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Supporting Information

Bioinspired Highly Electrically Conductive Graphene-Epoxy Layered Composites

Peng Ming,[a†] *Yuanyuan Zhang,*[a†] *Jianwen Bao,*[b] *Gang Liu,*[b] *Zhou Li,*[c] *Lei Jiang*[a] *and Qunfeng Cheng*[a*]

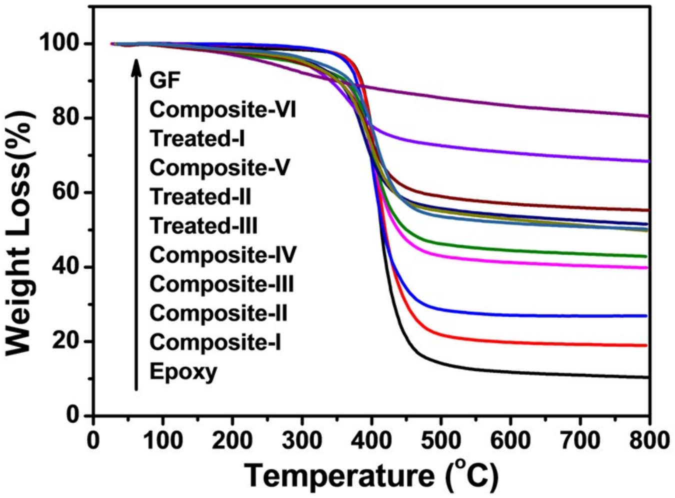

Figure S1. TGA curves of pure GF, epoxy resin, and bioinspired graphene-epoxy composites under N_2 with raising rate of temperature 20 °C/min.

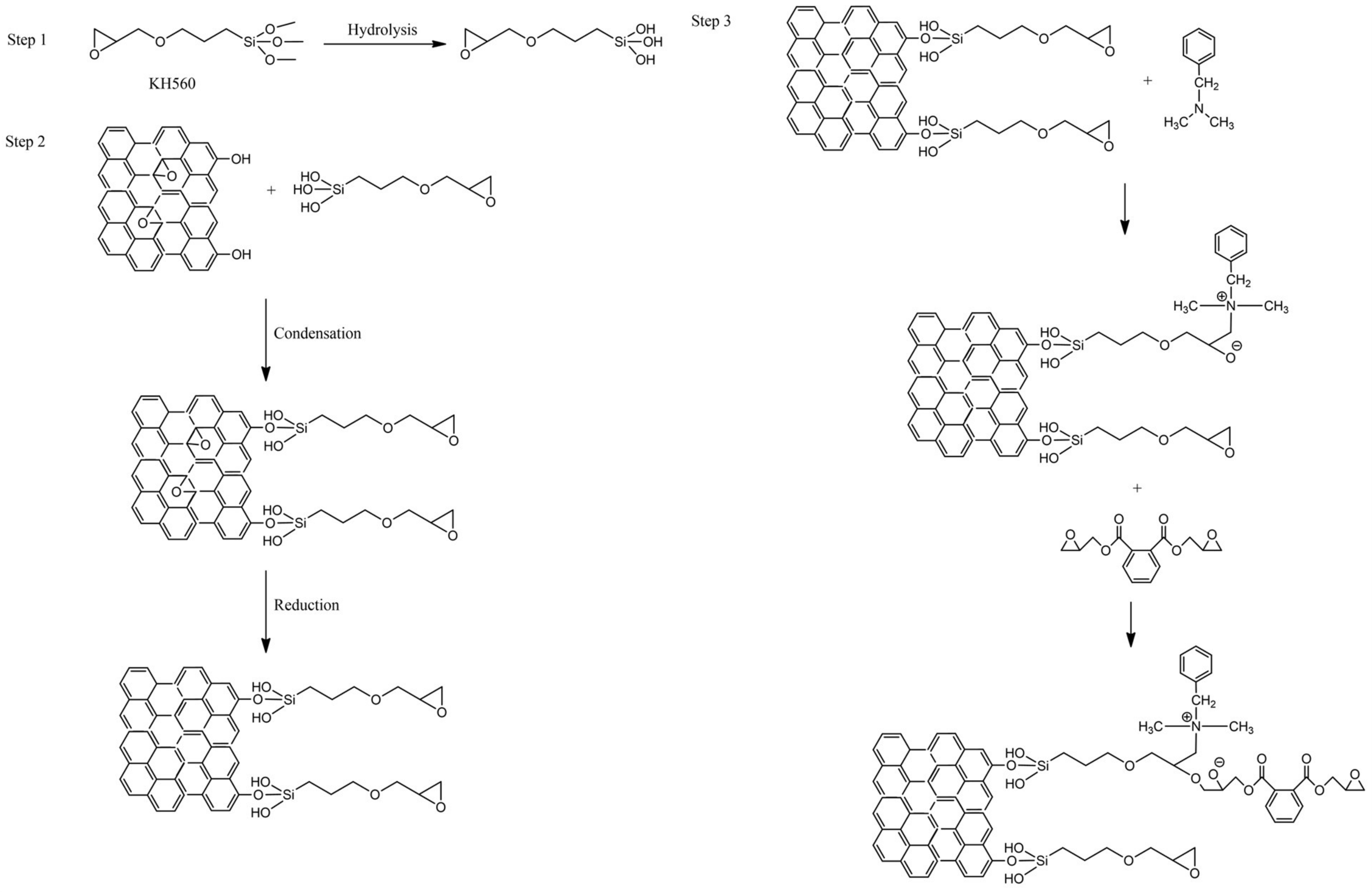

Figure S2. The Proposed cross-linking mechanism between (3-Glycidyloxypropyl)trimethoxysilane (KH-560) and epoxy resin system. Step 1: The KH-560 is hydrolyzed into the (3-Glycidyloxypropyl)trimethoxysilanol. Step 2: The graphene oxide reacts with (3-Glycidyloxypropyl)trimethoxysilanol through condensation of water into KH-560 grafted GO (KH-560-GO). Then the KH-560-GO is reduced with $N_2H_4 \cdot H_2O$ to remove the other groups on GO surface. Step 3: the epoxide groups on the KH-560-GF are then attacked by benzyldimethylamine (BDMA), yielding zwitterions that contain a quaternary nitrogen cation and an active anion. This type of ester is functionalized as the initiator of the chainwise polymerization, and then reacts with epoxy resin of glycidyl ester, leading to formation of an alkoxide species. Subsequently, the formed alkoxide species attack the BDMA again. It should be noted that the reaction in step 3 can happen at both ends of glycidyl ester with the KH-560-GF, eventually resulting in formation of graphene-epoxy composite having chemical cross-linking bonds.

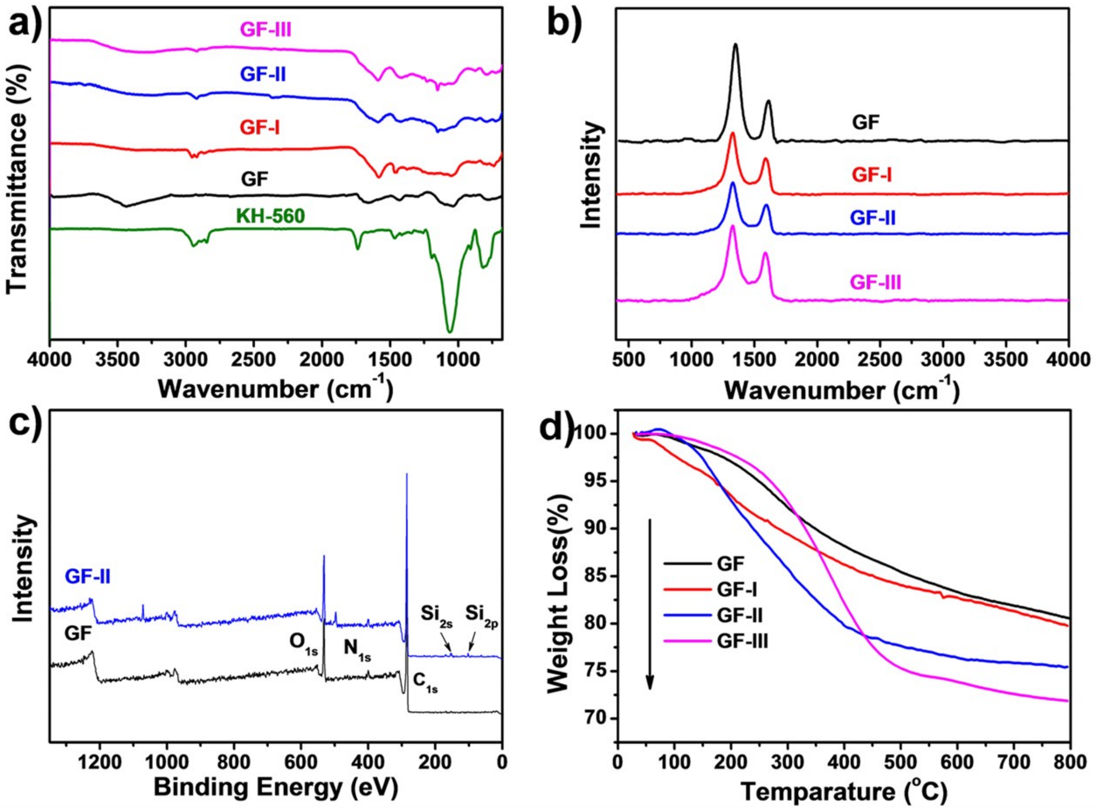

Figure S3. (a) FTIR was conducted to verify the grafting reaction between GF and KH-560. The absorption peaks vibration around 2800-2900 cm^{-1} of GF-I, GF-II, and GF-III corresponds to stretching vibration of -CH$_2$- and -CH-, which demonstrates that the silane coupling agent was successfully grafted on the surface of GF. (b) Raman spectrum was also conducted to confirm the grafting reaction between GF and KH-560. The I_D/I_G ratio was decreased from 3.4 for the GF to 2.7 for GF-I, 2.4 for GF-II and 2.3 for GF-III, respectively. (c) XPS results show that after grafting KH-560, the C$_{1s}$ and N$_{1s}$ peaks decrease 4.4% and 12.5%, respectively. The O$_{1s}$ peak increases 12.8%, because of the contribution of oxygen element in KH-560. Meanwhile, the appearance of silicon element peak on GF-II further confirms that KH-560 was successfully grafted to the surface of GF. (d) TGA results show that the weight loss of GF-I, GF-II, GF-III are 20.2%, 24.5%, 28.1% respectively, which is higher than the GF with 19.46% weight loss. This is direct evidence of the successful grafting KH-560 on the surface of GF.

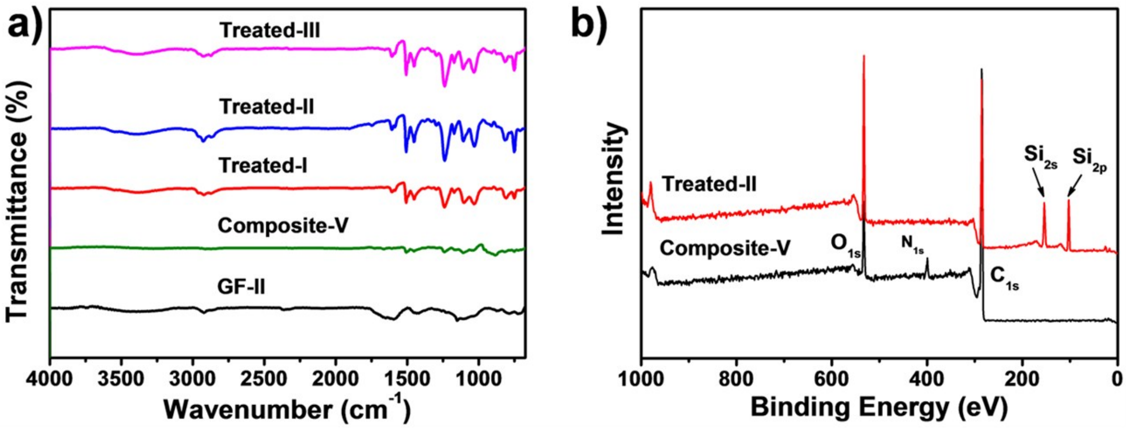

Figure S4. (a) The FTIR comparison of GF-II, Composites-V, Treated-I, Treated-II, and Treated-III. (b) The XPS spectrum of Composite-V and Treated-II.

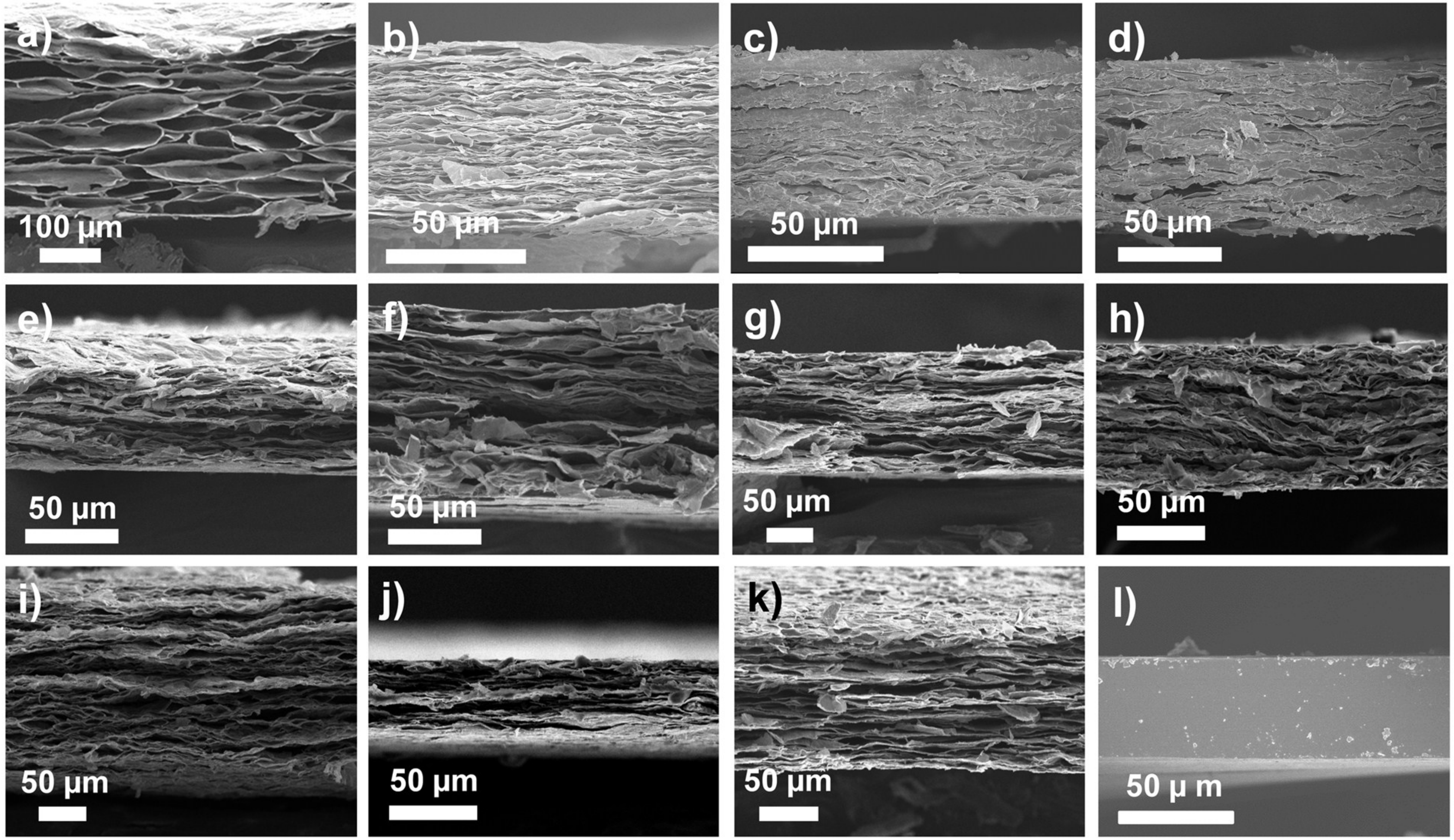

Figure S5. Cross-section surface morphology: (a) GF, (b) control GF, (c) Composite-I, (d) Composite-II, (e) Composite-III, (f) Composite-IV, (g) Composite-V, (h) Composite-VI, (i) Treated-I, (j) Treated-II, (k) Treated-III and (I) Epoxy resin, respectively.

Table S1. The mechanical properties of the pure GF, control GF, epoxy resin and bioinspired graphene-epoxy composites with differen graphene loadings.

Sample	**Stress(MPa)**	**Modulus(GPa)**
GF	2.9 ± 0.5	0.04 ± 0.01
Control GF	19.2 ± 1.2	0.6 ± 0.1
Epoxy	67.5 ± 4.9	2.6 ± 0.5
Composite-I	10.6 ± 0.5	1.2 ± 0.3
Composite-II	14.1 ± 1.6	1.3 ± 0.4
Composite-II	41.1 ± 4.3	4.6 ± 0.1
Composite-IV	48.1 ± 1.4	3.8 ± 0.6
Composite-V	55.1 ± 3.9	5.1 ± 0.6
Composite-VI	33.4 ± 2.6	2.4 ± 0.5
Treated I	55.4 ± 5.4	3.8 ± 0.8
Treated II	69.6 ± 6.3	5.5 ± 0.4
Treated III	44.4 ± 2.8	4.3 ± 0.4

Table S2. Concentrations of various elements on the GF and GF-II

Sample	**Concentrations of various elements (%)**			
	C	**O**	**N**	**Si**
GF	85.50	11.13	2.88	--
GF-II	81.71	12.56	2.52	1.51