Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supporting Information

Self-Healing of Electrical Damage in Thermoset Polymers via Anionic Polymerization

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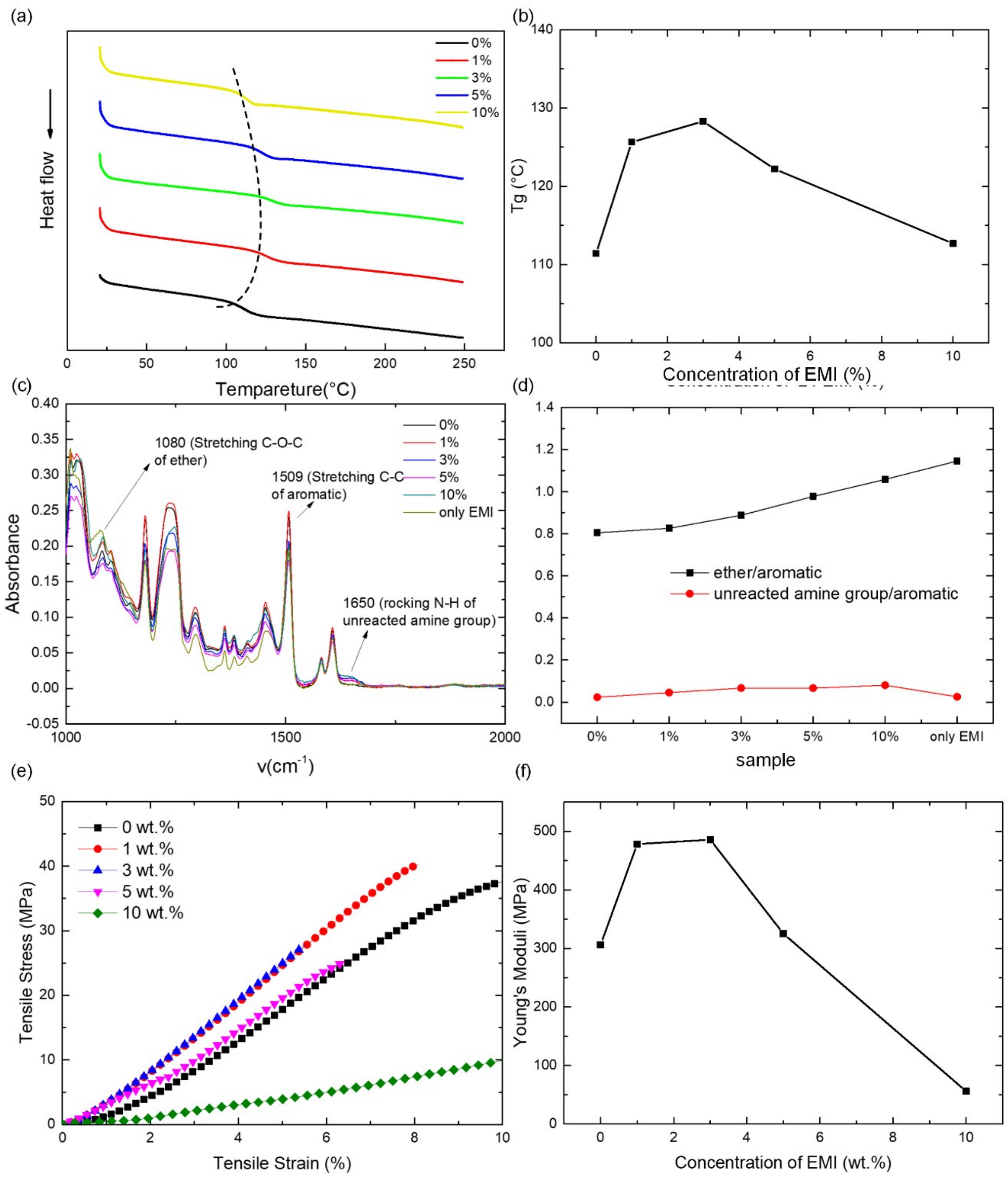

Figure S1. a) DSC results of EMI-modified epoxy with different concentrations of EMI. b) shows the adding of EMI increased the glass transition temperature when the concentration was below 5 wt.%. c) FITR results of EMI-modified epoxy slices (500 μm) with different concentrations of EMI. 1080 cm^{-1} is the stretching C-O-C of ether. 1509 cm^{-1} is the stretching C-C of aromatic rings. 1650 cm^{-1} is the rocking N-H of unreacted amine group. d). shows the ratio of the ether's peak strength to aromatic' and the ratio of the unreacted amine group's peak strength to aromatic'. The group of 'only EMI' represents the epoxy cured without PAA but only by 3 wt.% EMI and is presented for compare. As the imidazole content increased, the ether's peak strength and amine group's peak strength both increased gradually, which implied that partial epoxy groups were cross-linked by etherification reactions initiated by EMI-adducts. Strain-stress curves (e) and the corresponding Young's moduli (f) of the various samples with changing the EMI content. The trend of Young's modulus of the samples matches that of the breakdown strength, which implies that the electromechanical failure caused by mutual Coulombic force from the opposite electrodes under an applied field may account for the dependence of breakdown strength on EMI content.

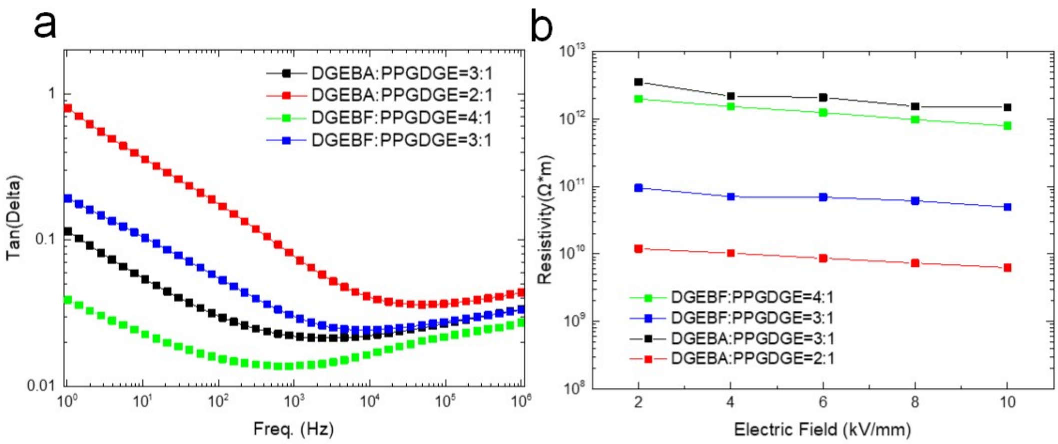

Figure S2. Dielectric loss tangent (a) and resistivity (b) of different formulas of healing agent cured by 3 wt.% EMI.

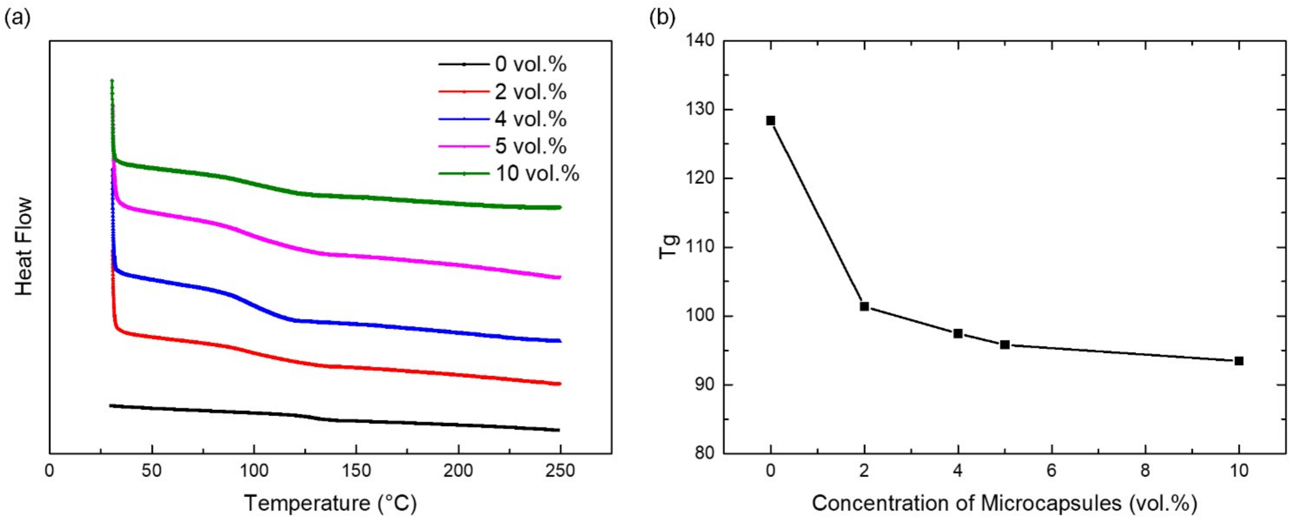

Figure S3. DSC results (a) and Tg (b) of the self-healing materials with different concentrations of microcapsules.

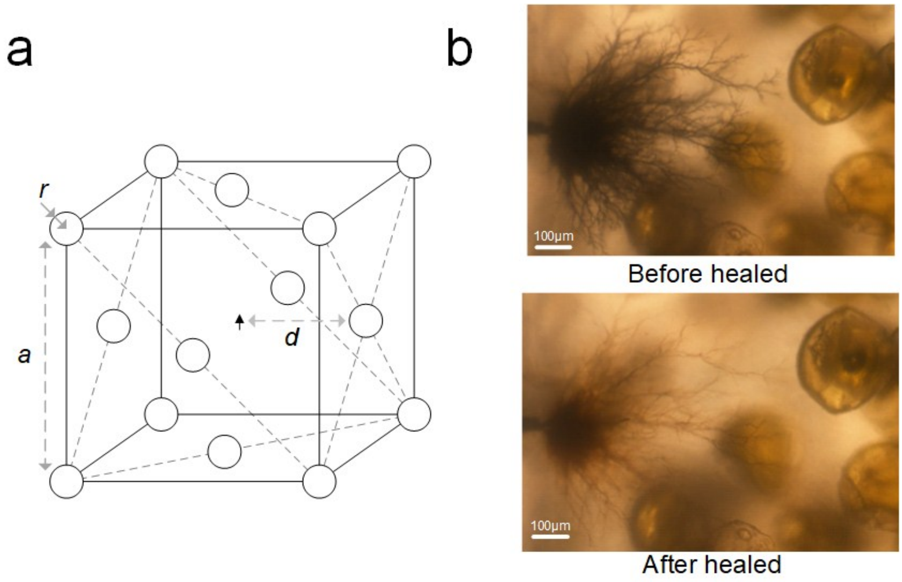

Figure S4. a) The model of cubic closest packing which was used to estimate the longest distance *d* between microcapsules and possible existed defects if the microcapsules evenly distributed in the material like cubic closest packing. b) Electrical trees couldn't be healed well if the trees grew too long and too crowd since the material had been damaged too much.

If we want to heal the defects and recover the dielectric properties of the material, the electrical trees couldn't be too large or the insulating properties of the material might already fall too much. In addition, large trees with dense channels cannot be fully healed too. So we want the electrical tree to hit the microcapsules before the tree is too long. Supposing that microcapsules evenly distribute in the material like cubic closest packing and the radius is *r*.

As **Figure S4a** shows, If the content of the microcapsules is *a* (volume fraction), we can calculate the longest distance *d* between microcapsule and possible existed defect as follows.

$$\frac{\frac{4}{3}\pi r^3 * 8 * \frac{1}{8} + \frac{4}{3}\pi r^3 * 6 * \frac{1}{2}}{[2 * (d + r)]^3} = a \qquad (1)$$

From the formula (1), the longest distance d depends on microcapsules' radius r and content a. If a = 0.05, then d = 2.47 r. If a = 0.03, then d = 3.12r. This is the longest distance. In most situation, the distance between microcapsule and defect is much smaller. Above analysis assures most electrical trees can always hit a microcapsule when it is small. In our experiment, the average diameter of microcapsules was 200 μm. Therefore the longest distance was 312 μm if the concentration of microcapsules was 3 % (volume fraction). Experiment results showed that these lengths of tree channels didn't decrease electrical property too much and can be well healed. However, if the distance was too long, the tree would be too large and couldn't be healed well as Figure S4b shows.

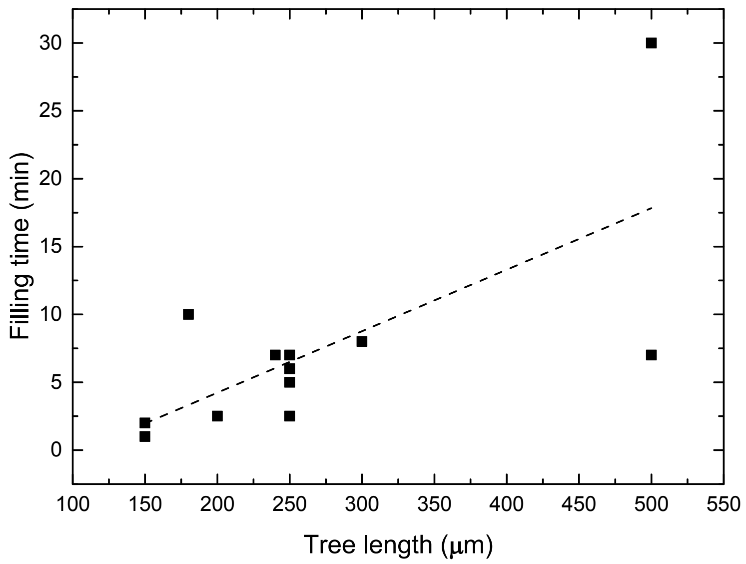

Figure S5. The time for healing agent to fill electrical tree channels completely after the tree hit the microcapsule. The filling time approximately behaved a linear relationship with tree length and ranges from several minutes to half an hour.

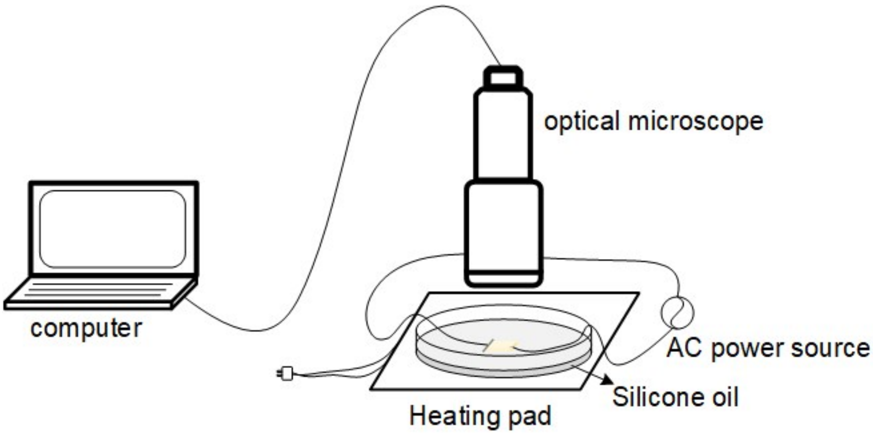

Figure S6. The Electrical tree aging platform for aging and healing test. The aging and healing process can be observed on the computer through optical microscope. Suitable conditions were controlled by the heating pad and AC power source. The AC power source was made up with a function generator (Tektronix AFG3102C) which could generate any type of waveform and a power amplifier (TREK, model 50/12-H-CE) which could amplify the waveform to 5000 times of its origin voltage.

Table S1. The dielectric properties of different healing agent group.

EP828：PPGDGE	**3：1**	**2：1**
Resistivity (Ω*cm)	1.48×10^{14}	6.25×10^{11}
Dielectric loss angle tangent	0.0343	0.2112
Viscosity (cST)	2440	1223
EP862：PPGDGE	**4：1**	**3：1**
Resistivity (Ω*cm)	0.79×10^{14}	4.94×10^{12}
Dielectric loss angle tangent	0.0168	0.0647
Viscosity (cST)	1493	1225

Table S2. The apparent discharge magnitudes of self-healing samples with different concentrations of EMI before aging, after aging and after healing when applied voltage was 8kV.

Concentration of EMI	Before aging (pC)	After aging (pC)	After healing (pC) [a]
0 wt%	0.17 [b]	20~200 [c]	20~200
1 wt%	0.17	20~200	0.17
3 wt%	0.17	20~200	0.17
5 wt%	0.17	20~200	0.17

[a] after healing means after running at 55 °C for 150 hours; [b] 0.17 pC was the minimum value which the partial discharge detector can measure; [c] because the apparent discharge magnitudes of samples with different shapes of electrical tree were different, all kinds of results fell between 20 pC~200 pC.

Movie S1. The flowing process of the healing agent into the tree channels.