Supplementary Information

## High-Performance Transparent Pressure Sensor Based on Sea-Urchin Shaped Metal Nanoparticles and Polyurethane Microdome Arrays for

## **Real-Time Monitoring**

Donghwa Lee, Jongyoun Kim, Honggi Kim, Hyojung Heo, Kyutae Park, and Youngu Lee\*

Department of Energy Science & Engineering, Daegu Gyeongbuk Institute of Science and Technology (DGIST), 333, Techno Jungang-Daero, Hyeonpung-Myeon, Dalseong-Gun, Daegu, 42988, Korea

\*Corresponding Author, E-mail: youngulee@dgist.ac.kr



**Figure S1**. Atomic force microscopy (AFM) topographies of sapphire wafer as a mater mold: (a) 2D image, (b) phase image, and (c) 3D image. It shows that microdome arrays were characterized by a height of ~1.6  $\mu$ m, diameter of ~2.7  $\mu$ m, and pitch of ~3  $\mu$ m.



**Figure S2.** Photographs of nanoimprinting process: (a) imprinting process and (b) separation of mater mold and replicated product. An intermediate polymer substrate (IPS) template with membrane shield is pressed like a stamp onto the microdomes applied to the wafer surface. The heat is used to form the microdome patterns, after which the IPS template is separated from the master mold. A replicated product, IPS replica, appears the transparent rainbow pattern. (c) Top-view and magnified scanning electron microscopy (SEM) images of IPS replica mold (characterized by a diameter of ~2.64  $\mu$ m, and pitch of ~3  $\mu$ m.).



**Figure S3.** Photographs of demolding process that an IPS replica mold was easily demolded from patterned SSNPS-polyurethane (PU) layer because of the intrinsic properties of IPS template. After attaching another indium tin oxide (ITO) film deposited on a transparent polyethylene terephthalate (PET) substrate, the pressure sensor composed of PET/ITO/microdomes SSNPs-PU/ITO/PET shows high transparency.



**Figure S4.** Cross-sectional SEM image of the microdomes (characterized by a height of ~1.6  $\mu$ m, a diameter of ~2.6  $\mu$ m, and a pitch of ~3  $\mu$ m).



Figure S5. SEM image of microdome SSNPs-PU film. All samples tend to have similar thickness (approximately  $60 \ \mu m$ ).



**Figure S6.** Optical transmittance of pressure sensors with microdome arrays fabricated using different concentrations of SSNPs ( $0.6 \sim 6.0 \text{ mg/mL}$ ).



**Figure S7**. Illustration of a computer controlled custom-made measurement system composed of a force gauge with motorized test stand, a digital source meter for I-V measurement, various standard weights, and custom jig connected copper wires.



**Figure S8.** Photographs of fabrication process using a conventional poly(dimethylsiloxane) (PDMS) mold: (a) removal of air bubble in PDMS mixture, (b) curing of PDMS with master mold, and (c) demolding of replica PDMS from a master mold. (d) SEM image of a replica PDMS. It shows the uniformity and integrity of microholes without structural defects.



**Figure S9.** SEM images of a (a) master mold, (b) replica IPS mold, (c) SSNPs-PU layer, and (d-f) their magnified patterns. It is obvious that the IPS molding process ensures the uniformity and integrity of microdome arrays of a SSNPs-PU composite without additional processes such as surface treatment unlike the PDMS mold.



**Figure S10.** (a) Illustration of the conduction mechanism of pressure sensor based on microdome compared with planar structure. (b) Resistance chance of pressure sensor based on planar and microdome structures under different pressure. The increase of contact area leads to a reduction in resistance by amplifying the possibility of tunneling. Note that in comparison with tunneling resistance, the effect of the SSNPs resistance and contact resistance is found to be quite low and therefore it is neglected.



**Figure S11**. A plot showing the sensitivity (*S*) defined with the same equation (defined as  $S = (\Delta R/R_0)/\Delta P$ ) and the detection limit of reported piezoresistive pressure sensors. Note that the reported piezoresistive pressure sensors [ref. 1 to 9] are all opaque due to the high concentration of conductive fillers. Detailed values of the sensitivity and detection limit are described in Table S1.

Materials	Transmittance	S function	Sensitivity	Detection limit	ref
PEDOT:PSS- SWCNT/PDMS bump	-	$S = (\Delta R/R_0)/\Delta P$	0.5 kPa <sup>-1</sup>	28 Pa	1
AgNWs/WPU	-		0.12 kPa <sup>-1</sup>	10mg (24.5 Pa)	2
Interlocked microdome CNT/PDMS	-		15.1 kPa <sup>-1</sup>	0.2 Pa	3
Cotton fiber decorated with rGO/AgNWs	-		5.8kPa <sup>-1</sup>	0.125 Pa	4
Double-layered graphene	91.7% (active layer)		0.039 kPa <sup>-1</sup>	0.3 Pa	5
vertically aligned CNT/PDMS	-		0.3 kPa <sup>-1</sup>	20 Pa	6
Graphene/PDMS with random spinosum	-		25.1 kPa <sup>-1</sup>	16 Pa	7
Graphite nanoplatelet- CNT/PDMS	-		0.06 kPa <sup>-1</sup>	0.32 MPa	8
SSNPs/PU	85% (active layer)		2.53 kPa <sup>-1</sup>	0.3 kPa	9
Microdomes SSNPs/PU	77.7% (whole device)		71.37 kPa <sup>-1</sup>	4 Pa	This work
Konjac-derived CNFAs	-	$S = (\Delta I/I_0)/\Delta P$	1.02 kPa <sup>-1</sup>	10 Pa	10
AuNWs-impregnated tissue paper/PDMS	-		1.14 kPa <sup>-1</sup>	13 Pa	11
Aligned CNT/Graphene	81.4% (active layer)		19.8 kPa <sup>-1</sup>	0.6 Pa	12
CNTs-AgNPs/Sponge	-		9.08 kPa <sup>-1</sup>	11 kPa	13
Hollow structured graphene/PDMS	-		15.9 kPa <sup>-1</sup>	50 Pa	14
MXene/rGO aerogel	-		22.56 kPa <sup>-1</sup>	10 Pa	15
Carbon black-decorated fabric	-		0.585 kPa <sup>-1</sup>	1 kPa	16
PVDF@rGO nanofiber	-		15.6 kPa <sup>-1</sup>	1.2 Pa	17
Microdomes SSNPs/PU	77.7% (whole device)		99.88 kPa <sup>-1</sup>	4 Pa	This work

Table S1. Summary of the performance of piezoresistive sensors reported in the literature.<sup>1-17</sup>



**Figure S12.** Difference in the pressure sensitivity (*S*) calculated from (a)  $R/R_0$  changes and (b)  $I/I_0$  changes.



**Figure S13.** (a) Response and relaxation time of a piezoresistive pressure sensor based on a microdome structure. (b) Response time of a piezoresistive pressure sensor based on a microdome structure under different pressures (19.6, 78.4, and 346 Pa).



**Figure S14.** Augmentation index (AIx) and reflection index (RI) parameters for 1 min (33-year-old man). Note that average values of Aix and RI are about -16% and 15 m/s, respectively.

## References

- 1. E. Roh, H.-B. Lee, D.-I. Kim, and N.-E. Lee, Adv. Mater., 2016, 29, 1703004.
- 2. Y. Wei, S. Chen, X. Yuan, P. Wang, and L. Liu, Adv. Funct. Mater., 2016, 26, 5078-5085.
- J. Park, Y. Lee, J. Hong, M. Ha, Y.-D. Jung, H. Lim, S. Y. Kim, and H. Ko, ACS Nano, 2014, 8, 4689-4697.
- 4. Y. Wei, S. Chen, X. Dong, Y. Lin, and L. Liu, Carbon, 2017, 113, 395-403.
- 5. S. Chun, Y. Kim, H.-S. Oh, G. Bae, and W. Park, Nanoscale, 2015, 7, 11652-11659.
- K.-H. Kim, S. K. Hong, N.-S. Jang, S.-H. Ha, H. W. Lee, and J.-M. Kim, ACS Appl. Mater. Interfaces, 2017, 9, 17499-17507.
- Y. Pang, K. Zhang, Z. Yang, S. Jiang, Z. Ju, Y. Li, X. Wang, D. Wang, M. Jian, Y. Zhang, R. Liang, H. Tian, Y. Yang, and T.-L. Ren, *ACS Nano*, 2018, **12**, 2346-2354.
- 8. H. Zhao and J. Bai, ACS Appl. Mater. Interfaces, 2015, 7, 9652-9659.
- 9. D. Lee, H. Lee, Y. Jeong, Y. Ahn, G. Nam, and Y. Lee, Adv. Mater., 2016, 28, 9364-9369.
- 10. Y. Si, X. Wang, C. an, L. Yang, J. Yu, and B. Ding, Adv. Mater., 2016, 28, 9512-9518.
- S. Gong, W. Schwalb, Y. Wang, Y. Chen, Y. Tang, J. Si, B. Shirinzadeh, and W. Cheng, *Nat. Commun.*, 2014, 5. 3132.
- M. Jian, K. Xia, Q. Wang, Z. Yin, H. Wang, C. Wang, H. Xie, M. Zhang, and Y. Zhang, *Adv. Funct. Mater.*, 2017, 27, 1606066.
- H. Zhang, N. Liu, Y. Shi, W. Liu, Y. Yue, S. Wang, Y. Ma, L. Wen, L. Li, F. Long, Z. Zou, and Y. Gao, ACS Appl. Mater. Interfaces, 2016, 8, 22374-22381.
- N. Luo, Y. Huang, J. Liu, S.-C. Chen, C. P. Wong, and N. Zhao, *Adv. Mater.*, 2017, 29, 1702675.
- Y. Ma, Y. Yue, H. Zhang, F. Cheng, W. Zhao, J. Rao, S. Luo, J. Wang, X. Jiang, Z. Liu, N. Liu, and Y. Gao, *ACS Nano*, 2018, **12**, 3209-3216.
- N. Luo, W. Dai, C. Li, Z. Zhou, L. Lu, C. C. Y. Poon, S.-C. Chen, Y. Zhang, and N. Zhao, *Adv. Funct. Mater.*, 2016, 26, 1178-1187.
- 17. Z. Lou, S. Chen, L. Wang, K. Jiang, G. and Shen, Nano Energy, 2016, 23, 7-14.