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High-quality graphene films produced by freeze-stretching alignment

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Monolayer graphene has the highest tensile strength, Young's modulus, and electrical conductivity of known materials, and it is expected to be used in aerospace, automobiles, flexible electronics, etc. [1]. However, the extraordinary properties of graphene are on the molecular level and have not been realized when assembled into macroscopic materials, thereby hindering its practical applications. There exist two pivotal scientific issues in the assembly of graphene: (1) the misaligned and wrinkled structure of graphene platelets and (2) weak interfacial interactions among graphene platelets [2], which have not yet been adequately addressed. The fabrication of high-performance graphene macroscopic assemblies such as films and fibers is therefore a great challenge and important goal.

In a recent article published in *Nature Materials* [3] by Prof. Qunfeng Cheng's group from Beihang University in China and collaborators from the University of Texas at Dallas led by Prof. Ray H. Baughman, this challenge has been effectively addressed by a strategy of sequential covalent and π-π inter-platelet bridging under stretching, as shown in Fig. 1a. This joint research team demonstrated that sequential bridging is essential to permanently freeze the stretching-induced alignment, leading to simultaneous improvement in alignment, stacking compactness, and interlayer connectivity of the graphene. The resulting reduced graphene oxide (rGO) films have a record high isotropic in-plane tensile strength (1.55 GPa, Fig. 1b), and a high Young's modulus, toughness, electrical conductivity, and electromagnetic interference (EMI) shielding capacity.

Cheng and co-workers [3] used a focus ion beam (FIB) to cut and tailor the rGO films and observed that there are numerous voids between the wrinkled graphene platelets, which, together with poor alignment, results in the poor properties of the films. They also found that any stretching-induced alignment decreases when the stretching force is removed unless the graphene platelets are bridged during stretching. Considering the two dimensionality of graphene platelets, biaxial stretching can improve the isotropic in-plane alignment of the films, and this may be conveniently adjusted by changing the stretching force. Because of the increased alignment, the tensile strength, Young's modulus and electrical conductivity of the films gradually increase while their plastic deformation gradually decreases. Specifically, the tensile strength, Young's modulus, toughness, electrical conductivity, and EMI shielding effectiveness (SE) of the optimized sequential bridged (SB) and biaxially stretched (BS) rGO (SB-BS-rGO) films are 3.6, 10.6, 3.3, 1.5, and 1.5 times those of untreated rGO films (Fig. 1c), respectively. Moreover, the strength and toughness of these films are superior to those of in-plane carbon fiber fabric composites that are currently used in a wide range of commercial products.

More importantly, Cheng and co-workers [3] demonstrated that the rGO films can be easily bonded together by using a commercial resin without significantly decreasing their performance, which provides a method for the fabrication of large-area, thick high-performance graphene films. By combining doctor blade casting and this novel strategy of sequential bridging under stretching, they reported the fast fabrication of large-area, thick graphene films, whose properties are similar to those of small, thin films fabricated by vacuum filtration, indicating the great potential of this process for large-scale commercial applications. This work may also suggest a method for fabricating the high-performance assembled materials of other two-dimensional nanoplatelets.

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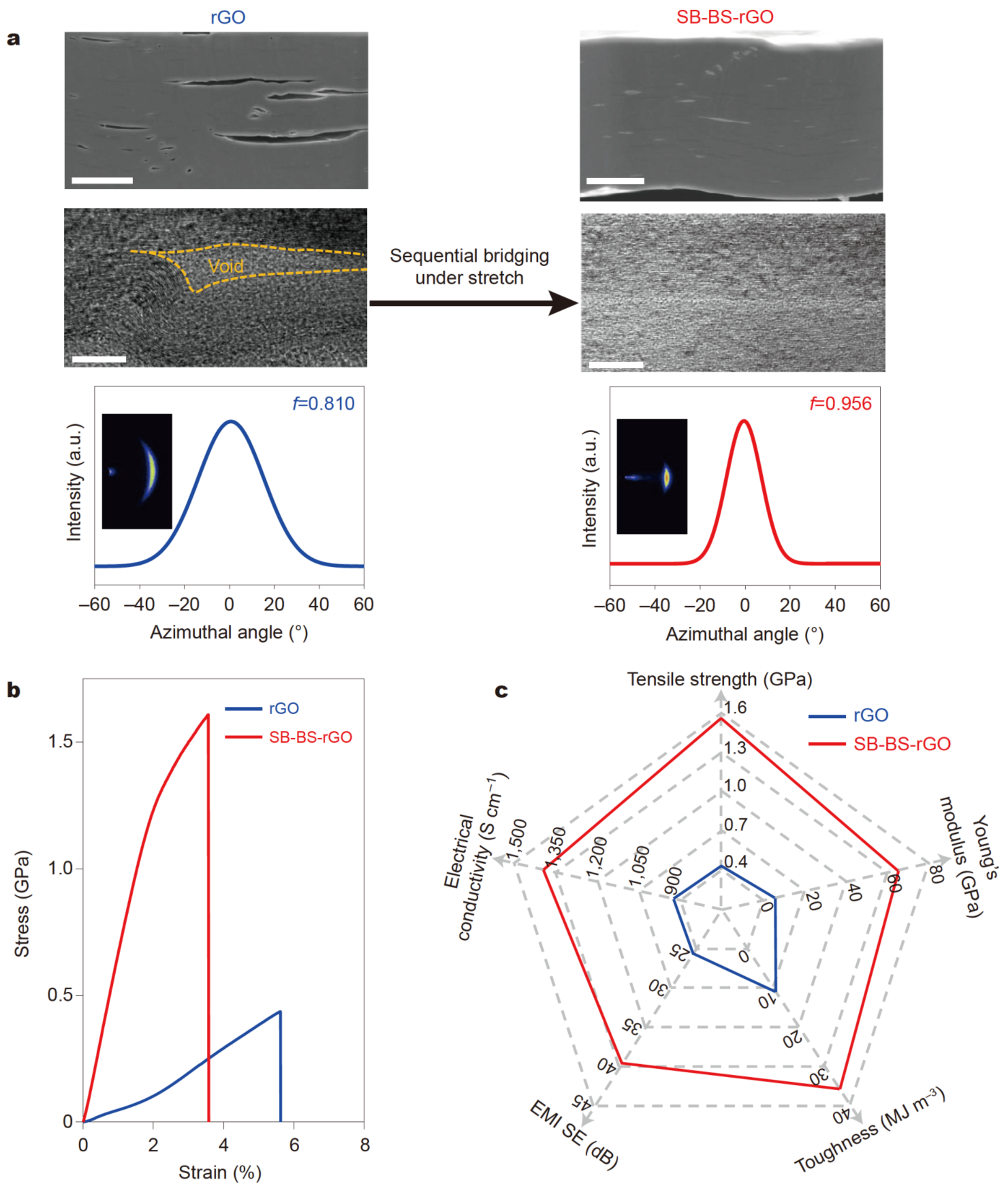

Figure 1 (a) Scanning electron microscopy images (top: scale bar 1 μm) and transmission electron microscopy images (middle: scale bar 10 nm) of cross-sections cut by FIB, and the corresponding wide-angle X-ray scattering patterns (bottom) for rGO (left) and SB-BS-rGO (right) films. (b) Typical tensile stress-strain curves of rGO and SB-BS-rGO films. (c) A radial plot comparing the tensile strengths, Young's moduli, toughnesses, electrical conductivities, and EMI SEs of rGO and SB-BS-rGO films.

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