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Supporting Information

Three-dimensional Multi-recognition Flexible Wearable Sensor via Graphene Aerogel Printing

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Experimental Section

Preparation of graphene oxide

Graphene oxide (GO) was obtained by oxidizing expandable graphite (EG). 3g EG power was mixed with 40ml concentrated sulfuric acid (98%, H_2SO_4), 2.5g phosphorus pentoxide (P_2O_5) and 2.5g potassium permanganate ($K_2S_2O_8$), the mixture was kept at 80℃ for 5 hours. After cooling to room temperature, the mixture was diluted with ice water and filtering. Dried at room temperature for 2 days, obtained the pre-oxidized EG. Then the pre-oxidized EG was added into 120ml H_2SO_4, 9g $KMnO_4$ was added slowly with stirring. Then the mixture was heated to 35℃ and stirred. 2 hours later, diluted with 1L water, and added dropwise 5ml 30% H_2O_2 until the mixture color become bright yellow. The mixture was wash three times with 10% hydrochloric acid and centrifuge. Finally, load in the dialysis bag and soaked in deionized water until pH=7.

Ink preparation

The synthesized GO dispersion was concentrated by solvent evaporation method with a 60 °C water bath. Then the pastes were gentle stirring to prepare inks with concentrations of 5, 10, 15, 20 and 25 mg/ml, respectively.

Micro extrusion printing of GO ink

The patterns were printed using a multiaxis dispensing system (2400, EFD). The GO inks (the concentrations from 5 to 25 mg/ml) were loaded in a syringe (3cm^3 barrel, EFD Inc.) attached by a luer-lok to a micronozzle (200 μm inner diameter). Extrude the ink through the nozzle via an air-powered fluid dispenser (Ultimus I, EFD, Inc.) which used to provide appropriate pressure. The printed patterns were put in a frozen atmosphere for 12 h and then vacuum freezing-dried at -60 °C for 3 h.

Chemical reduction of GO patterns

The printed GO patterns were placed in a sample bottle with HI (15%) aqueous solution. Then the bottle was put in a drying oven and heated to 95 °C and kept there for 3 h. After the sample cooled to room temperature, the patterns were washed by water and vacuum-dried at -60 °C for another 3 h. Then, successfully obtained reduced graphene oxide (rGO).

Characterization

The surface morphology and structure were characterized by field emission scanning electron microscopy (FE-SEM, Hitachi S4800), atomic force microscope(AFM, Fastscan Bio) and X-ray diffraction (Empyrean diffractometer using monochromatic Cu Kα1 radiation (λ = 1.5406 Å) at 40 kV). The resistances of printed patterns were characterized by four-probe method (Keithley 4200

Semiconductor Characterization System) and UNI-T analyzer (UT39A).

Calculation of the porosities of the aerogels

Calculation of the porosity according to the following equation: [1, 2]

$$\varepsilon = 1 - \rho / \rho_0$$

Where ε denotes the porosity, ρ is the density of fabricated graphene aerogel, and ρ_0 is the density of graphite, which hypothesized to be 2200mg/ml.

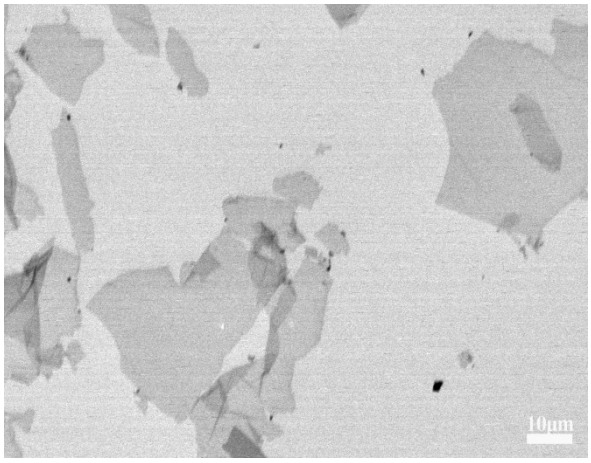

Fig. S1 SEM image of GO, the lateral size of GO sheets distributes from 50 μm to 80 μm.

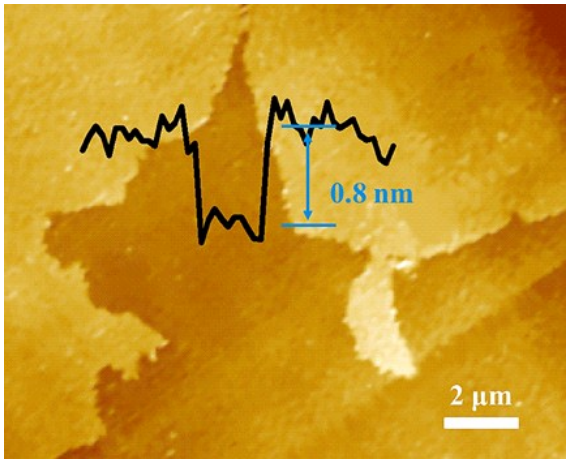

Fig. S2 AFM image of GO, the representative thickness is characterized as 0.8 nm.

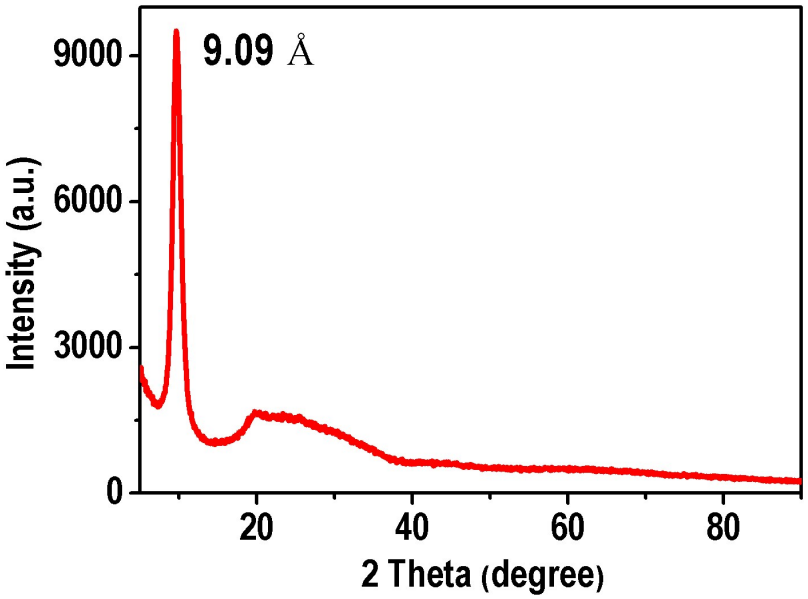

Fig. S3 XRD image of GO, the diffraction peak at 9.72° corresponds to the characteristic 001 peak of GO, the *d* spacing of 9.09 Å indicates the dense stacking of the GO sheets. Besides, a weak diffraction peak at 20.32° is close to the 002 peak located in 26.4° from natural graphite.

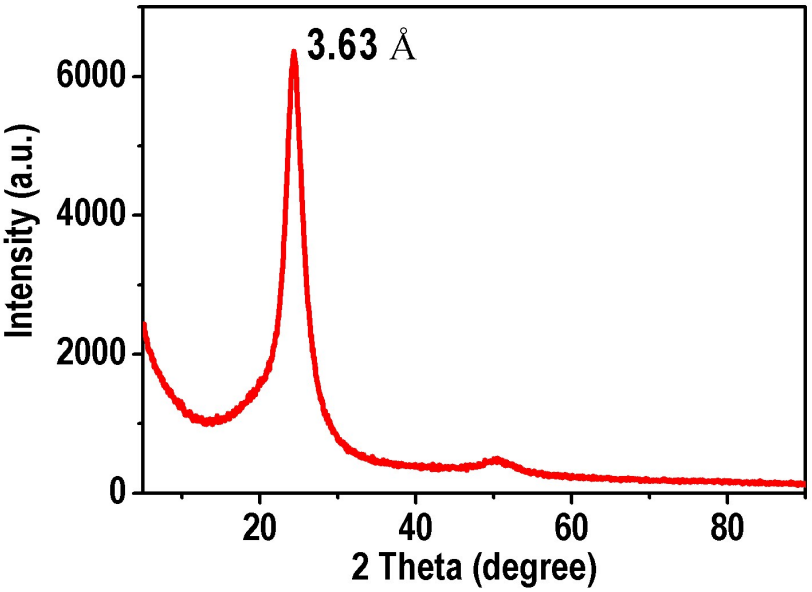

Fig.S4 XRD image of rGO, the XRD pattern of rGO displays a wide peak at 24.52°, with a *d* spacing of 3.63 Å. The interlamellar spacing of rGO is much more narrow than that of GO and quite close to that of natural graphite. This is due to partial removal of the oxygen-containing groups in the reduction process.

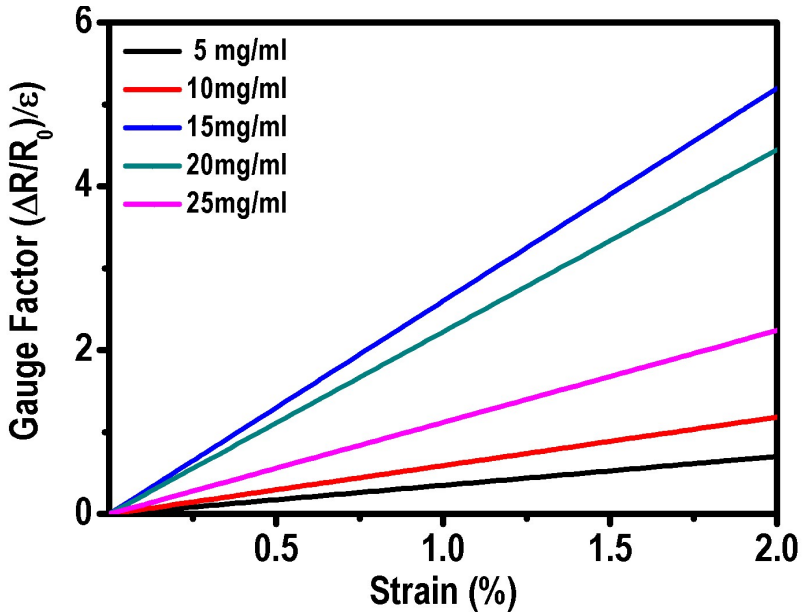

Fig.S5 Gauge factors under different strains for the printed graphene aerogel with different concentrations (from 5mg/ml to 25mg/ml).

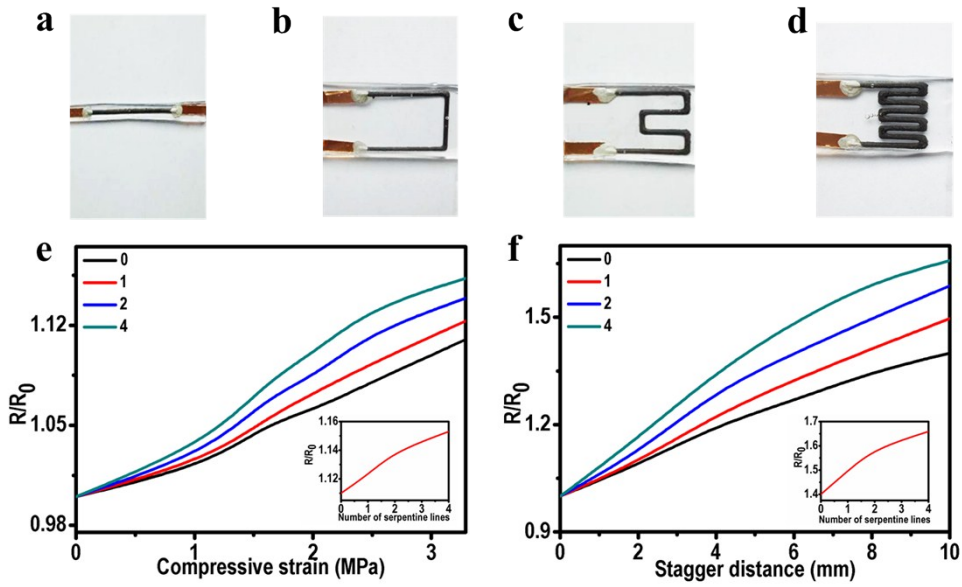

Fig. S6 Photographs showing (a) straight line (b) one serpentine line (c) two serpentine lines (d) four serpentine lines printed graphene aerogel sensors. (e) Compression test (f) Staggering test with resistivity changes curve. (inset: curve trend chart)

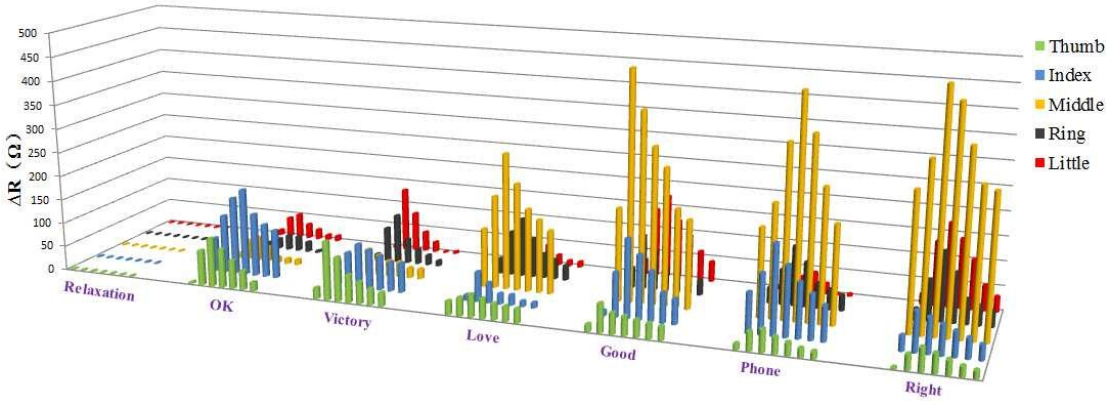

Fig. S7 The 3D chart displays resistance changes of 7 different gestures which were monitored at 5 figures joint with electronics data logger. Seven characteristic vales were readout at the peak or nadir of the monitoring resistance curves. There were 35 variables for each gesture to descript to the detail movement responses.

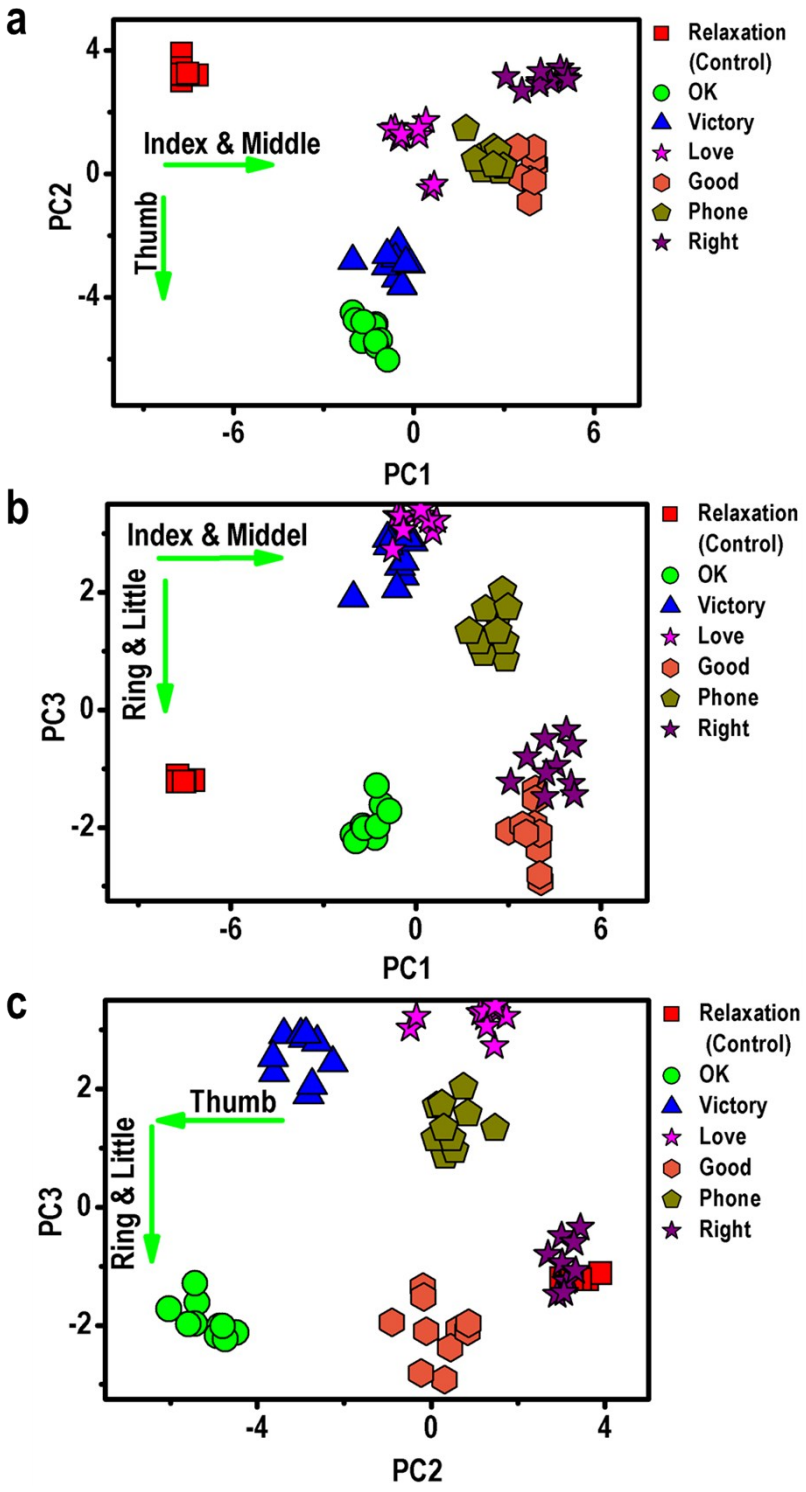

Fig. S8 PCA score plots of the first three principal components of statistical significance for 7 different gestures. (a), (b), (c) represent the PCA result at different projection respectively.

References

1. H. Hu, Z. B. Zhao, W. B. Wan, Y. Gogotsi, and J. H. Qiu, *Adv. Mater.*, 2013, **25**, 2220.
2. Y. Liu, W. Z. Qian, Q. Zhang, A. Y. Cao, Z. F. Li, W. P. Zhou, Y. Ma, and F. Wei, *Nano Lett.*, 2008, **8**, 1324.