

Supplementary Information

**Low viscosity and Low Temperature Curing Reactive POSS/Epoxy  
Hybrid Resin with Enhanced toughness and Comprehensive Thermal  
Performance**

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## S1 Synthesis and Characterization of OG-POSS

In this procedure, 200 mL of isopropyl alcohol (IPA) and 15.61 g of a 5% tetramethylammonium hydroxide (TMAH) aqueous solution were combined in a 500 mL three-necked flask and mixed thoroughly. Separately, 30 mL of IPA and 60.38 g (0.255 mol) of KH-560 was measured and successively added to a constant-pressure dropping funnel. The IPA solution of KH-560 was dropwise added to the aforementioned mixture under stirring. This addition was completed over the span of one hour, followed by stirring at room temperature for six hours. Upon completion of the reaction, the solvent IPA was removed by rotary evaporation. Subsequently, 200 mL of toluene was added for dissolution. The mixture was then washed with saturated brine until it was neutral, dried with anhydrous magnesium sulfate for 12 hours, and the toluene was removed by rotary evaporation. The product was dried in a vacuum oven at 70 °C for 24 hours to yield a hydrolyzed intermediate product.

Into a three-necked flask containing a predetermined mass of the hydrolyzed product, 100 mL of toluene and a specified quantity of a 10% tetramethylammonium hydroxide (TMAH) aqueous solution was introduced. The mixture was heated to 108 °C and reacted for 6 hours, with the timer starting at the onset of toluene reflux. After the reaction was complete, the mixture was washed with saturated brine until neutral, dried with anhydrous magnesium sulfate for 12 hours, and the toluene was removed by rotary evaporation. The product was dried in a vacuum oven at 70 °C for 24 hours to yield a pale-yellow viscous liquid, which is OG-POSS, with a yield of 57.50% and an epoxy value of 0.56 mol (100 g)<sup>-1</sup>.

OG-POSS was synthesized utilizing the hydrolytic condensation reaction of KH-560, as evidenced by Fourier Transform Infrared Spectroscopy (FT-IR) and Nuclear Magnetic Resonance (NMR) techniques, as shown in Fig. S1 (a-d). From the FTIR spectrum of OG-POSS (Fig. S1(a)), the absorption peak at 1085 cm<sup>-1</sup> corresponds to the Si-O-C stretching vibration of KH-560. This peak disappears after the reaction, indicating its involvement in the hydrolytic condensation process. The absorption peaks at 1103 cm<sup>-1</sup> and at 910 and 852 cm<sup>-1</sup> represent the Si-O-Si stretching vibrations and the epoxy framework stretching vibrations of OG-POSS, respectively. Additionally, there is no noticeable O-H stretching vibration absorption peak around 3300 cm<sup>-1</sup>, suggesting a comprehensive condensation of the hydrolyzed intermediate. Concurrently, from the <sup>1</sup>H NMR (Fig.

S1(b)) and  $^{13}\text{C}$  NMR (Fig. S1(c)) spectra of OG-POSS, the methoxy groups in KH-560 that undergo hydrolytic condensation can be observed. The hydrogen and carbon atoms chemically shift in the methoxy groups connected to Si at 3.46 ppm and 50.39 ppm, respectively, and these peaks disappear post-reaction. Furthermore, the  $^{29}\text{Si}$  NMR spectrum (Fig. S1(d)) confirms the presence of the Si-O-Si bond in OG-POSS with chemical shifts at 66.73 ppm, 68.54 ppm, and 70.91 ppm, suggesting that OG-POSS comprises a mixture of cage structures  $\text{T}_8$ ,  $\text{T}_{10}$ , and  $\text{T}_{12}$ .

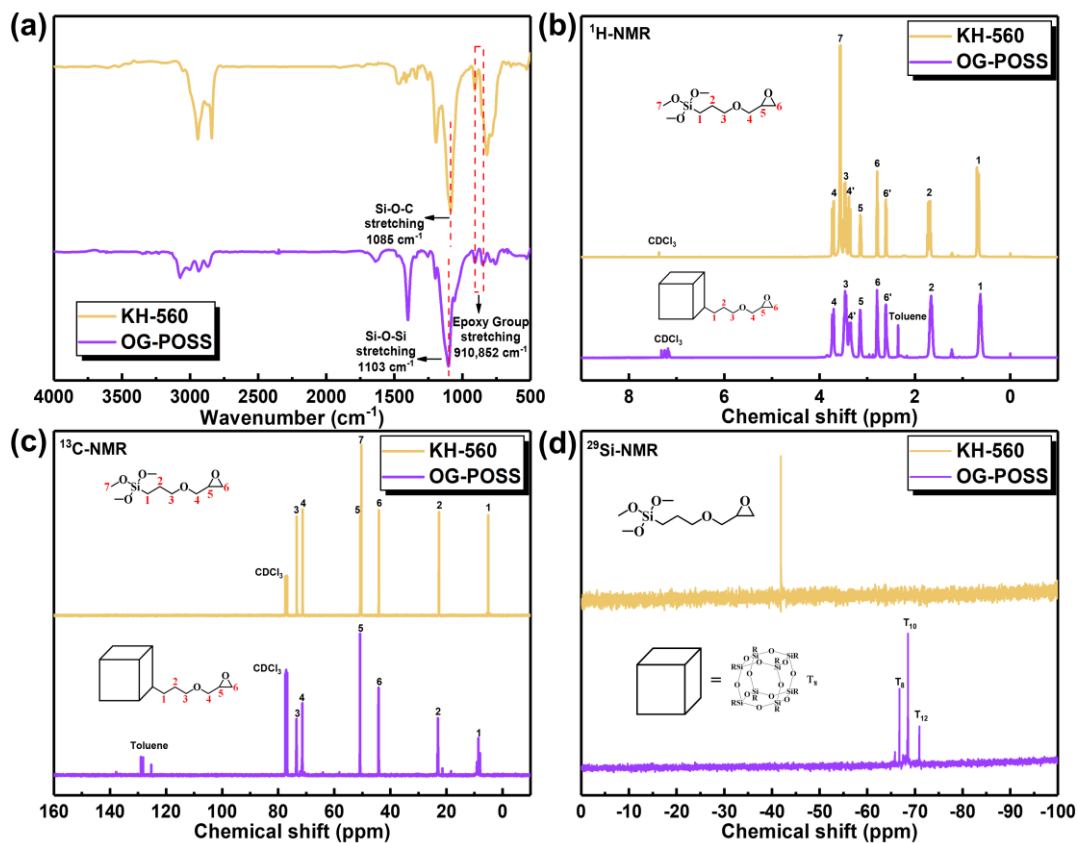


Fig. S1 (a) FTIR spectrum of KH-560 and OG-POSS; (b-d)  $^1\text{H}/^{13}\text{C}/^{29}\text{Si}$  NMR spectrum of KH-560 and OG-POSS.

## S2 Processability and Curing Reactivity of OG-POSS in OPEP hybrid resin

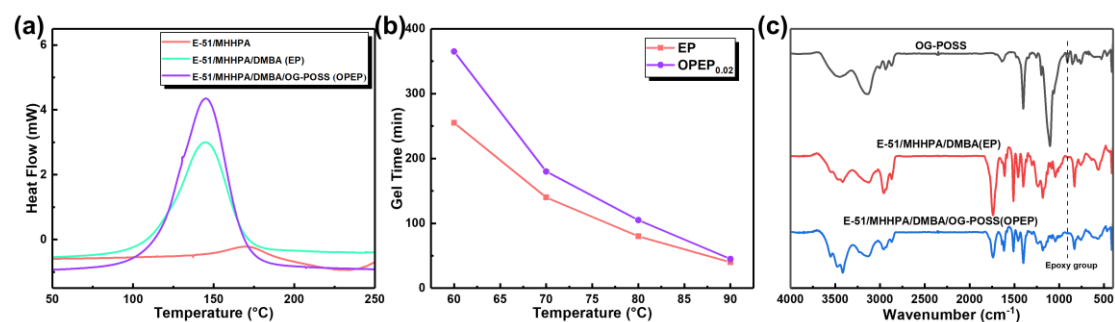


Fig. S2 (a) DSC curves of different systems ( $\beta = 5 \text{ K min}^{-1}$ ); (b) Gel time curves of EP and OPEP<sub>0.02</sub> samples; (c) FTIR spectra of different samples after complete curing.

Table S1 Viscosity data of EP and OPEP<sub>0.02</sub> samples.

Temperature / Time	25 °C / 0 h	70 °C / 1 h	70 °C / 2 h	70 °C / 3 h	70 °C / 4 h	70 °C / 5 h	70 °C / 6 h
Viscosity of EP / (mPa s)	771.82	204.77	255.41	466.21	994.57	2686.79	5857.43
Viscosity of OPEP <sub>0.02</sub> / (mPa s)	751.47	141.18	184.70	407.65	686.79	1972.32	4678.95

(a) The viscosity of E-51 at 25 °C is 12940 mPa s.

(b) Temperature / Time represents the curing time of the sample at a certain temperature.

As shown in Fig. S2(a), the DSC curve of E-51/MHHPA system, without the addition of curing accelerator (DMBA), shows an extremely weak curing exothermic peak at 173 °C, indicating that the curing system has low reactivity and inapparent thermal effect, which the epoxy/anhydride system (without curing accelerator) cannot satisfy the requirements of low-temperature curing. Conversely, an outstanding curing exothermic peak appeared in the DSC curves of EP and OPEP systems, containing curing accelerator, and the initial curing temperature ( $T_i$ ) was less than 100 °C (as shown in Table 1), which provides a theoretical possibility for the low-temperature curing of epoxy resin.

Table S1 and Fig. S2(b) indicate the viscosity data and gel time curves of EP and OPEP<sub>0.02</sub> systems respectively to explore the influence of OG-POSS on the resin molding process. It is obvious that the addition of MHHPA significantly reduces the viscosity of the system (the viscosity of E-51 at 25 °C is 12940 mPa s), which is conducive to its further mixing and reflects the excellent processability of the liquid anhydride curing agent (OG-POSS has little effect on the viscosity of the resin system compared to MHHPA). Regarding gel time, the OPEP system exhibits a

significantly longer gel time at lower temperatures than the pure system. However, this difference diminishes as the temperature increases, with the gel times of the two systems becoming almost identical at 90 °C. This indicates that the addition of OG-POSS does not significantly impact the curing process of the resin.

As shown in Fig. S2(c), after complete curing of the EP and OPEP systems, the stretching vibration peak associated with the epoxy group's skeleton is no longer observable in the FT-IR spectra, which indicates that the reactive OG-POSS containing a multitude of epoxy groups can react with anhydride curing agents, co-curing with the epoxy resin.