Ultralight and Highly Compressible Nitrogen-doped Magnetic Carbon Nanospheres/Graphene Composite Aerogel

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Introduction

Ultra-light compressible graphene aerogel has extensive application prospects in many fields.[1-2] In this study, graphene oxide nanosheet (GO) was used as precursor and weakly alkaline ammonium citrate was used as reducing agent and nitrogen source. Then, by introducing the positively charged magnetic carbon nanospheres, nitrogen-doped magnetic carbon nanospheres/graphene composite aerogels (MCNS/NGA) were first obtained by a one-step hydrothermal electrostatic self-assembly strategy. The MCNS/NGA exhibited ultra-low density, superelasticity and good Magnetism. More importantly, this work provides a practical method for the synthesis of 3D graphene-based composites.

Materials and Methods

Graphite powder (325 mesh) was purchased from Aladdin Reagent Company. Ammonium citrate was obtained from Tianjin Guangfu Technology Development Co., Ltd. All chemicals were used without further purification. GO was prepared from graphite powders by a modified Hummers method.[3] MCNSs were originally fabricated using ferrocene, hydrogen peroxide, 1,5-dihydroxynaphthalene and PF127 by one-step hydrothermal method. MCNS/NGA were prepared using GO, ammonium citrate and MCNSs by a one-step hydrothermal in-situ electrostatic self-assembly strategy.

Results and Discussion

The cylinder-shaped monolith of MCNS/NGA with an ultra-low density of 12.07 mg cm^{-3} can freely stand on dandelion (**Fig. 1a**). MCNS/NGA have a 3D interconnected microstructure with numerous open pores (**Fig. 1b**), which can effectively inhibit the aggregation of graphene sheets. Highly wrinkling regions and uniformly dispersed magnetic carbon nanospheres (**Fig. 1c**) can be observed on the surface of the graphene. Meanwhile, the selected area electron diffraction pattern shows diffusion rings, indicating MCNS/NGA have good crystallinity. In the FTIR spectra (**Fig. 1d**), the $-NH_2$ and -CN stretching vibration bands exist at 1588 and 1250 cm^{-1}, respectively. In addition, the characteristic band at 582 cm^{-1} is attributed to Fe-O of Fe_3O_4. From magnetic performance test (**Fig. 1e**) the magnetic saturation strength (Ms) of MCNS/NGA is 22.47 emu g^{-1}, indicating good superparamagnetism. The mechanical property test (**Fig. 1f**) shows that the

MCNS/NGA can undergo reversible deformation under different strains, i.e., 30%, 60%, 90% and 95%, respectively, exhibiting excellent resilience. With the strain of 95%, the maximum stress is up to 138.79 kPa, superior to most reported materials to-date.

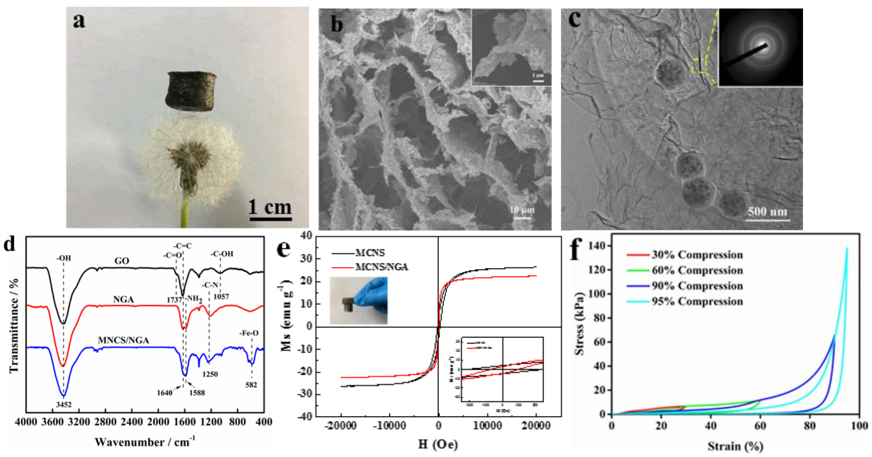

Figure 1 a) Photograph of a MCNS/NGA standing on dandelion; b) SEM and c) TEM images of MCNS/NGA; d) The FTIR spectra of GO, NGA and MCNS/NGA; e) Magnetic hysteresis loop of MCNS and MCNS/NGA; f) The stress-strain curves of MCNS/NGA at different strain of 30%, 60%, 90% and 95%.

Conclusions

In conclusion, the graphene composite aerogel in this work can effectively overcome the accumulation and irreversible agglomeration of graphene nanosheets, and improve the pore structure and mechanical properties. It has an ultra-light, stable and repeatable superelasticity (up to 95%, maximum stress up to 138.79 kPa), and good magnetism (magnetic saturation strength of 22.47 emu g^{-1}). Therefore, the MCNS/NGA offer attractive prospects extending to the applications as adsorbents, supporter for sensors, and electrode materials for supercapacitors.

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