Build a Rigid Flexible Graphene/Silicone Interface by Embedding SiO_2 for Adhesive Application

Hualan Wang, [a,b,*] *Cheng Yang,* [a] *Risheng Liu,* [a] *Kai Gong,* [b,*] *Qingli Hao,* [c] *Xin Wang,* [c] *Jirong Wu* [a] *Guodong Zhang,* [a] *Yingqian Hu,* [a] *Jianxiong Jiang,* [a,*]

[a.] Key Laboratory of Organosilicon Chemistry and Material Technology, Ministry of Education, Hangzhou Normal University, No. 2318, Yu Hangtang Rd, Hangzhou, 311121, China.

[b.] School of Pharmaceutical Science, Jiangnan University, No.1800, Lihu, Wuxi, 214122, China.

[c.] Key Laboratory of Soft Chemistry and Functional Materials, Ministry of Education, Nanjing University of Science and Technology, No.200, Xiao Lingwei, Nanjing, 210094, China.

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[*] Corresponding authors. Tel/Fax: +86 571 28868903.

Email addresses: hualanwang@163.com (Hualan Wang), kingong222@163.com (Kai Gong), fgeorge@21cn.com (Jianxiong Jiang)

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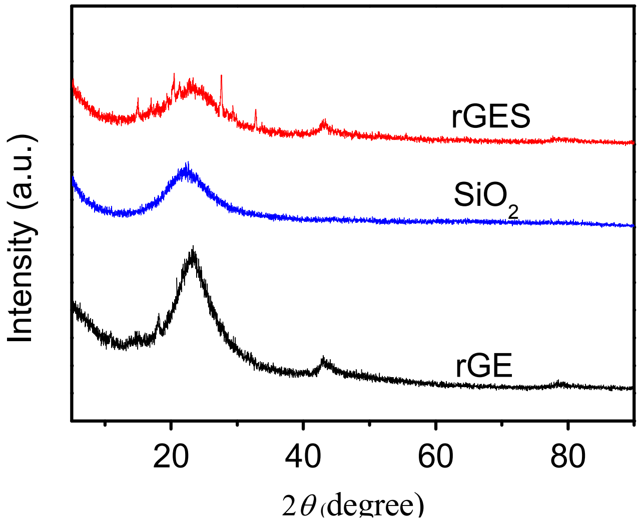


Figure S1. XRD of rGE, SiO_2 and rGES-2.

Figure S1 gives the XRD patterns of rGE, SiO_2 and rGES. The rGE shows a broad peak at 23.3°, the peak of SiO_2 was found at 22.6°, and rGES shows a comprehensive peak of rGE and SiO_2 at 22.9°, signifying a certain integration of SiO_2 and rGE. After integrated with rGE in rGES composites, SiO_2 shows some narrow and small new peaks in the range of 15–35°, indicating that the regularity of SiO_2 is enhanced. A more regular growth of SiO_2 is promoted by the existence of rGE. Moreover, no obvious graphite peak of 26.6°can be found in rGES, indicating the agglomeration is inhibited well by SiO_2 nanoparticles and a good dispersion of rGE in rGES.

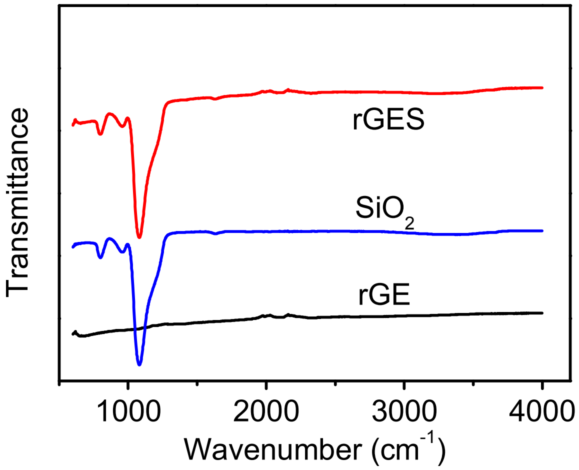
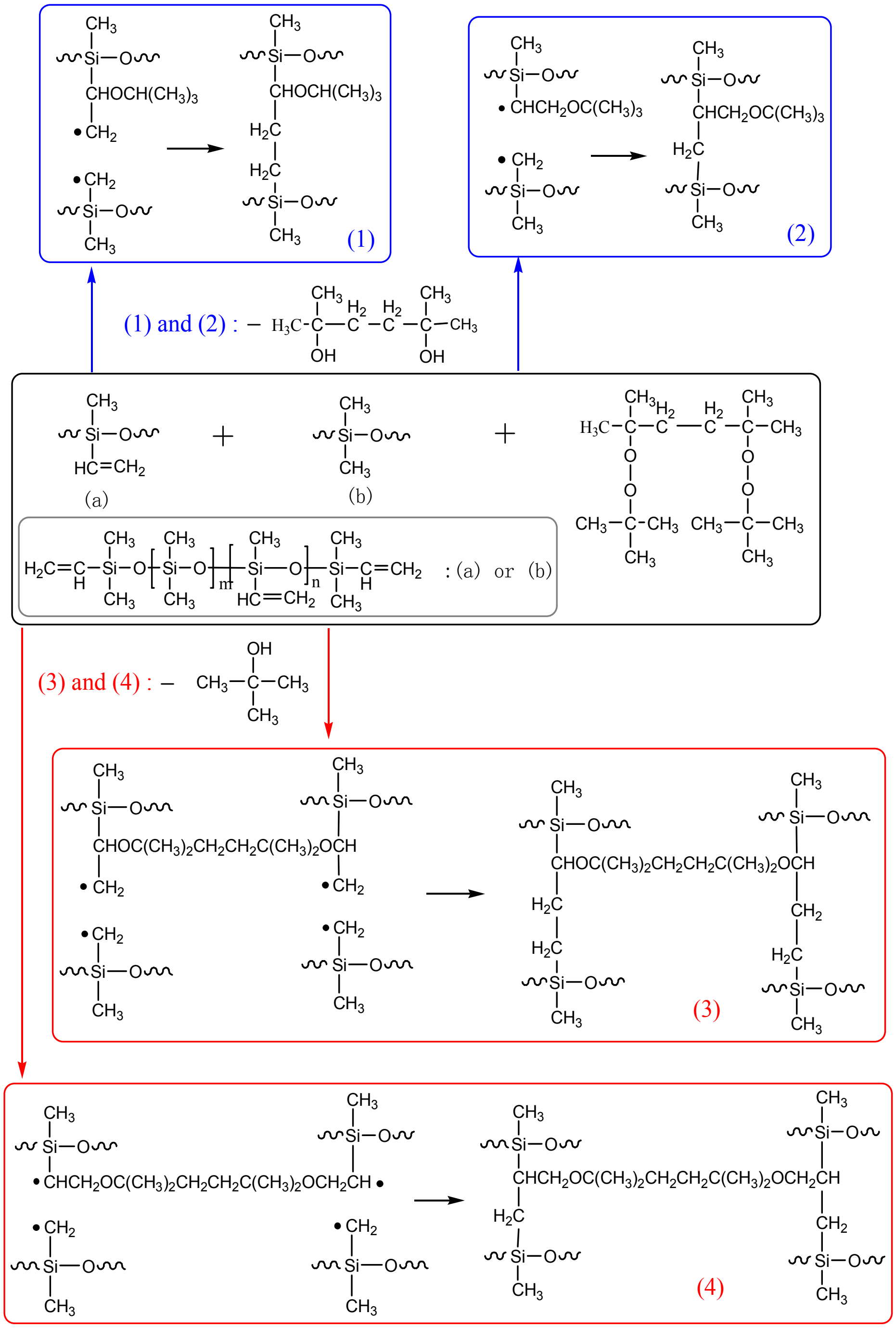


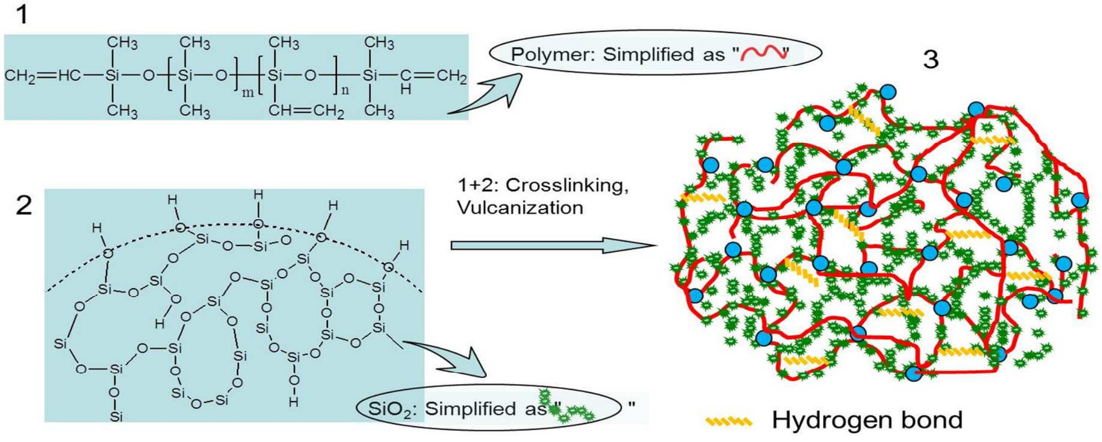
Figure S2. FTIR spectra of rGE, SiO_2 and rGES-2.

In order to further reveal the interaction between rGE and SiO_2, the FT-IR spectra of GO, rGE and rGES are compared in Figure S2. For the case of SiO_2, the peak at about 790 cm^{1} could be assigned to the typical vibration mode of framework Si–O–Si, 1064 cm^{1} is the asymmetric stretching vibration of Si O Si, and this peak also emerges in rGES just as shown in Figure S2. A weak band located at 950 cm^{1} for SiO_2 was mainly attributed to the stretching vibrations of Si–OH groups at defect sites. Most of the characteristic peaks of SiO_2 can be found in rGES, indicating that the components are well combined and successfully constitute the final composite.



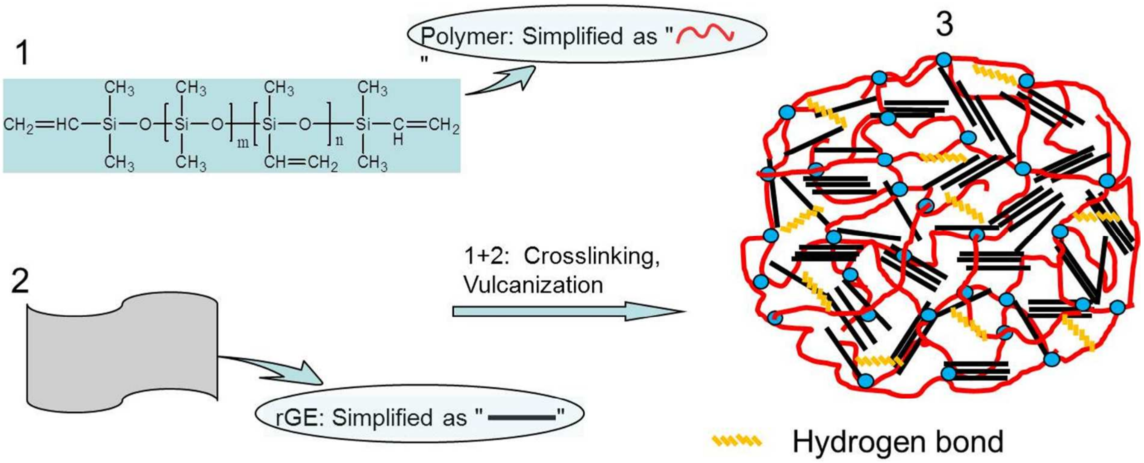
Scheme S1. Cross-linking reactions of polysiloxane.

Most unfilled silicone networks display extremely poor mechanical properties except self-enhanced matrixes. To understand the enhancing mechanism of rGES in methyl vinyl polysiloxane matrix, it is necessary to figure out the reinforcement principle of unfilled and filled polysiloxane by individual SiO_2 and rGE. Methyl vinyl polysiloxane is such a kind of silicone whose backbone consists of repeated units of —Si—O—,[S1] and each silicon atom is connected with methyl or vinyl. The cross-linking pots situate at the vinyl groups of one polysiloxane molecule and the methyl groups of another polysiloxane molecule.[S2, S3] For the peroxide curing agent used in this experiment, there are two possible dealcoholation processes,[S4] corresponding to four crosslinked forms or products, just as shown in Scheme S1. Therefore, the stress transmission and reinforcing of unfilled polysiloxane is realized mainly through vulcanizing agent, which is clearly insufficient to provide ideal mechanical strength as the vinyl content is only 0.16% mol for the silicone used in our experiments. In order to improve the mechanical properties of such kind of non-self-reinforced polysiloxane molecules, suitable reinforcing filler is essential to help form stronger network structure.



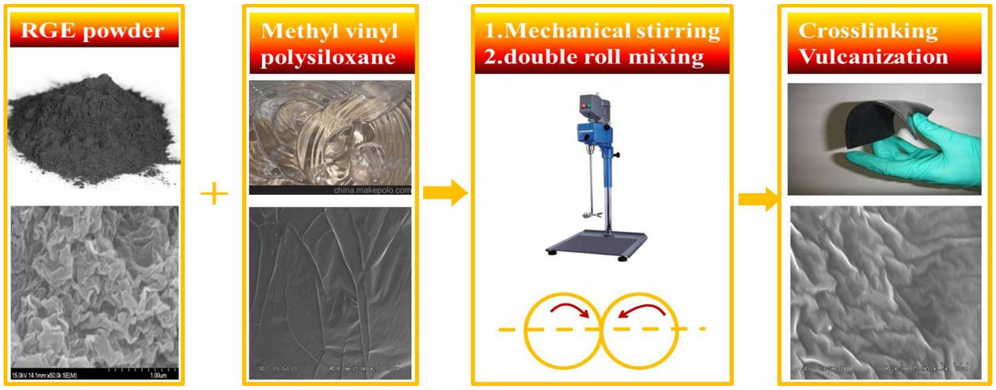
Scheme S2. Schematic interaction between polysiloxane and SiO_2 in a cross-linked system.

The most frequently used filler for silicone matrixes is precipitated or fumed SiO_2, which helps to form more developed network structures and stronger interactions in vulcanized polysiloxane matrixes.[S5, S6] Abundant hydrogen bonds between SiO_2 nanoparticles and polysiloxane form due to excellent compatibility and dispersion.[S7, S8] Besides the interaction provided by vulcanized agents, another strong interaction among different polysiloxane moleculars is achieved through bridging of SiO_2. These two modes, stable and more intertwined together, help show more effective interfacial stress transfer[S9, S10] and increased mechanical performances for polysiloxane matrix than a single crosslinking mode (Scheme S2).



Scheme S3. Schematic interaction between polysiloxane and rGE in the SR matrix.

However, rGE is not as compatible with polysiloxane as SiO_2, and the trend of graphitization leads to a large number of multilayer dispersion of rGE in polysiloxane (Scheme S3). Thicker rGE causes higher rigidity and harder blend with flexible polysiloxane due to lack of buffer or transition at the incompatible interface. As a result, the extraordinary performances of rGE are not effectively elaborated, and the rGE/polysiloxane interface is easily out of control or failure under external physical and chemical changes. One way to solve the problem is to overcome the stack of rGE and improve the compatibility with polysiloxane. Chemical modification and nano compounding are demonstrated able to prevent the rGE layers from stacking.[S11, S12] Since SiO_2 performs well in filled silicone matrixes, we would like to try the nano modification of rGE with nanoscale SiO_2.



Scheme S4 The blending processes of rGES/SR composites via green solvent-free process.

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