Supporting information for

Highly sensitive pressure switch sensor and enhanced ultraviolet photodetector based on 3D hybrid film of graphene sheets decorated with silver nanoparticles

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Table S1. Detailed sample information for as-purchased graphene oxide (GO).

Graphene Oxide	
Synthesis Method	Hummers
Diameter	500nm-5μm
Thickness	0.8-1.2nm
Single layer ratio	~99%
Purity	>99wt%

The information of the as-purchased graphene oxide (GO) was acquired from the Nanjing XFNANO Materials Tech Co. ,Ltd.

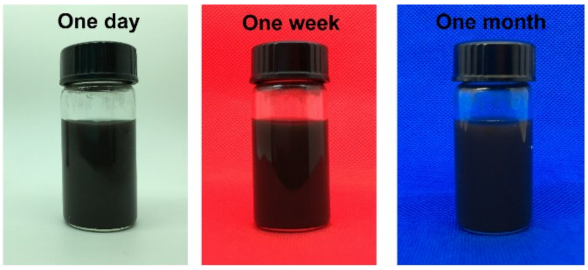

Figure S1. Photo images of the GNs/Ag colloid in different periods.

Photo images in different periods were shown in Fig. S1. The dispersion solution was stabilized by electrostatic repulsion, the GNs/Ag solution has no layering for a long time. The result confirm that the Ag NPs act as a useful spacer to prevent aggregation of GNs to form a stable solution.

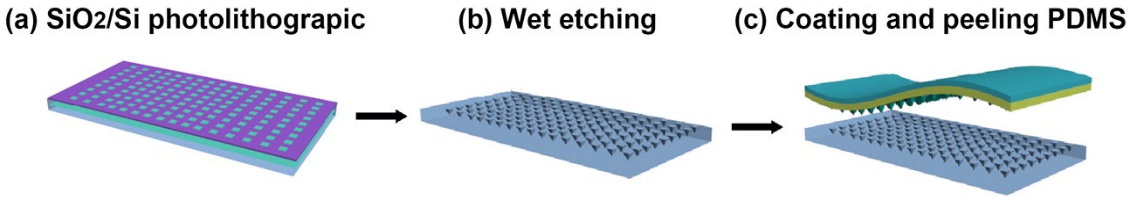

Figure S2. The flowchart schematic of fabricating pyramid arrays structured PDMS substrates.

The pyramid microstructured PDMS was prepared to disperse the external pressure and ensure the sensitivity and stability of the pressure switch device. The fabrication process of the sensitive pressure switch sensor is schematically illustrated in Fig. S2. Firstly, 10μm × 10μm SiO_2/Si arrays were obtained by lithography technique, and then the SiO_2 was etched by BOE (HF:NH_4F=6:1) and the arrayed Si was exposed to the air. The remaining SiO_2 act as the mask to etch the (001) Si arrays by the KOH and isopropyl alcohol mixed solution. Due to the anisotropic relative etching rates of Si {100} and Si {110} planes, the slowest etching plane aligned to the mask edge will form a sidewall. Based on this theory, the inverse square pyramid arrays structured Si wafer mould was obtained. (The specific etching parameters were listed in supporting information, Table S2) PDMS (20:1) dilute solution was coated on the as-prepared Si mould and vacuum-degassed for 30 minutes to fill up the entire substrate. After curing at 150 °C for 30 minutes, uniform size pyramid arrays structured PDMS was peeled off the Si mould. Similarly, PDMS (10:1) and PDMS (20:1) dilute solution were coated on normal flat Si substrates to obtain flat PDMS substrates, respectively.

Table S2. Specific wet-ecting parameters of Si wafer mould with inverse square pyramid arrays structure.

Step	Chemicals / Equipment	Temperature	Time
1	BOE ($HF:NH_4F$=6:1)	27 °C	30 minutes
2	Heater table	120 °C	2 minutes
3	BOE ($HF:NH_4F$=6:1)	27 °C	3 minutes
4	Acetone	27 °C	5 minutes
5	Deionized water	27 °C	5 minutes
6	KOH (20g) Deionized water(40mL) Isopropyl alcohol (50mL)	75 °C	15 minutes
7	Deionized water	27 °C	30 minutes

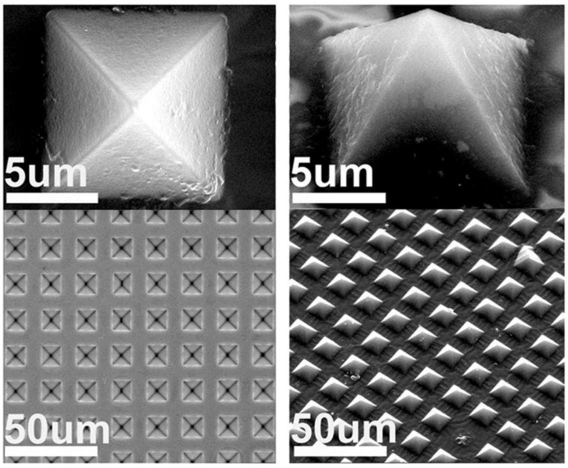

Figure S3. Top and side views of large-scope FESEM image of pyramid structural PDMS.

Top and side views of structured PDMS are show in Figure S3. Pyramidal PDMS (20:1) structure with sidewalls aligned to the mask edge was well produced.

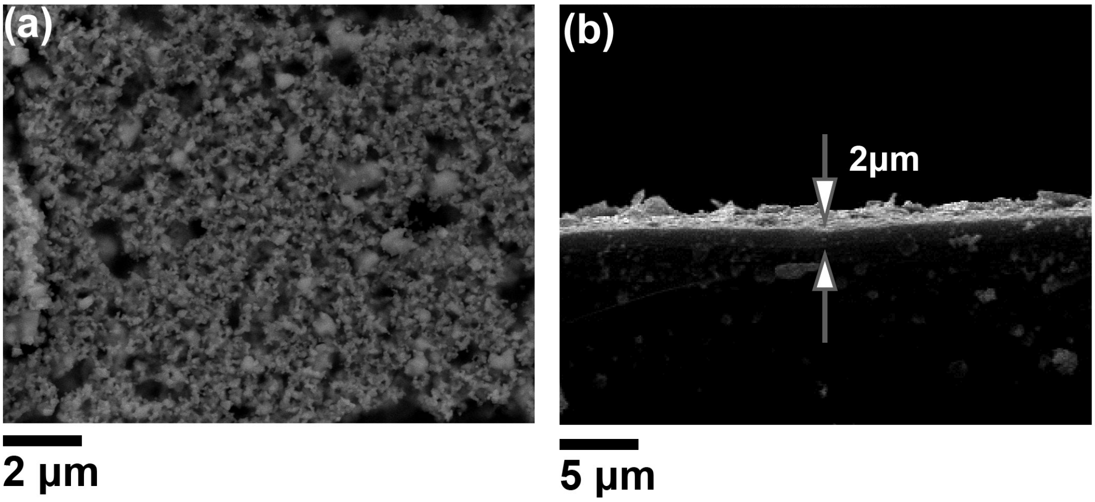

Figure S4. Top and cross-sectional SEM images of pressure switch sensor. (a) Surface morphology of pressure switch sensor. (b) Cross-sectional thickness images of the device.

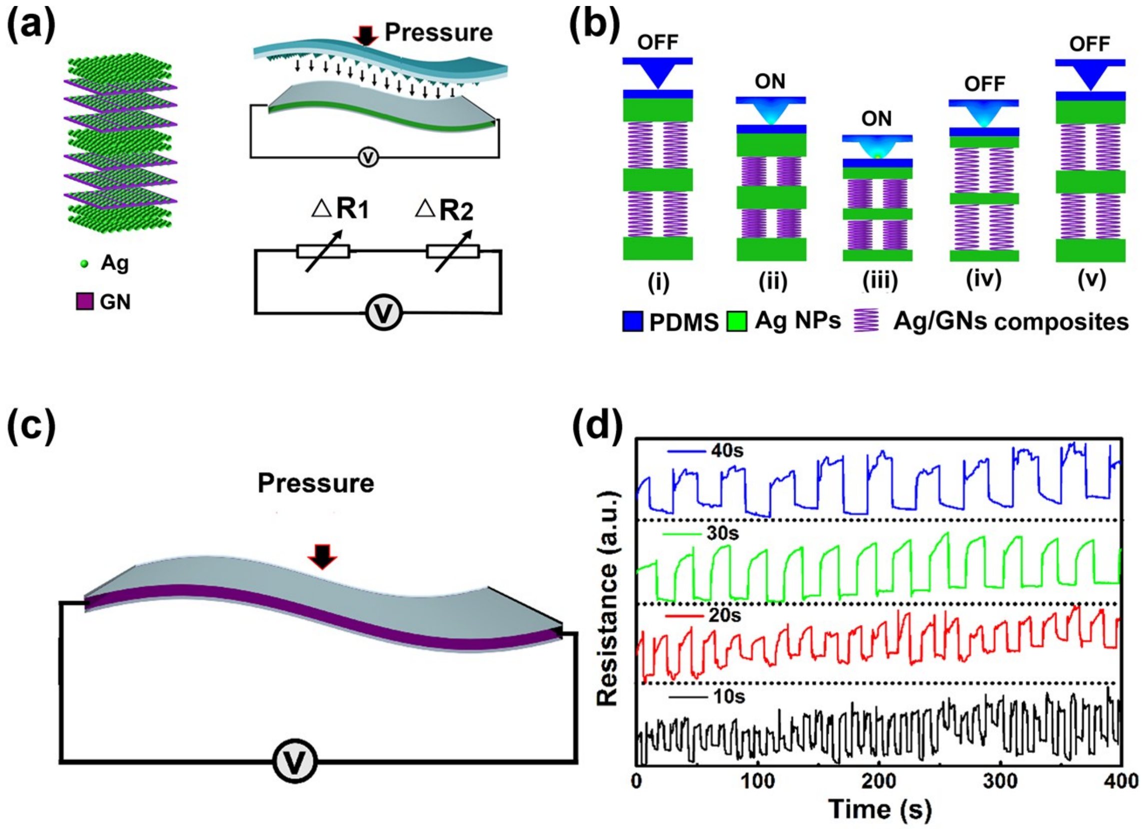

Figure S5. (a) An equivalent circuit for the contact resistance as a two-resistor network. (b) Schematic models for the working principle of the pressure switch sensors. (c) The model of unstructured pressure switch sensor. (d) Characterization of pressure response of unstructured devices under different periods.

As shown in Figure S5a, the resistance change of Ag NPs (the green region) was defined as ΔR_1, and the interface resistance change between Ag NPs and GNs (purple helical region) was defined as ΔR_2. The response to pressure can be attributed to the change of the contact resistance. The external pressure will be dispersed and evenly applied on the 3D Ag/GNs composites due to the pyramid structure. The Ag/GNs composites will act as a spring since the Ag NPs is an excellent nanospacer. With the external pressure applied, the alternate-superimposed 3D microstructure will be compressed, which provides more chance for Ag NPs touching with each other, resulting in the decrease of resistance. Conversely, spatial distance and the contact state will recover to the original state when the pressure is removed. When the external pressure is under the threshold, compression deformation of the Ag NPs (green region) and pyramid structured PDMS (blue region) can be ignored. The ΔR_1 can be considered as a constant value, and the response cycle can be divided into three circular steps: the original state (Figure S5b (i)); external pressure applied state (Figure S5b (ii)) and recovery state (Figure S5b (v)). With the pressure applied, the Ag NPs that inserted into the GNs contact instantly and form a loop with a small ΔR_2. The interface contact resistance will recover to the original value when the pressure is removed. When the pressure reaches the threshold, the response cycle will undergo all the steps (Figure S5b (i) to Figure S5b (v)). The ΔR_2 change rapidly between “ON” and “OFF”. However, the pressure, up to a certain degree, will lead to further compression of the device and reduce the resistance of Ag NPs (ΔR_1). On the other hand, compression deformation of pyramid structured PDMS (20:1) should be also considered. Both of them will retard the recovery and the combined effects can be considered to explain the phenomenon that ramps appear in the response process. Because the change of interface contact resistance (ΔR_2) play the main role in the pressure response cycle, the pressure switch sensor would operate normally.

The model of unstructured pressure switch sensor was shown in Figure S5c. Characterization of pressure response of unstructured devices under different periods was shown in Figure S5d. Though the device without pyramid PDMS structure also shows immediate response to the applied pressure, however, the device with structured

PDMS exhibits stable resistance response, whereas the response of the unstructured device is unstable. The results indicate that the structured device is more suitable for pressure switch sensing due to desirable switching between “ON” and “OFF”.