

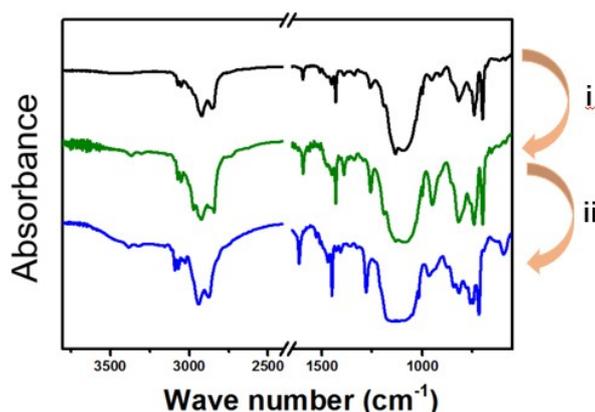
Simultaneous reinforcement and toughness improvement of epoxy-phenolic network with a hyperbranched polysiloxane modifier

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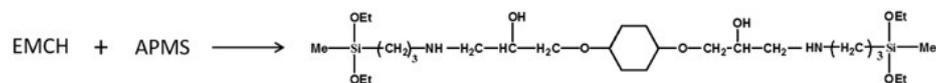
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i:



ii:

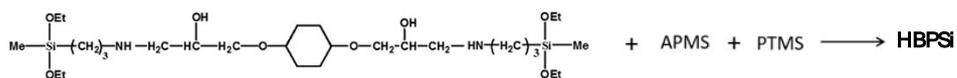


Fig.S1 The FTIR spectra of mixtures of reactants (i-ii)

After the first step of reaction, the absorption peak at 915 cm^{-1} , which is the characteristic stretching absorption peak of the epoxy group (C–O), disappears, indicating that all the epoxy groups on EMCH have been consumed. Then, in the spectrum of the product, the characteristic stretching absorption peak of hydroxy at 3400 cm^{-1} to 3600 cm^{-1} shows up, indicating the $-\text{Si-O-Me}$ and $-\text{Si-O-Et}$ have hydrolyzed into silanol groups (Si-OH).

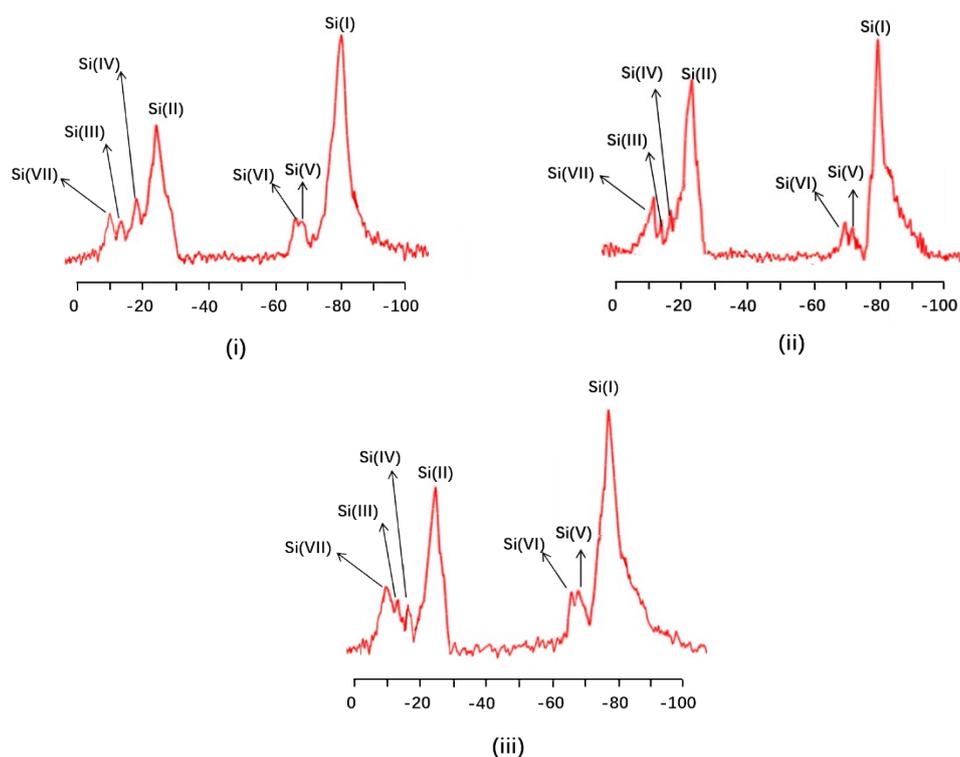


Fig.S2 ^{29}Si NMR spectrum of HBPSi-1 (i), HBPSi-2 (ii) and HBPSi-3 (iii)

The ^{29}Si NMR spectrum of HBPSi is depicted in Fig.S2. Attribution of peaks in the ^{29}Si NMR spectrum of HBPSi is summarized in Table S1. Peaks numbered as Si(I) and Si(II)

Table.S1 Attribution of peaks in the ^{29}Si NMR spectrum of HBPSi

D	Si(I)	Si(II)	
T	Si(VI)	Si(VII)	
L	Si(III)	Si(IV)	Si(V)

are assigned to dendritic units (D). Peaks numbered as Si(III), Si(IV) and Si(V) are assigned to linear units (L). Peaks numbered as Si(VI) and Si(VII) are assigned to terminal unit (T). The degree of branching (DB) of HBPSi can be calculated as

follows:

$$DB_1 = 2D / (2D + L) \quad \text{eq}_1$$

$$DB_2 = (D + T) / (D + T + L) \quad \text{eq}_2$$

Table.S2 The degree of branching of HBPSi.

Sample	DB ₁	DB ₂
HBPSi-1	0.92	0.85
HBPSi-2	0.94	0.89
HBPSi-3	0.97	0.90

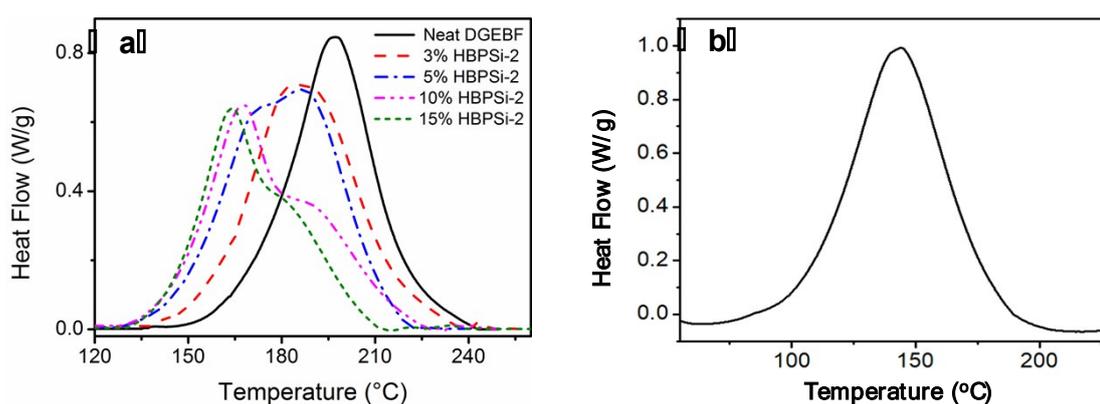


Fig.S3 Exotherm DSC curves for samples with varying HBPSi-2 contents (a) and system of DGEBF and HBPSi-2.

DSC was used to characterize the formation of HBPSi-modified epoxy networks. Because the modified epoxies containing HBPSi-2 show the best overall performance, the DSC results are only demonstrated for the case of epoxy-HBPSi-2 system. As shown in Fig.S3 (a), the initial reaction temperature and peak temperature both decrease with increasing HBPSi-2 content. When the HBPSi-2 loading is higher than 5%, two obvious peaks appear in the exothermic curves. The reaction process of the epoxy and HBPSi-2 was studied by DSC analysis of the system of DGEBF and

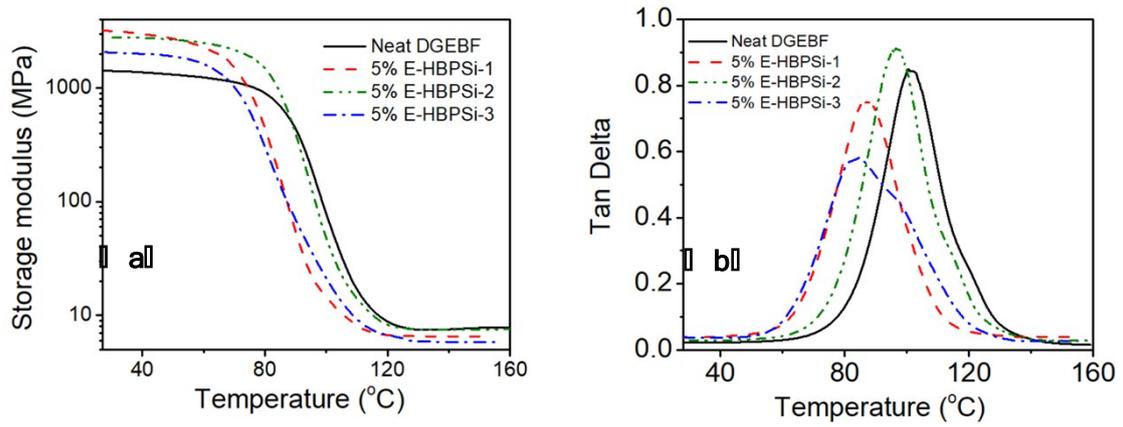


Fig.S6 Variation of storage modulus (a) and $\tan\delta$ (b) with temperature in neat DGEBF , 5% E-HBPSi-1,5% E-HBPSi-2 and 5% E-HBPSi-3.