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Bioinspired hierarchical graphene oxide wrinkles for flexible sensors, actuators and manipulators

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Recently, flexible and wearable devices are increasingly in demand and graphene has been widely used due to its exceptional chemical, mechanical and electrical properties. Building complex buckling patterns of graphene is an essential strategy to increase its flexible and stretchable properties. Inspired by the gyrification in the human brain, and the hierarchical micropapillae on the superhydrophobic surface of natural rose petals, herein we introduce a simple three-dimensional (3D) shrinking method to generate the cortex-like patterns using two-dimensional (2D) graphene oxide (GO) as the building blocks. And a facile dimensionally controlled four-dimensional (4D) shrinking method was further proposed to generate hierarchical buckling patterns on cylindrical substrates. The fluoroalkylsilane modified GO papillae array exhibits a combined performance of strong superhydrophobicity ($CA > 170^\circ$), tunable adhesive force (39.2~129.4 μN) and ultralarge liquid capacity (25 μL), which are promising for programmable manipulations of microdroplets and potential for multi-step microreactions. In response to some organic solvents, the square bilayer actuator exhibits excellent reversible, bidirectional, largedeformational curling properties on wetting and drying, which are promising for bionic actuators like mimosa. The reduced graphene oxide ridges (rGORs) generated on the spherical substrate seem isotropic, while those generated on the cylindrical substrate are obviously more hierarchical or oriented, especially when the cylindrical substrate are shrinking *via* two steps. The flexible rGORs-based strain sensors can be used to detect both large and subtle human motions and activities by achieving high sensitivity (maximum gauge factor upto 48), high unidirectional stretchability (300–530%), and ultrahigh areal stretchability (up to 2690%).

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