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Electronic Supplementary Information (ESI)

Construction of 3D aluminum flake framework by sponge template to prepare thermally conductive polymer composites

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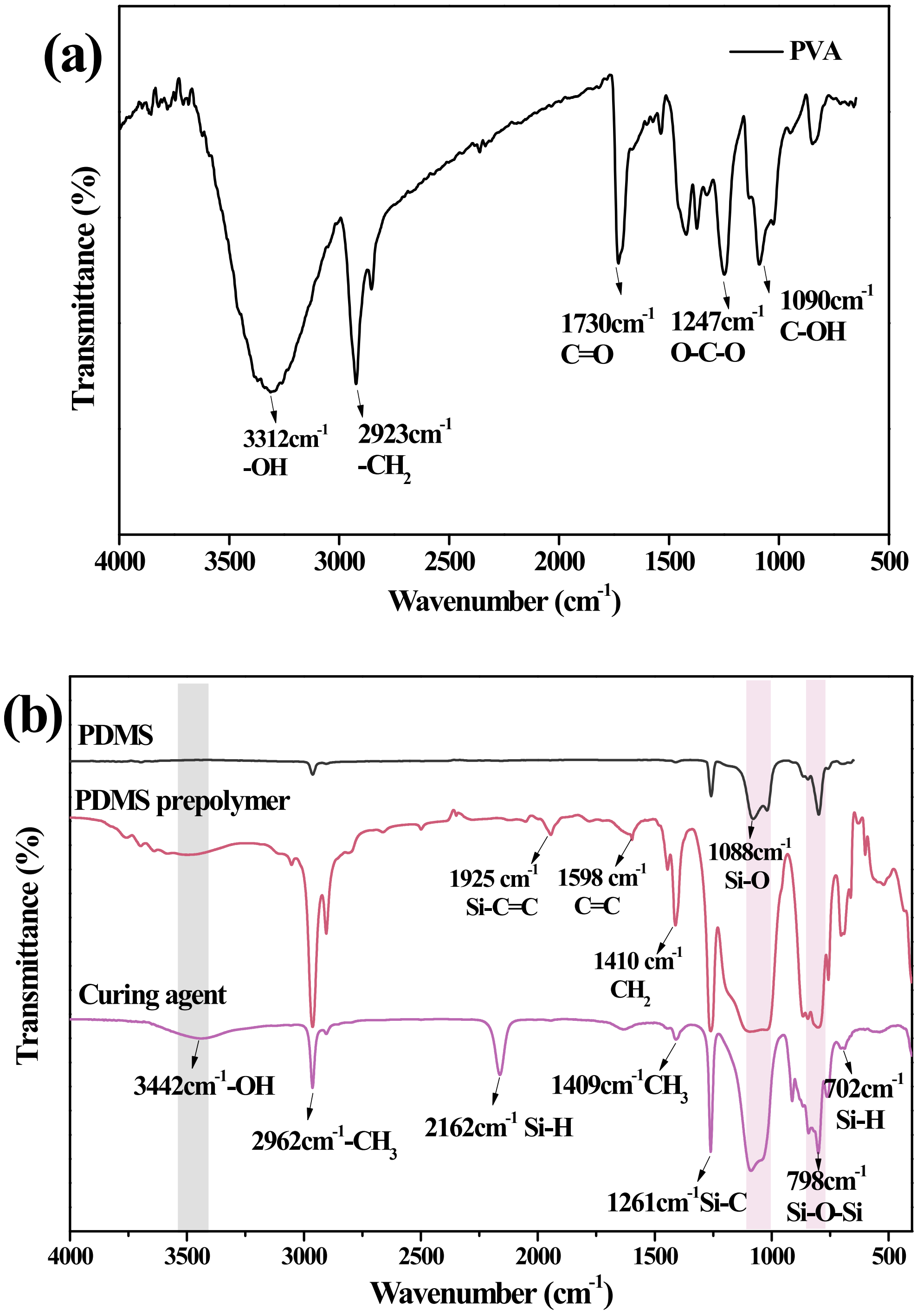


Fig.S1 FTIR spectra of PVA (a) and PDMS (b).

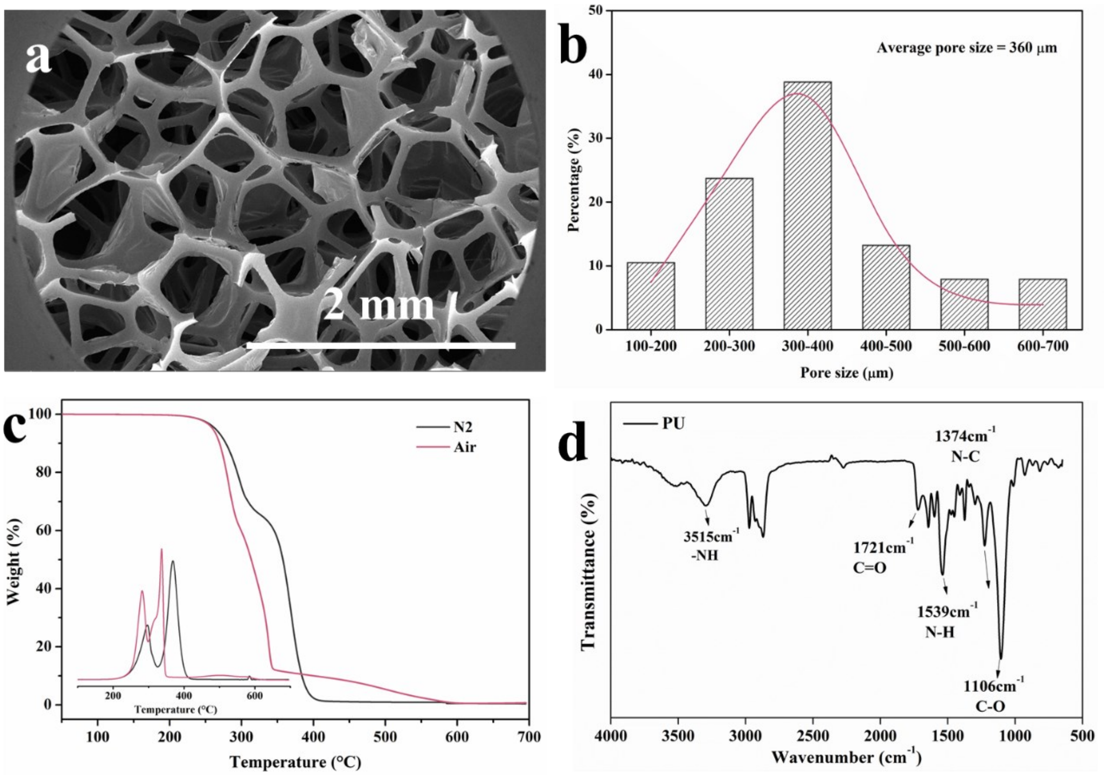


Fig.S2 Characterization of the PU sponge. (a) SEM, (b) caculated pore size, (c) DTG and FTIR of PU sponge.

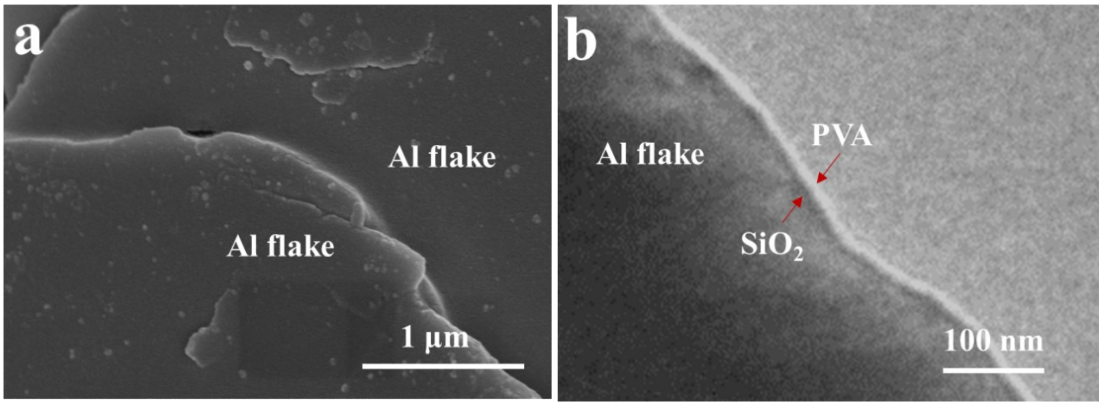


Fig.S3 High resolution SEM (a) and TEM (b) morphology of Al nanoflake in the surface of PU sponge

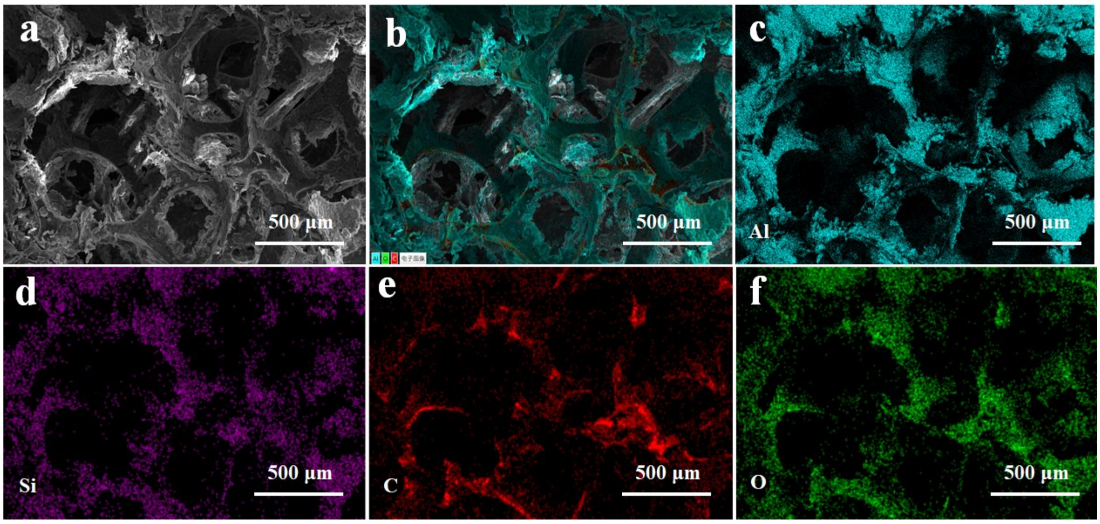
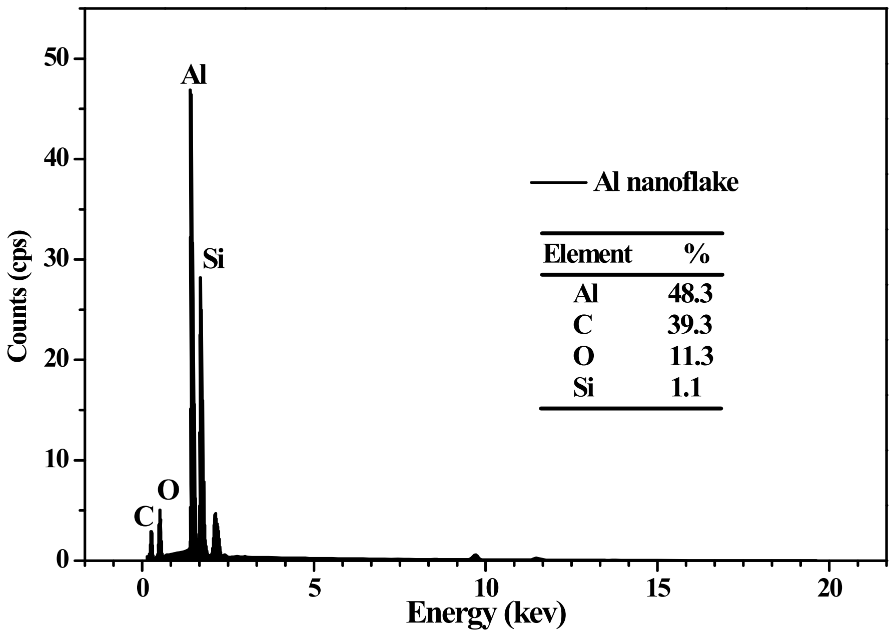


Fig.S4 (a-b) Selected SEM area for PU@Al$_{30}$ and the corresponding element mapping image of (c) Al, (d) Si, (e) C, and (f) O.



Element	%
Al	48.3
C	39.3
O	11.3
Si	1.1

Fig.S5 EDX analysis and element content in the surface of Al nanoflake.

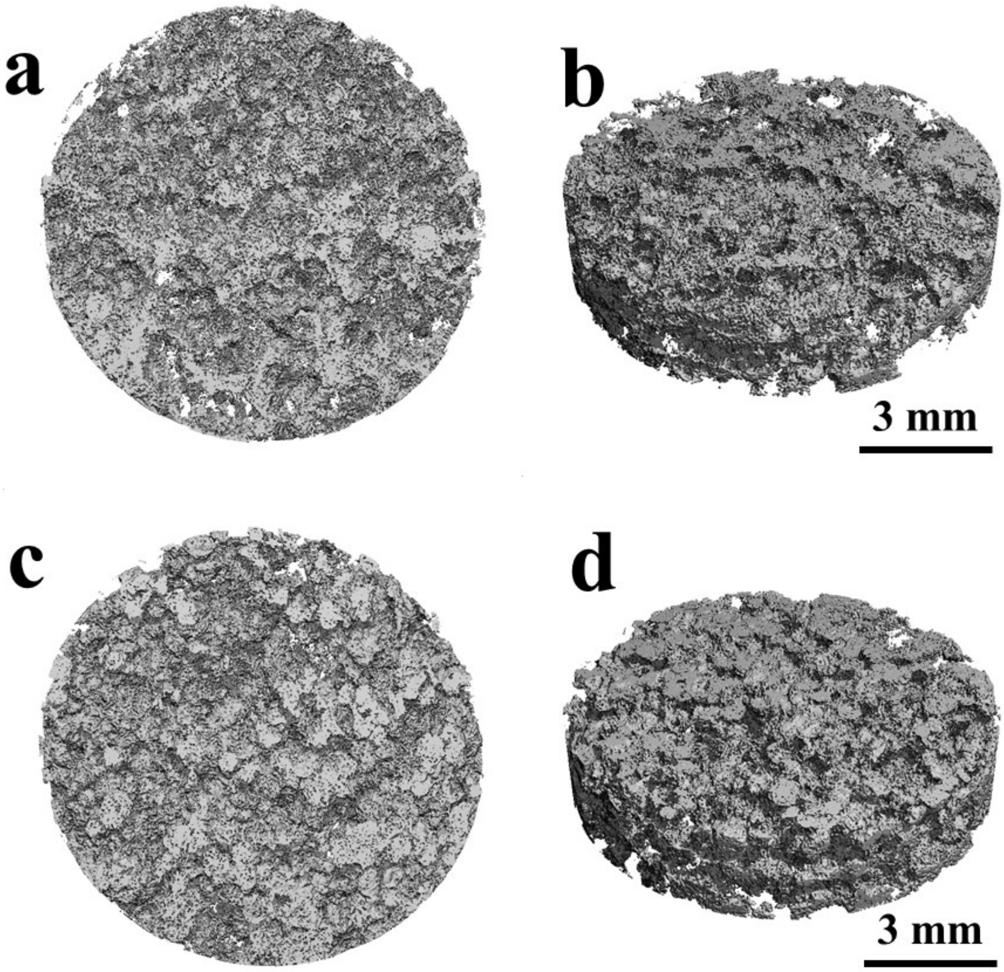


Fig.S6 Micro CT images of the (a-b) PDMS/PU@Al_{15} and (c-d) PDMS/PU@Al_{30} composites.

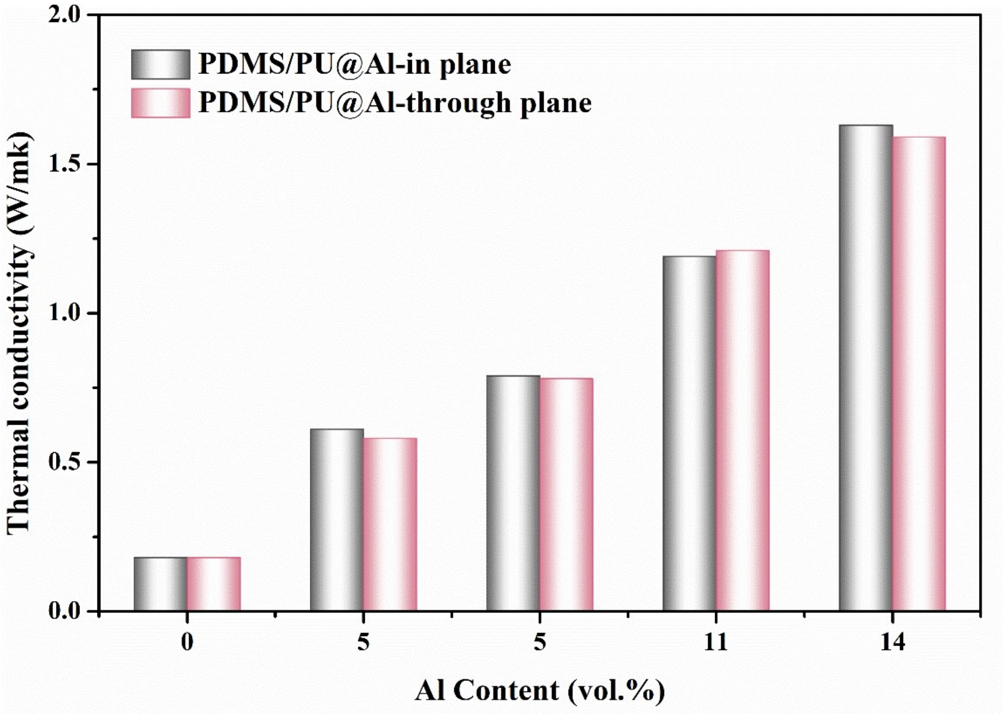


Fig.S7 Thermal conductivity and of PDMS/PU@Al composites at in plane and through plane direction.

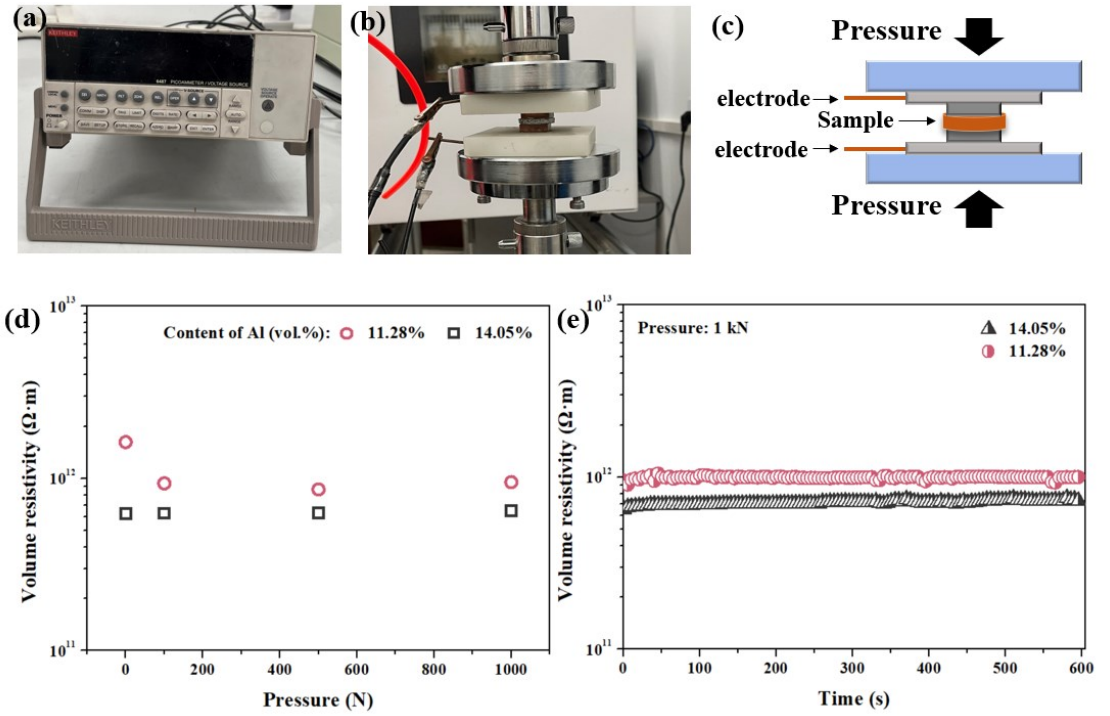


Fig.S8 Keithley 6487 (a). Resistance measurement process under pressure (b) and the schematic diagram (c). Volume resistivity-pressure (d) and volume resistivity-time (e) curves of PDMS/PU@Al composites.

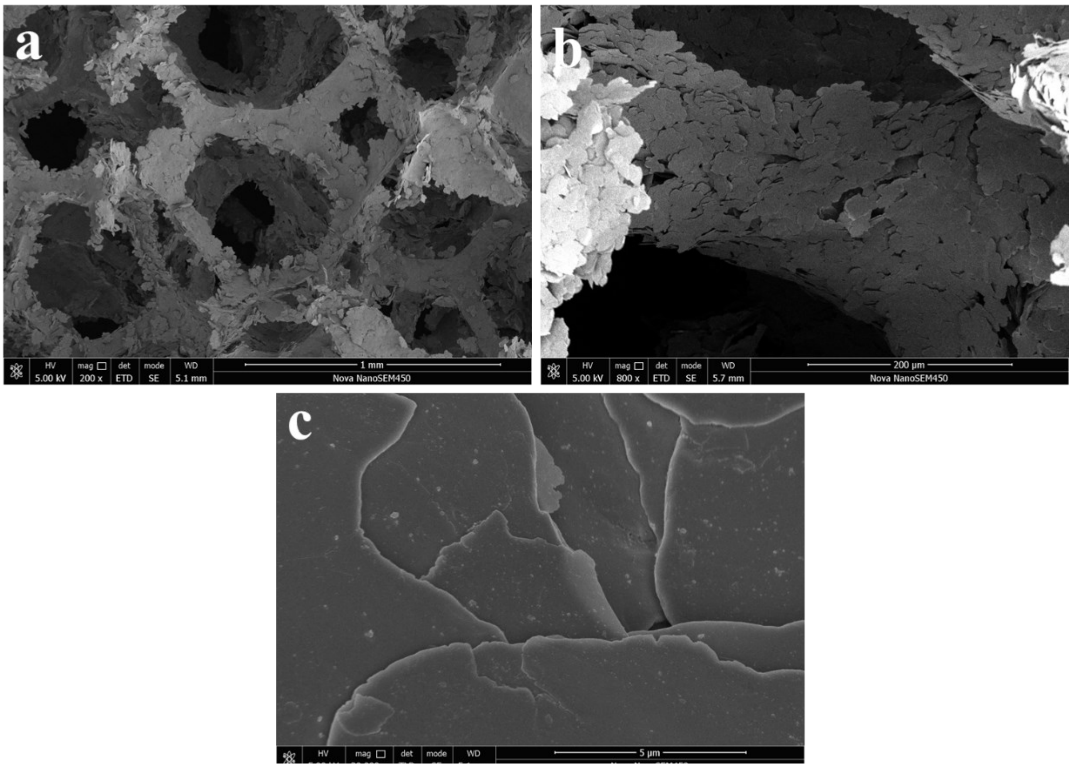


Fig.S9 SEM images of PU@Al_{30} sponge after compression with different magnification.

Method.S1 Calculation of interfacial thermal resistances of PDMS/Al

To further investigate the thermal conductive mechanisms of PDMS/Al and PDMS/PU@Al composites, the interfacial thermal resistance (ITR) are simulated by the effective medium theory (EMT) and Foygel's theory, respectively (Figure.S1). As for the PDMS/Al composites, ITR can be calculated using the EMT model, because Al flakes are randomly dispersed and the content of fillers was below 40 wt.%. [1-3] The EMT model is as follows:

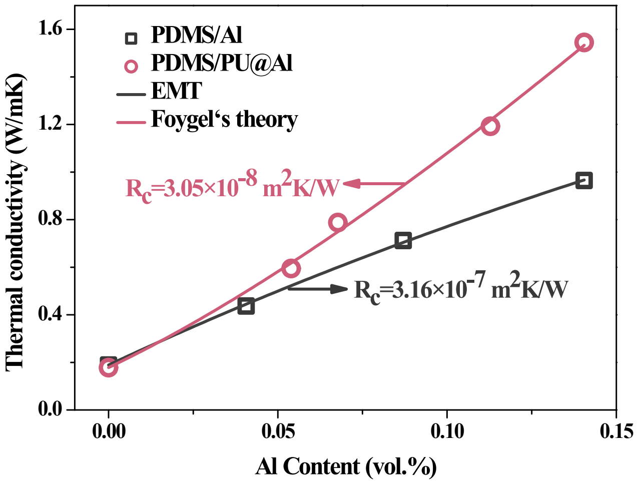
$$K = K_m \frac{3 + \varphi(\beta_1 + \beta_2)}{3 - \varphi\beta_1}$$

with

$$\beta_1 = \frac{2\left[d\left(K_{Al} - K_m\right) - 2R_{c1}K_mK_{Al}\right]}{d\left(K_{Al} + K_m\right) + 2R_{c1}K_mK_{Al}}$$

$$\beta_2 = \frac{L\left(K_{Al} - K_m\right) - 2R_{c1}K_{Al}K_m}{LK_m + 2R_{c1}K_mK_{Al}}$$

Where K, K_{Al}, and K_m are, respectively, the thermal conductivities of the composites, Al flakes (270 W/mK) and the pure PDMS (0.19 W/mK). φ is the volume fraction of the Al fillers, and d (80 nm) and L (15 μm) represent the diameter and length of Al flakes. By fitting the thermal conductivities of PDMS/Al composites in fitting curves, the interfacial thermal resistance (R_{c1}) between Al flakes and PDMS is 3.16×10^{-7} m^2K/W.



Fitting ITR results by EMT and Foygel's theory

Method.S2 Calculation of interfacial thermal resistances of PDMS/PU@Al

Compared with PDMS/Al composites, Al flakes in PDMS/PU@Al composites are closely connected to each other, and the ITR between fillers becomes the most important factor in thermal conductivities in PDMS/PU@Al composites. Thus, the nonlinear Foygel's model is proposed to calculate the ITR in PDMS/PU@Al composites, which can be expressed as follows[2, 4, 5]:

$$K = K_0(\varphi - \varphi_c)^{\tau}$$

$$R_c = \left(K_0 L \varphi_c{}^{\tau}\right)^{-1}$$

Where K_0 is a pre-exponential factor related to contact between Al flakes. φ_c is the critical volume fraction of Al flakes. τ is a conductivity exponent connected with the aspect ratio of the Al flakes. By fitting the thermal conductivity of PDMS/PU@Al composites, the Rc is calculated as 3.57×10^5 K/W. Considering the average overlap area between Al flakes, the ITR between Al flakes and PDMS can be obtained by the following equation:

$$R_{c2} = R_c \times \bar{A}_s$$

where

$$\bar{A}_s = \frac{2d^2}{\pi}\delta(p)$$

$$\delta(p) = \ln\left[\frac{\sqrt{1+p^{-1}}+\sqrt{1-p^{-1}}}{\sqrt{1+p^{-1}}-\sqrt{1-p^{-1}}}\right]$$

$$p = \frac{L}{d}$$

The resultant R_{c2} of PDMS/PU@Al composites is 3.05×10^{-8} m^{2}K/W.

Table.S1 Major parameters used in the simulation

Parameters	**PU**	**Al**	**PDMS**
Thermal conductivity (W/(m K))	0.25	219	0.16
Specific heat capacity (J/(g K))	1380	880	270
Density (kg/m^3)	1250	2700	1030

Reference

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