Electronic Supplementary Material (ESI) for Soft Matter.

Supplementary Information

Investigation of Thermal Conductivity for Liquid Metal Composites Using Micromechanics-Based Mean-Field Homogenization Theory

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1. Results using other mean field homogenization methods

Theoretical predictions from the Maxwell-Garnett's model and Bruggeman's theory are presented in previous studies [7], [8-10].

Maxwell-Garnett's model

$$\kappa_{eff} = \kappa_m \left(\frac{(1-c_1)(\kappa_1 + 2\kappa_0) + 3c_1\kappa_1}{(1-c_1)(\kappa_1 + 2\kappa_0) + 3c_1 k_0} \right)$$

Bruggeman's theory

$$\left(\frac{\kappa_1 - \kappa_{eff}}{\kappa_1 - \kappa_0} \right) \left(\frac{\kappa_0}{\kappa_{eff}} \right)^L = 1 - c_1 \text{ where } L = \frac{1}{3} \text{ for spherical inclusion}$$

We note that the predictions from Mori-Tanaka method and Maxwell's model are identical as in previous study [28]. Bruggeman's model shows similar results with double inclusion model with interpolation function $\zeta(c_1) = c_1$.

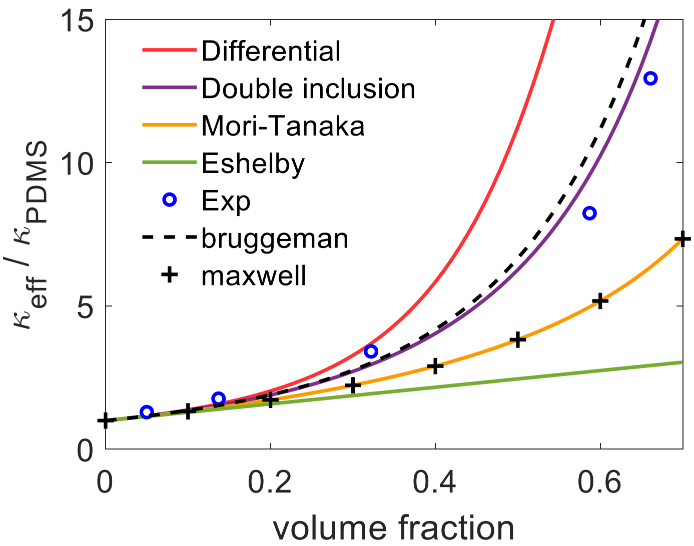

Fig. S1. Different homogenization models for effective thermal conductivities of LMCs. Predictions from Maxwell-Garnett's model and Bruggeman's theory are added. The thermal conductivities are normalized by the thermal conductivity of PDMS.

2. Results with variation of aspect ratio

Normalized effective thermal conductivities of LMCs when aspect ratios are 1, 2, 3, 4 and 5 are presented in Fig. S1. As the aspect ratio increases, the effective thermal conductivity increases. It is difficult to synthesize LMCs with elongated inclusions since the surface tension suppresses the presence of inclusions with a high aspect ratio. Otherwise, an elastomer matrix needs to be deformed, which is different from an original shape. This result shows that LMCs can have better thermal conductivity if experimental techniques are supported to facilitate the synthesis of LMCs with elongated inclusions.

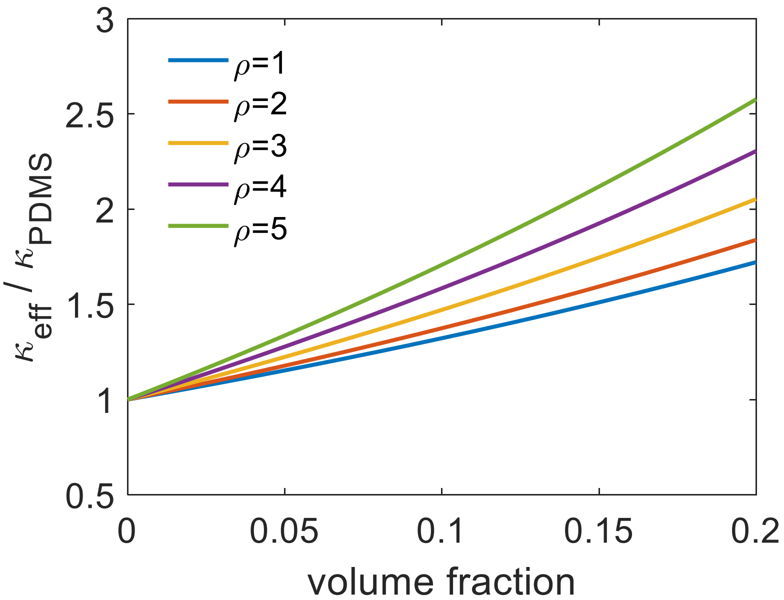

Fig. S2. Normalized effective thermal conductivities of LMCs when aspect ratios are 1, 2, 3, 4 and 5, respectively. Ellipsoidal LM inclusions are randomly oriented.

3. Results for PDMS with 10% increased thermal conductivity

We found that, if the thermal conductivity of PDMS within LMCs increases by 10% (from 0.17 W/m·K to 0.187 W/m·K), the theoretical prediction matches better with the experimental data as shown in Fig. S2 and Table S1.

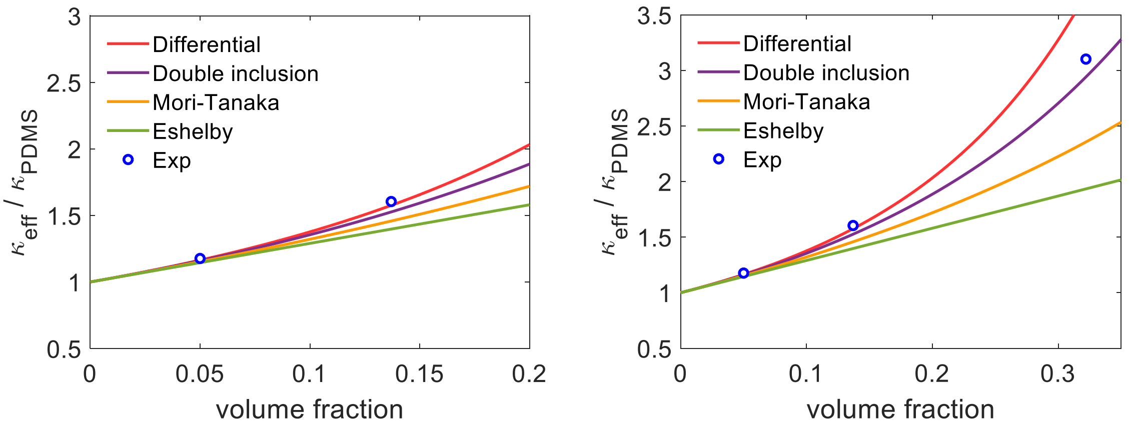

Fig. S3. Different homogenization models for effective thermal conductivities of LMCs when the thermal conductivity of the PDMS is increased by 10% from the experimental data.

LM Volume fraction	5.0%	13.7%	32.2%
Eshelby	2.67%	12.9%	37.6%
Mori-Tanaka	2.04%	9.11%	24.0%
Differential	1.03%	1.78%	18.8%
Double inclusion	1.40%	4.87%	5.18%

Table S1. Relative errors for each homogenization method compared to the experimental data when the thermal conductivity of the PDMS is increased by 10% compared to the measured value.

4. Convergence of theoretical prediction of the effective thermal conductivity

As far field strain increases, the aspect ratio of liquid metal inclusion increases continuously, and our theoretical prediction converges to the upper limit, the thermal conductivity of laminated composite consisting of long liquid metal fibers.

$$\text{upper limit} = (1 - c_1)\kappa_0 + c_1\kappa_1$$

In reality, LMC fractures before the effective thermal conductivity reaches upper limit.

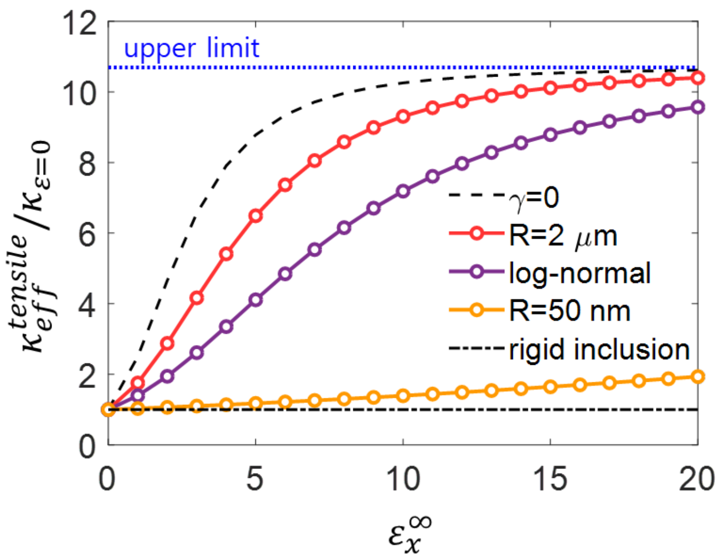

Fig. S4. Effective thermal conductivities in the tensile direction under the far field strain at 20% of volume fraction. Thermal conductivities are normalized by thermal conductivity at zero strain.