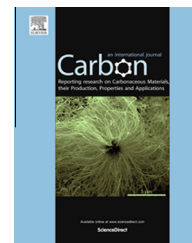


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# Elastic improvement of carbon nanotube sponges by depositing amorphous carbon coating

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## ABSTRACT

Porous materials such as foams, sponges and aerogels typically exhibit large energy dissipation during deformation, resulting in structural weakening and incomplete recovery. Here, we adopted a vapor deposition method to introduce a uniform amorphous carbon (AC) coating with controlled thickness throughout the porous network, on the surface of individual carbon nanotubes (CNTs) as well as their junctions, forming AC-CNT sponges. The AC-CNT sponges show enhanced mechanical properties (compressive strength, modulus), and in particular, significantly narrowed stress–strain loops with small energy loss coefficients compared with previous CNT or graphene-based aerogels typically having large hysteresis. Mechanism study has revealed several important factors that contribute to the observed superelasticity, including the thickening of nanotube struts, welding of nanotube junctions, and inhibition of bundle formation. Our method could systematically tailor the structure and properties of porous CNT (or other nanostructure) sponges, which have applications in microelectromechanical systems and mechanical energy storage.

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## 1. Introduction

Porous materials are widely used in structural, environmental and energy-related fields [1–3]. Carbon nanotubes (CNTs) and graphene sheets are strong and flexible nanostructures, and have been constructed into various porous structures such as foams, sponges, rubbers and aerogels [4–12]. The combination of excellent mechanical properties and electrical conductivities in CNT and graphene three-dimensional networks

enables numerous potential applications as scaffolds for functional composites, high performance electrodes for batteries and supercapacitors, and flexible devices that can work under deformation [13–20]. Compared with traditional materials such as polymeric foams and inorganic aerogels [21,22], macroscopic structures made from CNTs and graphene represent a new generation porous materials with ultra-light weight, high flexibility, significantly improved properties and multifunctionality.

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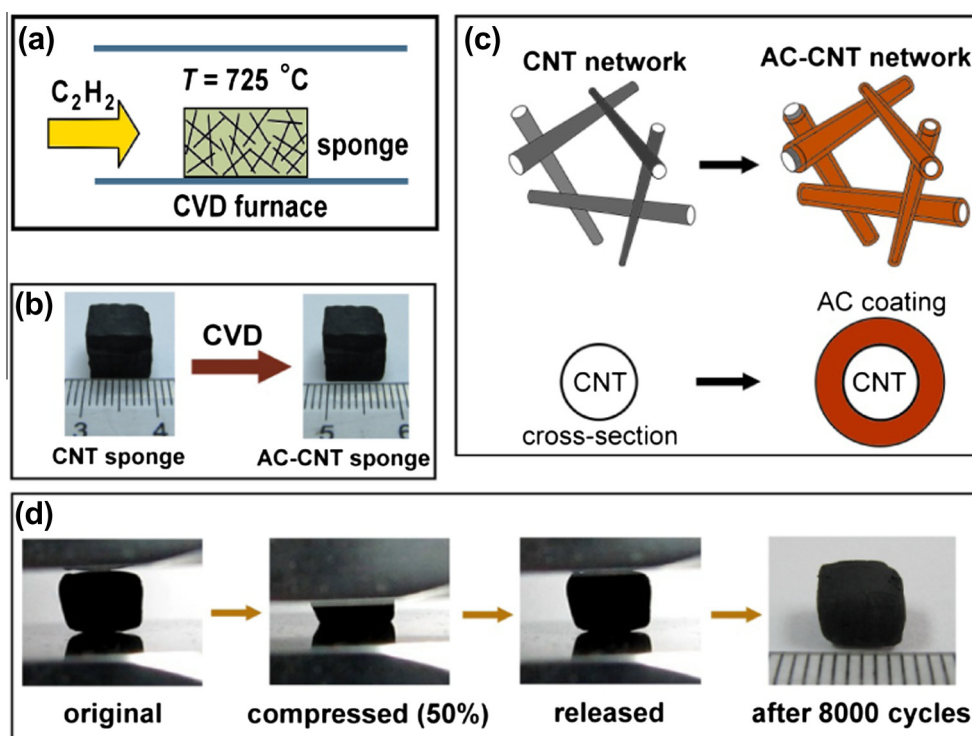
Although CNT and graphene-based arrays and networks can be compressed to large strains without collapse, associated energy dissipation processes (internal friction, zipping and un-zipping effect, buckling of nanotubes or graphene walls) cause gradual structural weakening and plastic deformation during cyclic compression [4,7,11,12]. Significant energy dissipation is reflected in the stress–strain curves with large hysteresis, especially at large strains, similar to the behavior of typical cellulose and biological polymers [23–25]. The compressive strength and modulus also degrade with increasing cycles, indicating structural weakening due to damage of nanostructure units and their interconnection. Intensive efforts have been focused on improving related mechanical properties, for example, a superelastic single-walled nanotube aerogel has been produced by coating few-layer graphene nanoplates through polymer infiltration and a two-step pyrolysis treatment, which is a relatively complex method [26]. For graphene aerogels, microwave treatment or creating CNT-coated graphene walls by synergistic assembly have enhanced the elasticity and structure recovery under repeated compression [11,12]. However, most of CNT and graphene porous structures show large hysteresis loops (with energy loss coefficients up to 75%) [4,11,12,19]. Given the high surface area and close packing of CNTs and graphene sheets, interaction between these structural units (e.g., relative motion, slipping) would cause friction and zipping effect (when two flexible nanotubes come into contact), resulting in considerable energy dissipation.

Here, we present a highly elastic and low energy-loss CNT sponge by introducing a conformal coating of amorphous carbon (AC) on individual nanotubes as well as inter-tube junctions to tailor the nanotube diameter and reinforce the network. Related mechanical properties such as compressive strength, modulus and the structure–property relationship have been studied systematically. Distinct from previous CNT or graphene aerogels, our AC-reinforced superelastic CNT sponges show very small stress–strain loops under compression characterized by low energy loss coefficients (<20%) at small to large strains over many cycles. The revealed mechanisms including the thickening of nanotube struts, welding of nanotube junctions and preventing bundle formation, provide useful insights in future design and fabrication of superelastic porous materials for many applications.

## 2. Experimental

### 2.1. Synthesis of CNT sponges with controlled bulk densities

CNT sponges were synthesized by chemical vapor deposition (CVD) using ferrocene and 1,2-dichlorobenzene as the catalyst and carbon precursors. A solution of ferrocene in dichlorobenzene (0.06 g/mL) was injected into the CVD furnace by a syringe pump at a feeding rate in the range of 0.1–0.25 mL/min, and carried into the reaction zone by Ar/H<sub>2</sub> (2000 and 300 mL/min, respectively) to grow sponges. The reaction temperature was set as 860 °C, and the CVD time was 4 h.



**Fig. 1** – Fabrication of elastic AC-CNT sponges. (a) Experimental setup including a CNT sponge placed in the CVD furnace (reaction temperature = 725 °C) and C<sub>2</sub>H<sub>2</sub> as carbon precursor. (b) Photos of a CNT sponge before and after CVD. (c) Illustration of the network structure in the CNT and AC-CNT sponges. (d) Snapshots taken during cyclic compression on an AC-CNT sponge (to 50% strain) and after 8000 cycles without plastic deformation. (A color version of this figure can be viewed online.)

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