Supplementary Material

Flexible Thermal Conductive Al_2O_3@siloxane Composite with Rapid Self-healing Property Based on Carboxyl-amine Dynamic Reversible bonds

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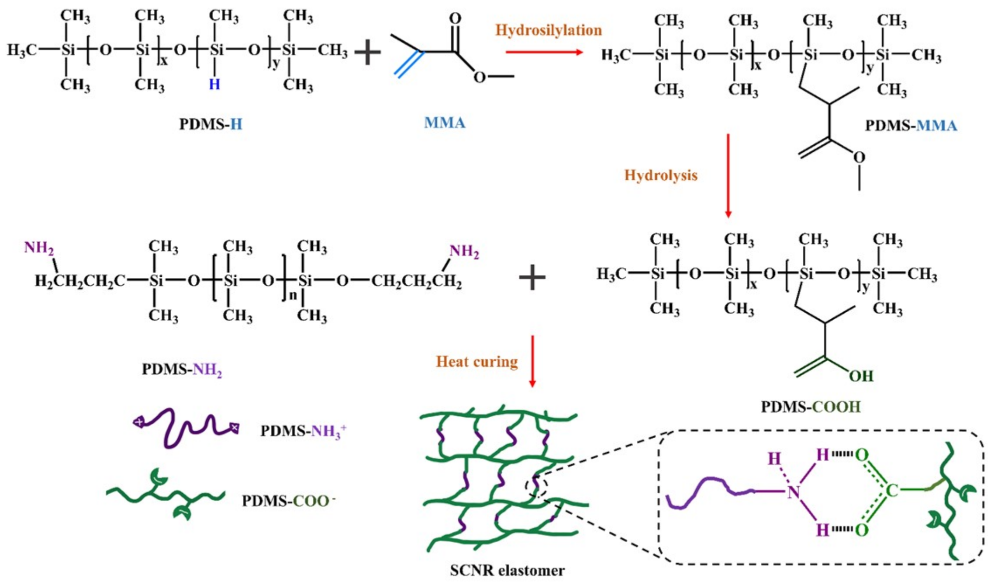
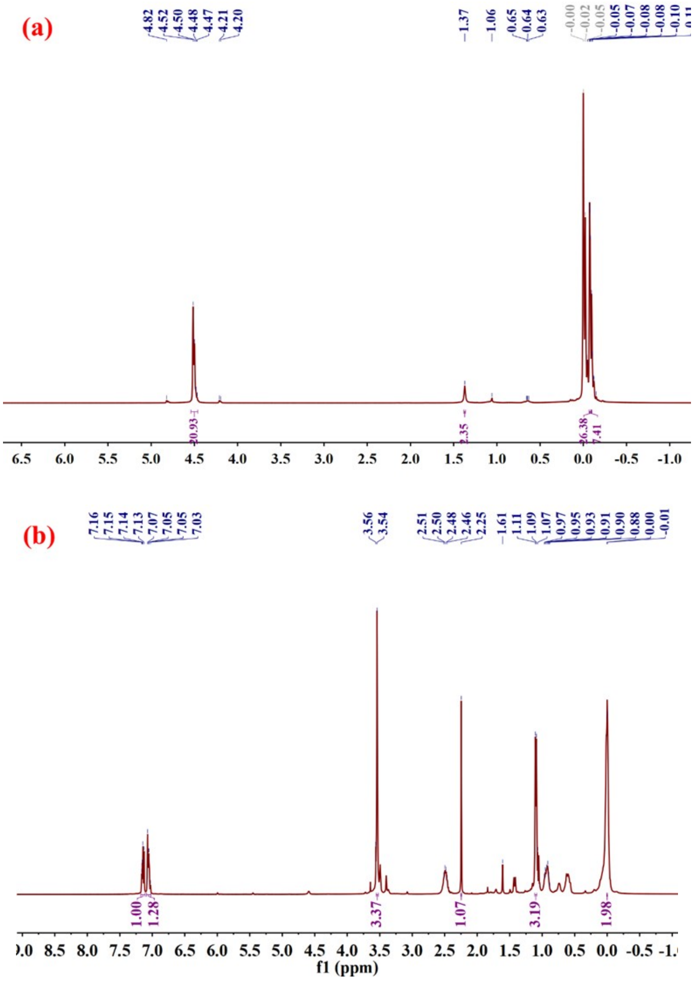


Fig. S1: Preparation process of PDMS-COOH and SCNR



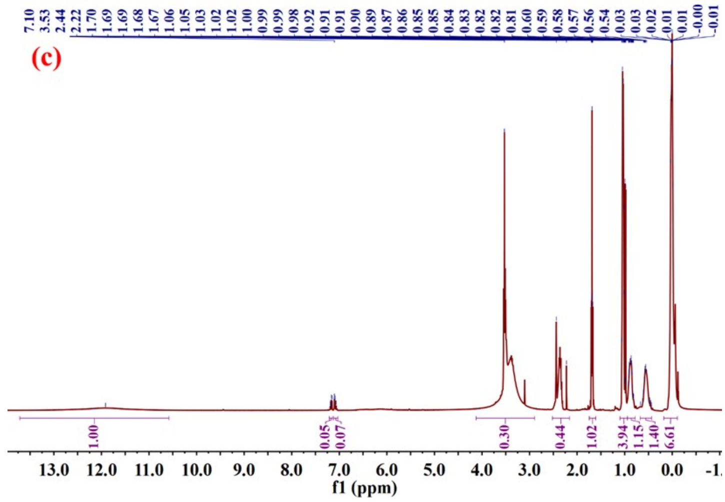


Fig. S2:(a) ^{1}H NMR spectrogram of PDMS-H; (b) PDMS-MMA; (c) PDMS-COOH

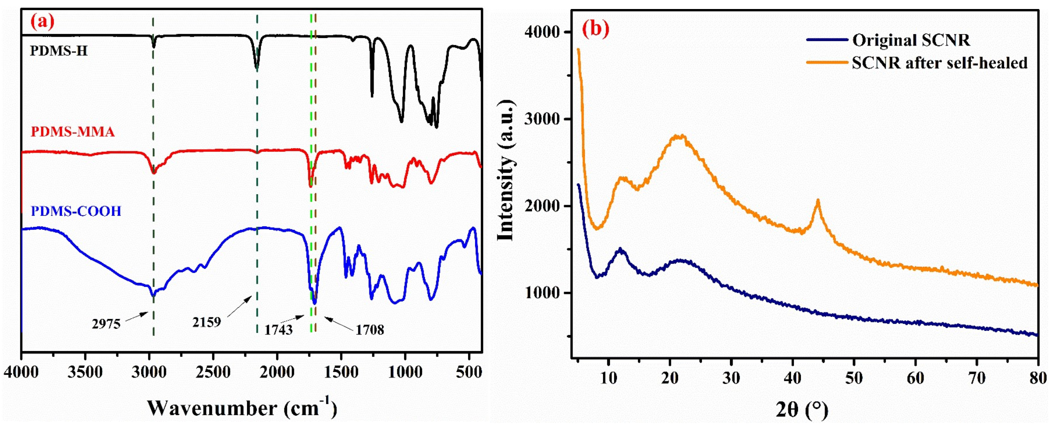


Fig. S3: (a) FTIR spectra of PDMS-H, PDMS-MMA, PDMS-COOH; (b) XRD patterns of the SCNR before and after self-healed

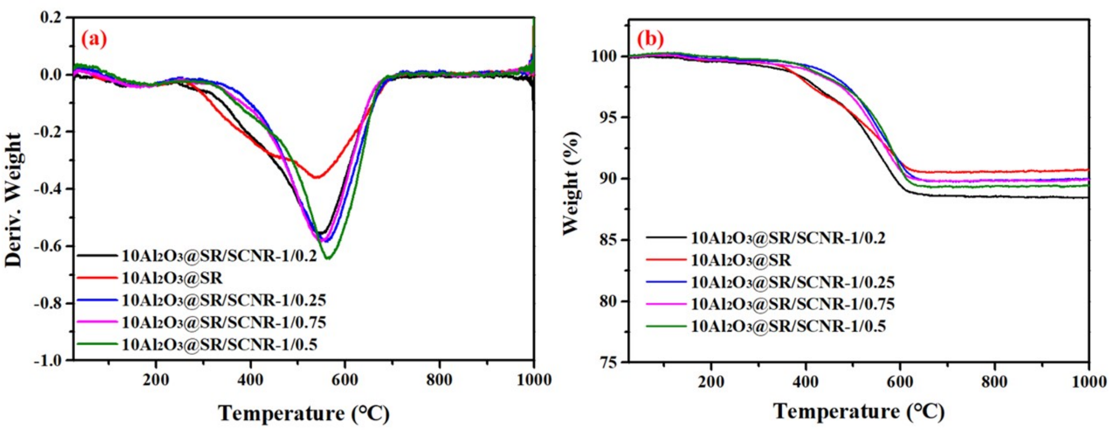
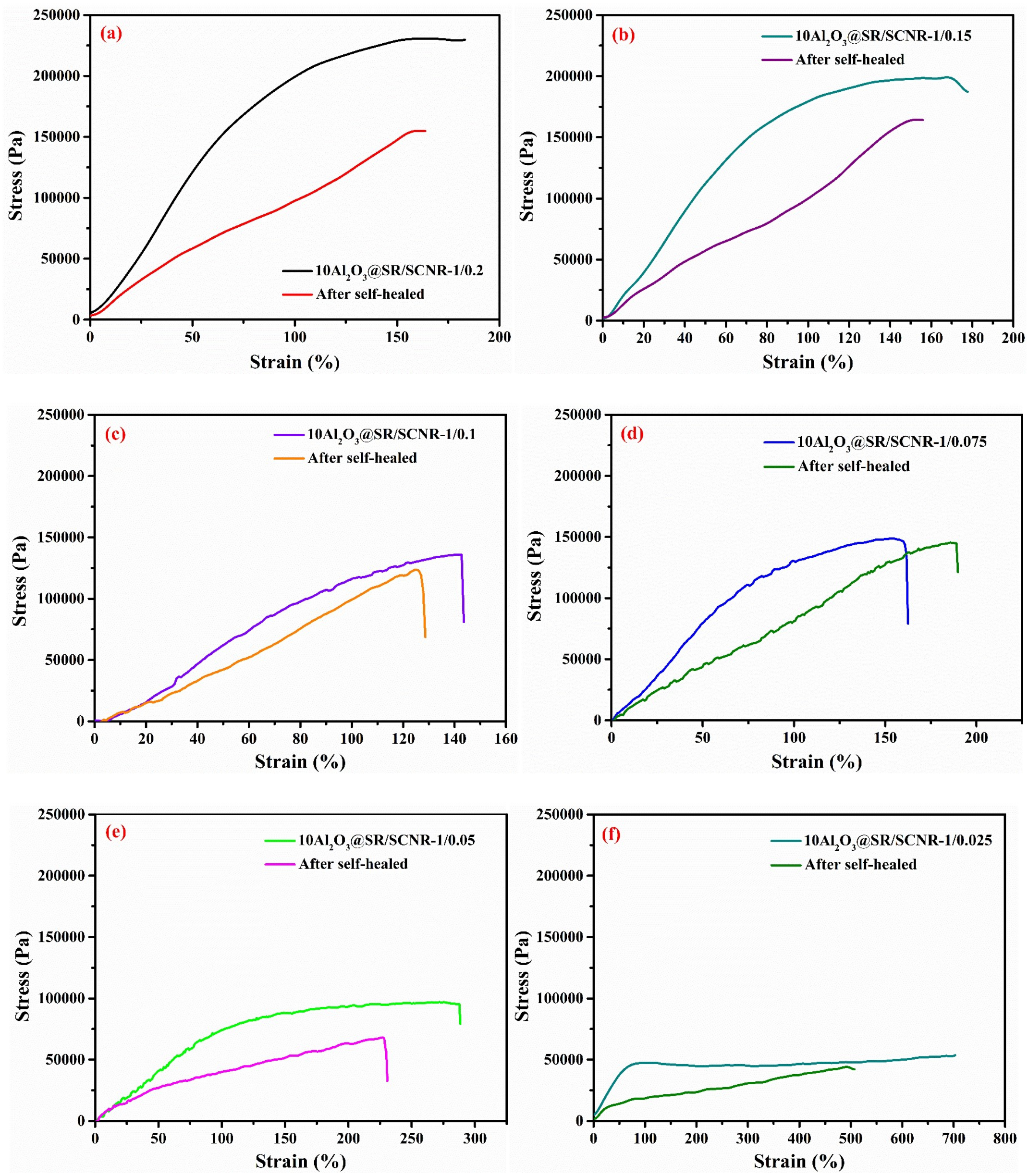


Fig. S4: (a) DTG curves of Al_2O_3@SR and Al_2O_3@SR/SCNR composites; (b) TG curves of Al_2O_3@SR and Al_2O_3@SR/SCNR composites;

Table S1 Date of TG and DTG of Al_2O_3@SR and Al_2O_3@SR/SCNR composites

Sample	$T_{d\ 5\%}$ (°C)	T_d max (°C)	Mass %
10Al_2O_3@SR	507	539	90.73
10Al_2O_3@SR/SCNR-1/0.025	562	569	89.96
10Al_2O_3@SR/SCNR-1/0.05	544	566	89.47
10Al_2O_3@SR/SCNR-1/0.075	530	548	89.92
10Al_2O_3@SR/SCNR-1/0.02	502	548	88.43



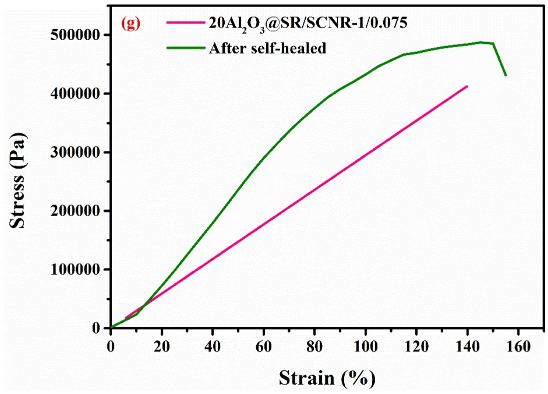


Fig. S5: (a-g) Self-healing properties of the Al_2O_3@SR and Al_2O_3@SR/SCNR composites with different SCNR wt% content

Fig. S6: Self-healing properties of the Al_2O_3@SR and Al_2O_3@SR/SCNR composites under 2 kPa pressure at 90 °C for 3 min

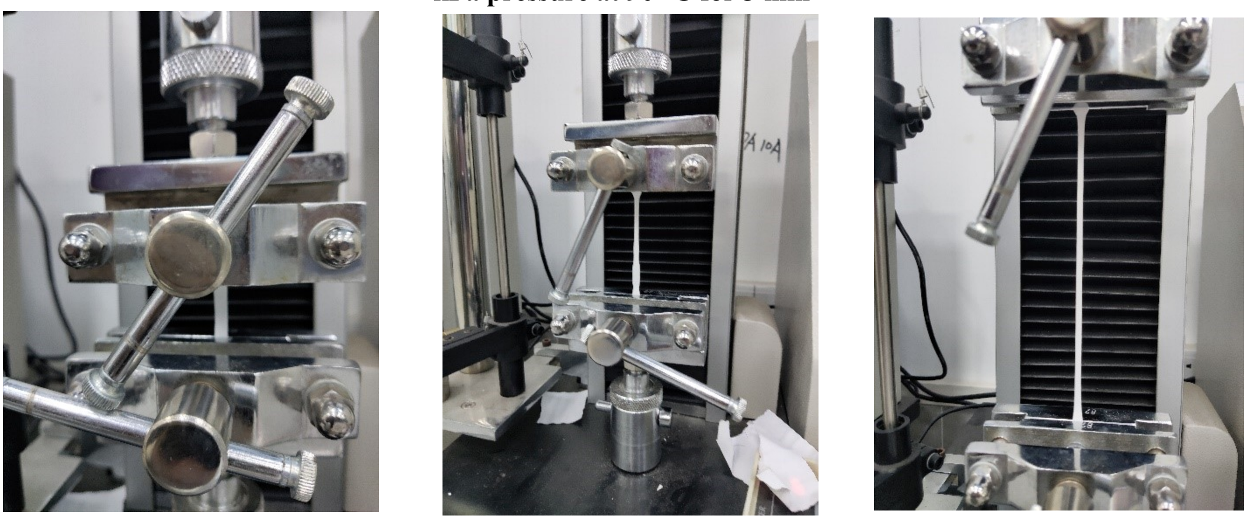


Fig. S7: Al_2O_3@SR/SCNR composites during tensile test

Table S2 A numerical contrast of the thermal conductivity, self-healing efficiency and the time on self-healing of Al_2O_3@SR/SCNR composites with recent works

Composites	Thermal conductivity (W/mK)	Self-healing efficiency (%)	Self-healing time (h)	References and year
Al_2O_3@SR/SCNR	5.8	95.6 %	0.05	This work
silicone/BN	0.8837	97.16	0.25	2020[S1]
EMPI@VACNTs	10.83	90.8	80	2021[S2]
Graphene/PDMS	0.826	78.83	6	2020[S3]
mBN/thiol-epoxy	1.058	85	1	2018[S4]
PDMS-COOH-CG	0.48	84.6	24	2021[S5]
LCEF	1.25	90.6	1	2021[S6]
BN/GO/ENR/PLA	0.36	86	1	2022[S7]
mBN-30/UPy-PDMS-UPy	0.48	84.6	24	2021[S8]

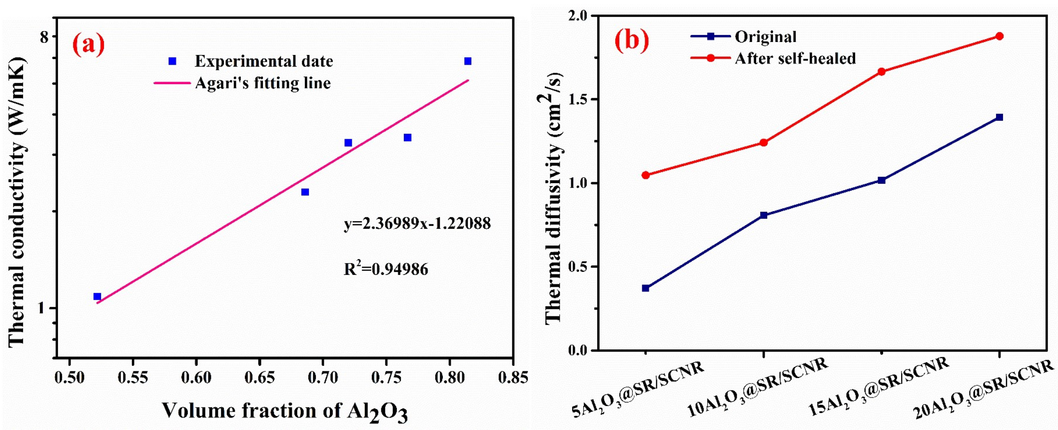


Fig. S8: (a) Experimental thermal conductivity data and the Agari's fitting line; (b) Thermal diffusivity of Al_2O_3@SR/SCNR before and after self-healed

The thermal conductivities of the composites with high filler content can be theoretically analyzed and predicted by classic **Agari's** model and the equation was as follows:

$$\log\lambda_c = V_f C_2 \log\lambda_f + (1-V_f)\log(C_1\lambda_p) \quad \text{..................(1)}$$

where λ_c, λ_f and λ_p are the thermal conductivities of the composite, filler and polymer matrix, respectively. In this work, the thermal conductivities of silicone matrix and Al_2O_3 are 0.2 W/mK and 27.5 W/mK, respectively. V_f is the volume fraction of the filler. The equation (1) can be transformed into equation (2):

$$\log\lambda_c = V_f[C_2 \log\lambda_f - \log(C_1\lambda_p)] + \log(C_1\lambda_p) \quad \text{........ (2)}$$

It can be seen from equation (2) that λ_c is a function of the V_f, and the relationship between λ_c and V_f was shown in. So an equation (3) for Agari's fitting line can be obtained as follows:

$$y = 2.4388x - 0.1.3287 \quad \text{.............................. (3)}$$

Therefore, from equations 2 and 3, the value of C_1 can be calculated firstly ($log(C_1\lambda_p)$ = -0.74). The calculated C_1 was 0.2345, and then C_2 can be calculated to be 0.83.

References:

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[S7] C. Jia, P. Zhang, S.M. Seraji. et al., Effects of BN/GO on the recyclable, healable and thermal conductivity properties of ENR/PLA thermoplastic vulcanizates, Composites. Part A, Applied science and manufacturing. 152 (2022) 106686.

[S8] C. Chen, H. Yu, Y Feng, et al., Polymer composite material with both thermal conduction and self-healing functions, ACTA POLYMERICA SINICA. 52 (3) (2021) 272-280.