Supporting Information

A highly stretchable dielectric elastomer based on core-shell structured soft polymer-coated liquid-metal nanofillers

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Experimental Details

Nanodroplets coating

The OA coated LM (EGaIn@OA) was synthesized as the precursor for the fabrication of EGaIn@PBMA. In this procedure, a certain amount of OA (Adamas, ACS, 99%) were mixed with 80ml tetrahydrofuran (THF, Sigma-Aldrich, ACS, 99%) and 200mg LM was dispersed in the solution under ultrasonication (180W) and vigorous stirring (600r/min) for 20min. The reaction was performed at 60℃ for 2h and then cooled to room temperature. The resulting EGaIn@OA nanodroplets were then treated by a free radical polymerization reaction using 25 mL of a THF solution consisting of 0.1-2ml BMA monomers (J&K, ACS, 99%) and a stoichiometric ratio of benzoyl peroxide (BPO, Aladdin, AR, >99%). The solution was then sonicated for 20 min and

transferred into a 50ml three-necked flask. The reaction was carried out with vigorous stirring under an N2 atmosphere and reflux in an oil bath at 60℃ for 24 h. The products were filtered using a 0.22μm PTFE membrane, washed with THF and ethanol twice to remove impurities, and then dried in a vacuum oven at 50 ℃ for 2 days. Four typical compositions of the reaction mixtures were shown in the Table S1 and the product was named as EGaIn@PBMA-1, EGaIn@PBMA-2, EGaIn@PBMA-3 and EGaIn@PBMA-4.

Nanocomposite fabrication

For the preparation of nanocomposites, the EGaIn@PBMA product was firstly ground into fine powders and then molded by hot-pressing at 160℃ under 5 MPa. The obtained EGaIn@PBMA samples were cut into 1mm(thickness)×20mm(width) dumbbell specimens coded as EGaIn-x@PBMA, where x (x=0,10,20,30,40) represented volume content of LM which was controlled by nanodroplets coating procedures. The parallel PBMA samples with uncoated LM were also prepared for comparative analysis by dispersing LM nanodroplets into BMA through in-situ polymerization without OA. The samples were coded as EGaIn-y/PBMA, where y (y=0,10,20,30,40) represented volume content of LM.

Characterization

The core-shell structure and elemental composition of EGaIn@PBMA were characterized using a high-resolution transmission electron microscope (HRTEM, JEOL, Japan) operated at 200kV. The characteristic functional groups on EGAIN@PBMA were characterized by Fourier transform infrared spectroscopy

(FTIR, Bruker Tensor 27, Germany). The morphology and microstructure of LM in nanocomposites were characterized using a scanning electron microscope (FE-SEM, JEOL, Japan). The tensile performance was tested on a universal testing machine (Instron-1121, UK) with a testing speed of 10 mm/min and five samples were tested for each kind of EGaIn@PBMA nanocomposites. Dielectric properties of the nanocomposites were measured on an impedance analyzer (LCR, Agilent 4294A, USA) within a frequency range from 40Hz to 30MHz and a 5mm diameter copper electrode was sputtered on both sides of the samples.
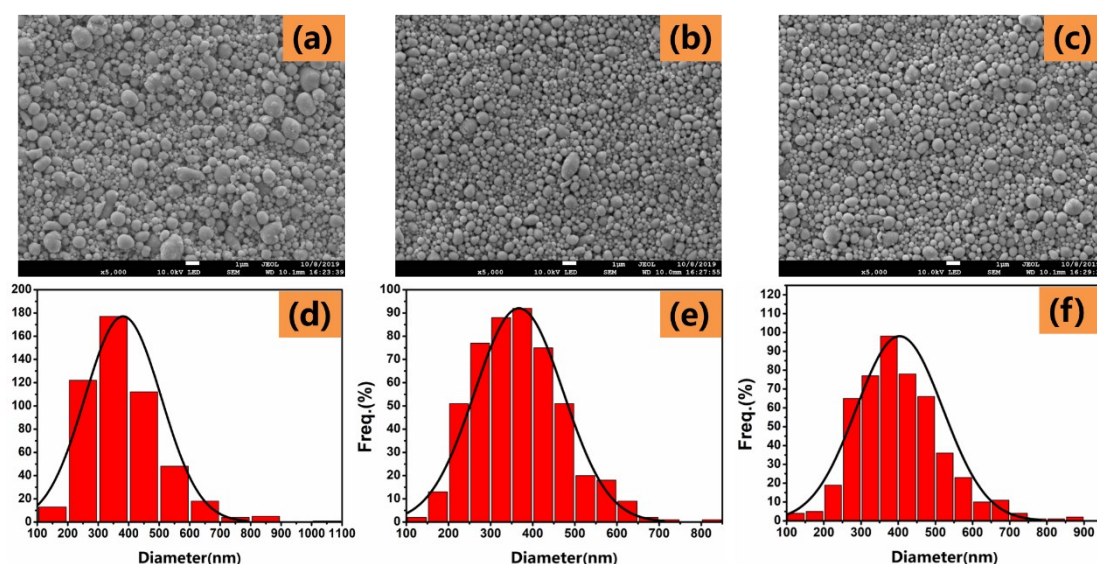

Fig S1. Particle size distribution of EGaIn@OA nanodroplets under different ultrasonic time: (a)(d) 10min; (b)(e) 15min; (c)(f) 25min.

Table S1. Compositions of the reaction mixtures

| Samples	LM	OA	BMA	BPO
EGaIn@PBMA-1	200mg	0.1ml	0.2ml	5mg
EGaIn@PBMA-2	200mg	0.1ml	1ml	18mg
EGaIn@PBMA-3	200mg	0.1ml	1.5ml	30mg
EGaIn@PBMA-4	200mg	0.1ml	2ml	40mg
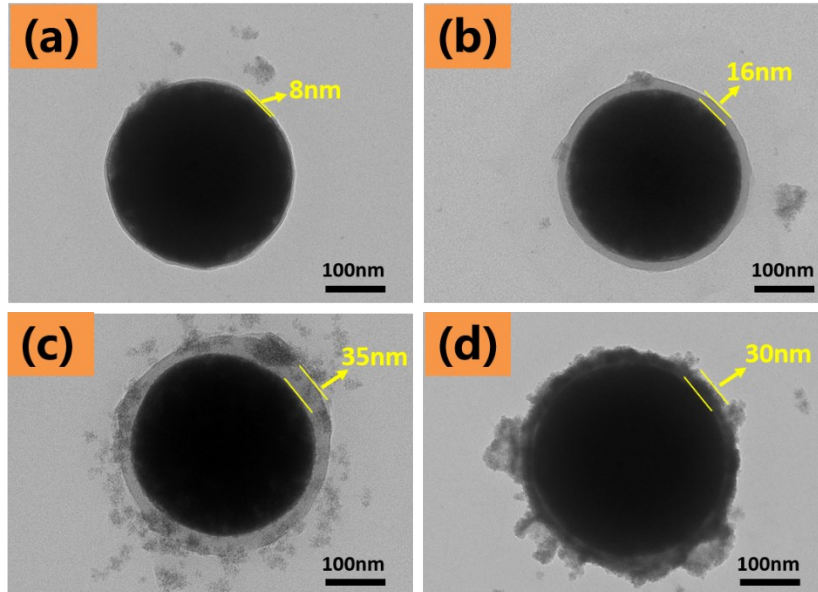
Fig S2. TEM images of EGaIn@PBMA with different polymer shell thickness: (a) EGaIn@PBMA-1 (b) EGaIn@PBMA-2 (c) EGaIn@PBMA-3 (d) EGaIn@PBMA-4
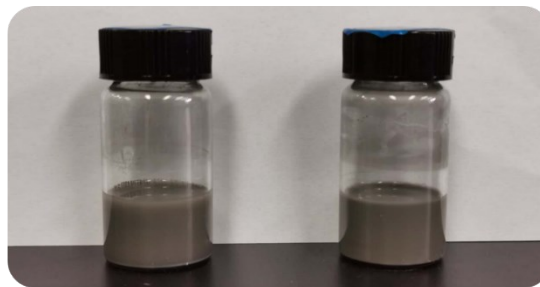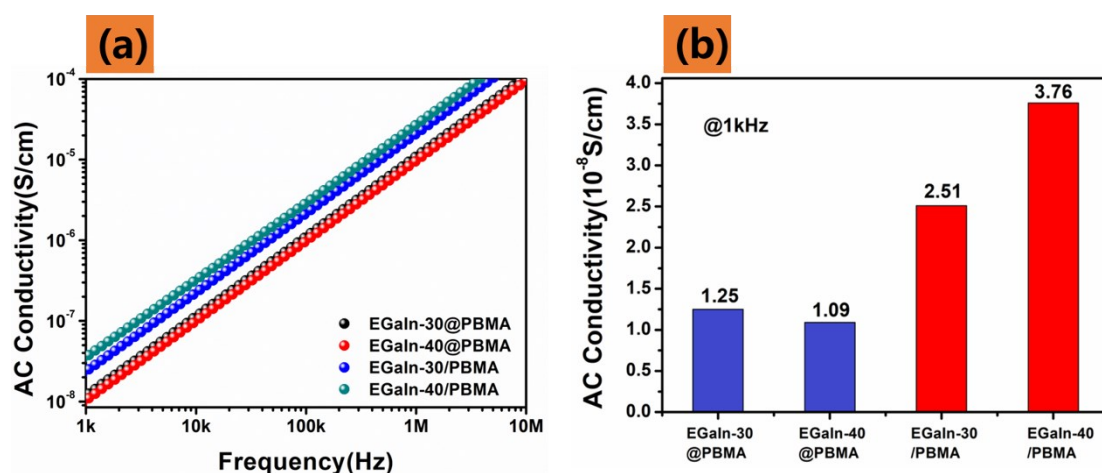

EGaIn@PBMA-1 EGaIn@PBMA-2

Fig S3. The dispersion state of EGaIn@PBMA-1 and EGaIn@PBMA-2 nanodroplets in the THF solvent in a week.
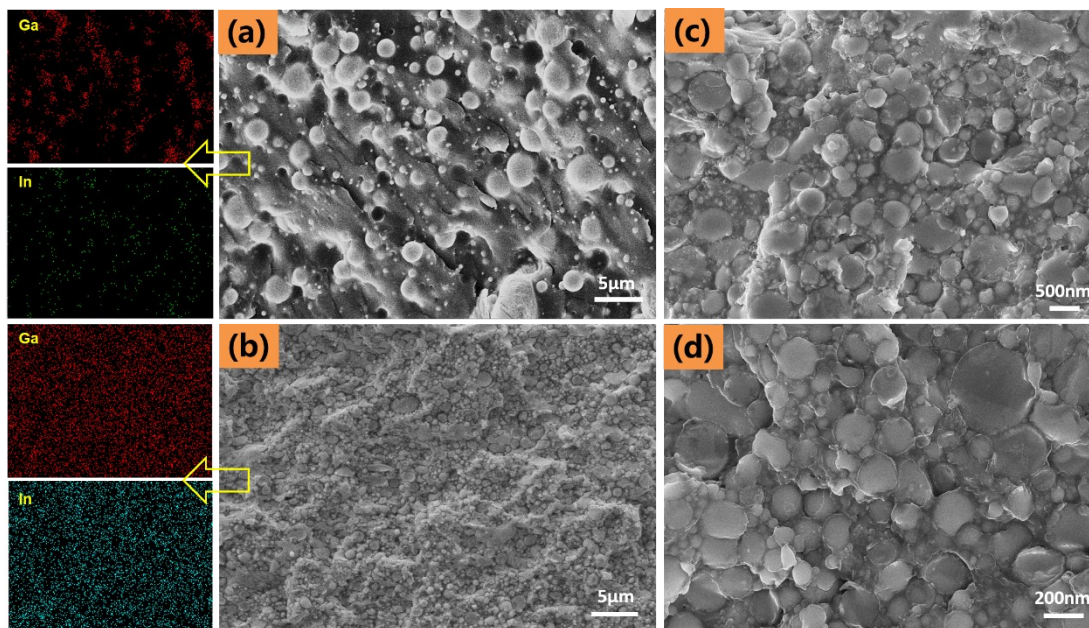
Fig S4. SEM images of fractured cross-sections of (a) EGaIn-40/PBMA and (b) (c) (d) EGaIn-40@PBMA composite. Corresponding EDS mapping was indicated by yellow arrow on the left.

Fig S5. (a) Frequency dependency on AC conductivity of EGaIn@PBMA and EGaIn/PBMA nanocomposites; (b) comparison of AC conductivity with different LM content at 1kHz.

It has been reported that a gallium oxide layer can be formed on the surface of EGaIn under ultrasonication in the air atmosphere.[20] Therefore, for composites filled with LM without modification, the conductive networks will not form even at a very high LM filling concentration. Fig S5. (a) shows that there was a frequency dependency for all nanocomposites with different LM volume content, i.e. the AC conductivity

increased with the rising frequency, implying no percolative networks were formed in both types of the systems. By comparing the conductivity of the two materials at 1kHz, it can be found that composites containing LM without PBMA insulating layers presented a higher conductivity value, which reflected the tunneling effect but not percolation caused by naked LM nanodroplets.