SUPPLEMENTARY MATERIALS

Large Magnitude Transformable Liquid Metal Composites

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Supporting information is included as follows:

Supporting text, the analytic relation of the expansion induced by the heating temperature

Supporting Figures and Legends S1-S7

Supporting Movie Legends S1-S8
Theoretical analysis of the expansion of LMEE matrix

To quantitatively predict the expansion and deformations of LMEE induced by heating, we derived the analytical relation of the expansion induced by the heating temperature. We assume that the ethanol droplet uniformly embedded in the LMEE matrix with total volume fraction $\eta$. The average radius is $R_0$ in the undeformed LMEE, and $R$ in the deformed elastomer due to the transition from liquid ethanol to vapor state. Then the volumetric expansion is estimated by

$$
\alpha = 1 + \eta \left[ \frac{R}{R_0} \right]^3 - 1
$$

For the small value of volume fraction $\eta$ (lesser than 20%), we suppose that the interaction of droplets is negligible. Thus, considering a single ethanol droplet with radius $R_0$ embedded in the infinite LMEE matrix illustrated in Fig. 1, it would boil and become vapor upon reaching the liquid-vapor transition temperature, and lead to significant expansion of the whole LMEE matrix.

We assume that $(R, \Theta, \Phi)$ and $(r, \theta, \phi)$ are the spherical coordinates in the undeformed and deformed configurations, respectively. With the assumption of centrally symmetric deformation, the three principal stretches are given by

$$
\begin{align*}
\lambda_r &= \frac{dr}{dR} \\
\lambda_\theta &= \frac{\lambda}{r} = \frac{r}{R} \\
\lambda_\phi &= \hat{\lambda} = \frac{r}{R}
\end{align*}
$$

When LMEE matrix is taken to be incompressible, $\lambda_r \lambda_\theta \lambda_\phi = 1$ would be met. And, the radial stretch is given by $\lambda_r = \hat{\lambda}^2$. Then the stress equilibrium equation of the symmetric sphere, in the absence of body forces in the radial direction, is expressed as:
\[ \frac{d\sigma}{dr} + 2\frac{\sigma_r - \sigma_\theta}{r} = 0 \]  
(3)

Here \( \sigma_r = \sigma_\theta \) is assumed due to the spherical symmetry. Constant uniform radial pressure \( P_v \) and \( P_0 \) are applied at the inner and outer surfaces of the sphere. Therefore, the boundary conditions are:

\[
\begin{aligned}
\sigma_r (r = R_0) &= -P_v + 2\gamma/a \\
\sigma_r (r = \infty) &= -P_0 
\end{aligned}
\]  
(4)

where \( \gamma \) denotes the surface tension of the ethanol vapor with LMEE matrix.

The stress model described by Eqs. (2-4) discussed here is very similar to the work of Zhu et al \([1]\), which is aimed to investigate snap-through expansion of a gas bubble in an elastomer. Thus, we give the main results, and the detailed discussion could be found in the Zhu work \([1]\). We adopt the Gent free-energy function \([1]\)

\[
W_{el} = \frac{\mu J_{lim}}{2} \ln \left(1 - \frac{J}{J_{lim}}\right) \\
\sigma(\lambda) = \sigma_r - \sigma_\theta = \frac{\lambda}{2} \frac{dW_{el}}{d\lambda} 
\]  
(5)

where \( J = 2\lambda^2 + \lambda^{-4} - 3 \), and \( J_{lim} = J(\lambda_{lim}) \) is a constant related to the limiting stretch \( \lambda_{lim} \).

Through integration of the stress equilibrium equation of Eq.(3), and combining with Eq.(4) and Eq.(5), we could obtain the relation \([1]\)

\[ P_v = P_0 + \frac{2\gamma}{R} + \frac{\mu}{2} \int_{R/R_0}^{\infty} \frac{(\lambda^{-2} + \lambda^{-5})}{1 - J_{lim}^{-1}(2\lambda^2 + \lambda^{-4} - 3)} d\lambda \]  
(6)

The above equation indicates that the ethanol vapor pressure is related to its radius stretch. For the given heating temperature, the saturation vapor pressure could be determined, which is equal to \( P_v \) under the thermal equilibrium. Thus, we could estimate the radius stretch \( R/R_0 \) through Eq. (6) for the given \( P_v \), and the volumetric expansion through Eq. (1) is then obtained. The thermal properties of ethanol saturation vapor pressure, and its saturation liquid and vapor density are given in Fig.4 \([2-3]\). In order to confirm whether liquid ethanol completely become vapor, the boiling ethanol vapor mass ratio could be estimated based on the conservation of mass, and given by

\[ m_v(T) = \frac{\rho_l(T) \left( \frac{R}{R_0} \right)^3 \rho_v(T)}{\rho_l(T_0) \left[ \rho_l(T) - \rho_v(T) \right]} \]  
(7)
where $\rho_l(T)$ denotes the liquid saturation ethanol density at the temperature T, and $\rho_v(T)$ for the vapor saturation ethanol density. $T_0$ denotes the ambient temperature, which is considered as 25°C here. $m_r(T) = 1$ indicates that the liquid ethanol is completely transited into vapor, and the ethanol may be the superheated vapor. For such case, the density cannot be determined by Fig.2. However, for most cases considered here, the liquid ethanol can not be completely transited into vapor due to the larger vapor saturation pressure.

References
[1] Zhu, J., Li, T., Cai, S., and Suo, Z. Snap-through expansion of a gas bubble in an elastomer. The Journal of Adhesion, 2011(87):466-481.
[2] https://www.thermalfluidscentral.org/encyclopedia/index.php/Thermophysical_Properties:_Ethanol;
[3] https://webbook.nist.gov/cgi/cbook.cgi?ID=C64175&Mask=4
Supporting Figures and Legends S1-S7

**Figure S1.** Microstructure change of composite when stretching.
Figure S2. Expansion mechanism of LMC. (a) Ethanol bubbles are distributed in the composites, scale bar is 20 μm. (b) Micro-CT scan showing the 3D microstructure of the LMC, the cavity is made by ethanol vapor and the white dots represent liquid metal droplets (c) Ethanol liquids evaporate in response to the heat.
Figure S3. Microstructure of LMC with different stirring time. (a) Metalloscope micrography images of the LMC, which is stirred for 50 seconds. (b) Metalloscope micrography images of the LMC, which is stirred for 500 seconds.
Figure S4. Deformation of LMC in response to heat. (a) LMC swells and deforms when subject to microwave heating (b). (c) (d) LMC with bar shape curves into a wave-shape on the hot plate. (e) Fabrication process of a three-layer configuration for electromagnetic induction heating.
**Figure S5.** The LMC ball deformation after repeated heating and cooling accompanied with surface color changing (grey and silver).

**Figure S6.** Expansion properties of LMC. (a) Swelling in local area of LMC and heaves itself after heating. (b) The bump of LMC are disappearing gradually. (c) The height of expansion over times.
of heating and cooling. After about 24 cycles of heating and cooling, LMC expansion height declines sharply.

![Figure S7](image)

**Figure S7.** Demonstration of various deformation behaviors. (a) A tri-leg shape LMC bends and up-warsps its three legs on hot plate. (b) LMC inflate like a puffer in response to heat.

**Supporting Movies and Legends:**

**Movie S1:** LMC expands like a balloon in response to heat, which exhibits the ultra elasticity.

**Movie S2:** LMC transforms between planar shape and 3D structure reversibly in response to thermal stimulus. This phenomenon is caused by the phase change of ethanol via temperature regulation.

**Movie S3:** LMC swells sequentially with selective heating.

**Movie S4:** A cylinder shape LMC hunches and recovers in the target regions when heated and cooled.

**Movie S5:** A 2D LMC-Made octopus inflate itself in response to the selective heating.

**Movie S6:** Inching behavior of a soft robot made of LMC when subject to heating and cooling circularly.

**Movie S7:** LMC gripper bends and grabs the object due to the uneven expansion in response to thermal stimulus.

**Movie S8:** A three-leg robot warps on a hot plate due to the uneven expansion.