinating materials, graphene-based meta-aerogels, that process excellent elasticity, ultralight specific weight (down to 0.1 mg·cm^{-3}), and superwide Poisson's ratio range ($-0.95 < v_{peak} < 1.64$) via facile and fast laser-engraving technique.

KEYWORDS

graphene, aerogel, laser engraving, compressible

Lightweight structural materials that integrate superior conductivity, unusual mechanical properties, and outstanding fatigue resistance are alluring in a variety of fields, from aerospace to energy-related areas. However, traditional materials in large fail to meet all these criteria. The discovery of the first two-dimensional atomically thin materials, graphene, makes these totipotent materials possible. Since 2010, tremendous scientific endeavors have been devoted to assembling two-dimensional (2D) graphene building blocks into macroscope three-dimensional (3D) bulk materials with extraordinary conductivity and mechanical strength. As a direct result, compressible, stretchable, ultralight and robust graphene-based aerogels were reported [1–4]. Parallelly, innovative assembly and processing techniques, such as hydrothermal selfassembly, ice-templating approach, and 3D printing are implemented to shape the aerogels to a desirable architecture [5–7]. Despite these encouraging advances, graphene aerogel with adequate mechanical performance, such as the ultrawide range of reversible compressibility and stretchability, and tunable structure across multiple length scales, remains a daunting task.

From the viewpoint of building blocks, pristine 2D graphene nanosheets alone cannot afford sufficient mechanical support for 3D skeletons under large deformation. More elegant structures combined with the binary/ternary system seem a promising solution, for example, 1D/2D hybrid composite. From a processing aspect, freeze-casting with the benefits of ice templating and the assistance of customized mold could enable oriented microstructure and duplicate the shape of the original mold. However, it is difficult to build up a sophisticated macrostructure that is commonly required for a specific application scenario. In this context, 3D printing emerges as an alternative strategy to construct graphene hydrogel/aerogel with customizable structures, while it demands an ultrahigh concentration of raw materials to guarantee suitable rheology attributes. Besides the time and energy-cost processing procedure for highly concentrated graphene inks, another side effect is their high viscosity, leaving an open question to regulate inter structure at a micro-/nano level accurately.

In a new paper published in *Nature Communications*, Qu et al. reported an ingenious laser-engraving strategy towards graphene meta-aerogels (GmAs) with unprecedented performance via multiscale structural regulation [8]. Inspired by natural leaves, 1D polyimide nanofibers with rigid aromatic chains were screened to reinforce the 2D graphene sheets, resulting in a veins-on-blade structure (Fig. 1(a)). Such 1D reinforced 2D microstructure enables a higher elastic bending stiffness of walls in GmAs, leading to a conversion of walls' deformation from microscopic buckling to a bulk mode and bettering mechanical properties in terms of robustness, strength, stiffness and shape recovery capacity. Besides, by leveraging bi-directional freeze casting, a long-range ordered lamellar microstructure up to several centimeters is achieved, which

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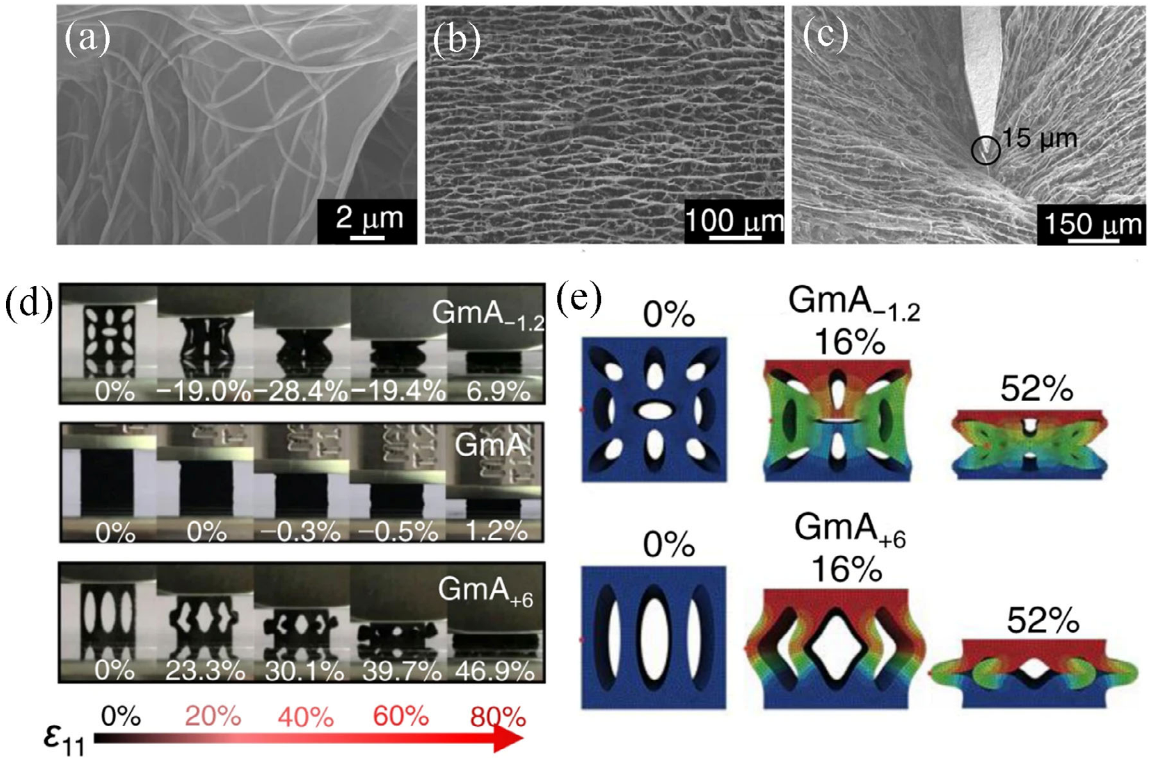

Figure 1 (a) and (b) cross-section SEM images of GmAs at different scale . (c) SEM image of GmA under blade cutting. The black circle marks the blade edge width of ~ 15 μm. (d) Snapshots of GmAs with different configurations during uniaxial compression. (e) The finite element calculation shows the compression process of samples of concave- ($GmA_{-1.2}$) and convex-shaped (GmA_{+6}) configurations.

can enhance mechanical performance (Fig. 1(b)). To note, the resultant GmAs deliver a remarkable fatigue resistance with 100% strain retention even after 1,000 cycles of 50% compression strain, surpassing most of the carbon-based aerogels. The GmAs can even withstand the cutting of a shape blade and fully recover their shape (Fig. 1(c)). Combined with structural characterization and molecular dynamic simulation results, the authors proposed a raised bending stiffness-induced bulk deformation mechanism. More interesting, further introducing laser engraving technique grants courtless meta-structures to the GmAs. With a delicate structure design, various Posison's ratios can be realized (Figs. 1(d) and 1(e)). It is worth highlighting that it is the first report regarding an ultrawide range of tunable Posison's ratio for graphene-based aerogel. Rich "graphene sculpture" combined with tailored Posison's ratio as well as excellent mechanical properties make the GmAs promising in functional and intelligent devices. Equally important, the authors demonstrate the versatility of GmAs by incorporating magnetic nanoparticles into the aerogel. Various magnetically responsive actuators are showcased with high shape fidelity. Even though the tremendous advances mentioned above, some challenges remain for this strategy. For instance, how to accurately control the inner structure and expedite the process of engraving when a large sample is met?

This paper achieves a milestone in graphene-based aerogel as it provides multiscale structural regulation and realizes multifaceted performance enhancement. Equally important, Qu and his coworkers open up a new strategy to shape soft, ultralight and conductive metamaterials, which will significantly gear up the development of 2D materials-based aerogels beyond graphene.

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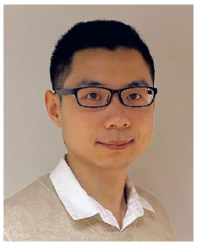

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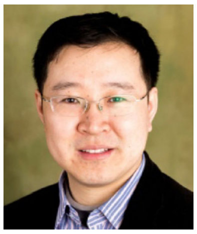

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