Electronic Supplementary Material (ESI) for Materials Horizons.

Electronic Supporting Information

Interlayer Crosslinking to Conquer the Stress Relaxation of Graphene Laminated Materials

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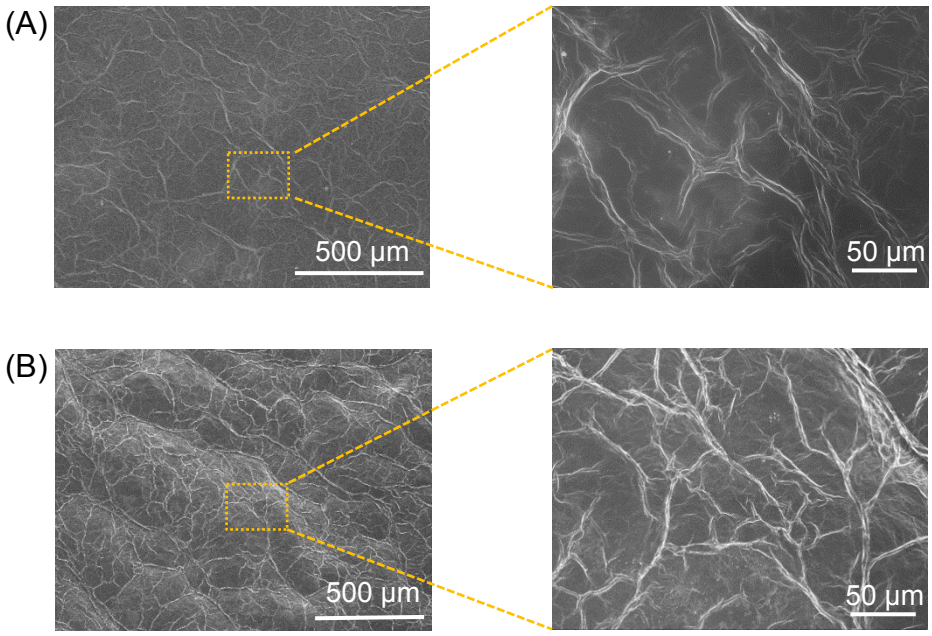

Fig. S1 Surface morphology of (A) GO and (B) RG papers by SEM.

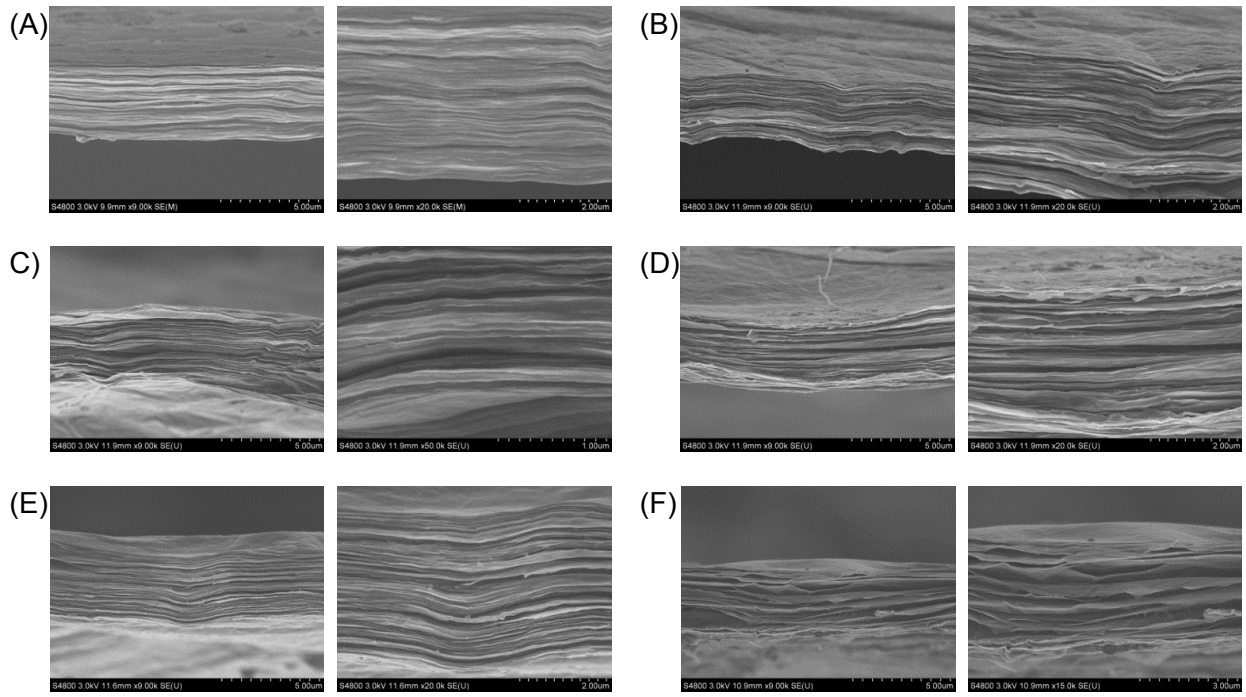

Fig. S2 Cross section of graphene papers by SEM. (A) GO, (B) RG, (C) RG-Mg, (D) RG-Ca, (E) RG-Fe, and (F) RG-S papers.

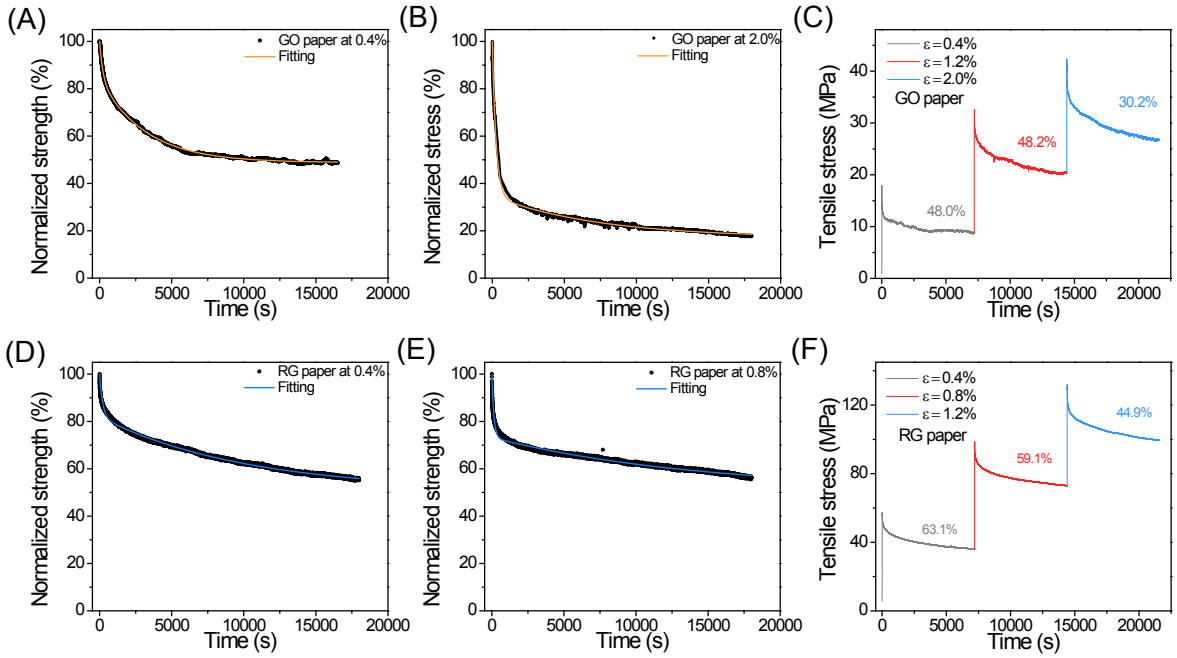

Fig. S3 Blade cast GO and RG papers at different relaxation strains. (A) and (B) are GO paper at strains of 0.4% and 2.0% for 5 h, respectively. (C) GO paper is relaxed at strain of 0.4%, 1.2% and 2.0%, gradually. Each strain is relaxed for 2 h. (D) and (E) are RG paper at strain of 0.4% and 0.8% for 5 h, respectively. (F) RG paper is relaxed at strain of 0.4%, 0.8% and 1.2%, gradually. Each strain is relaxed for 2 h.

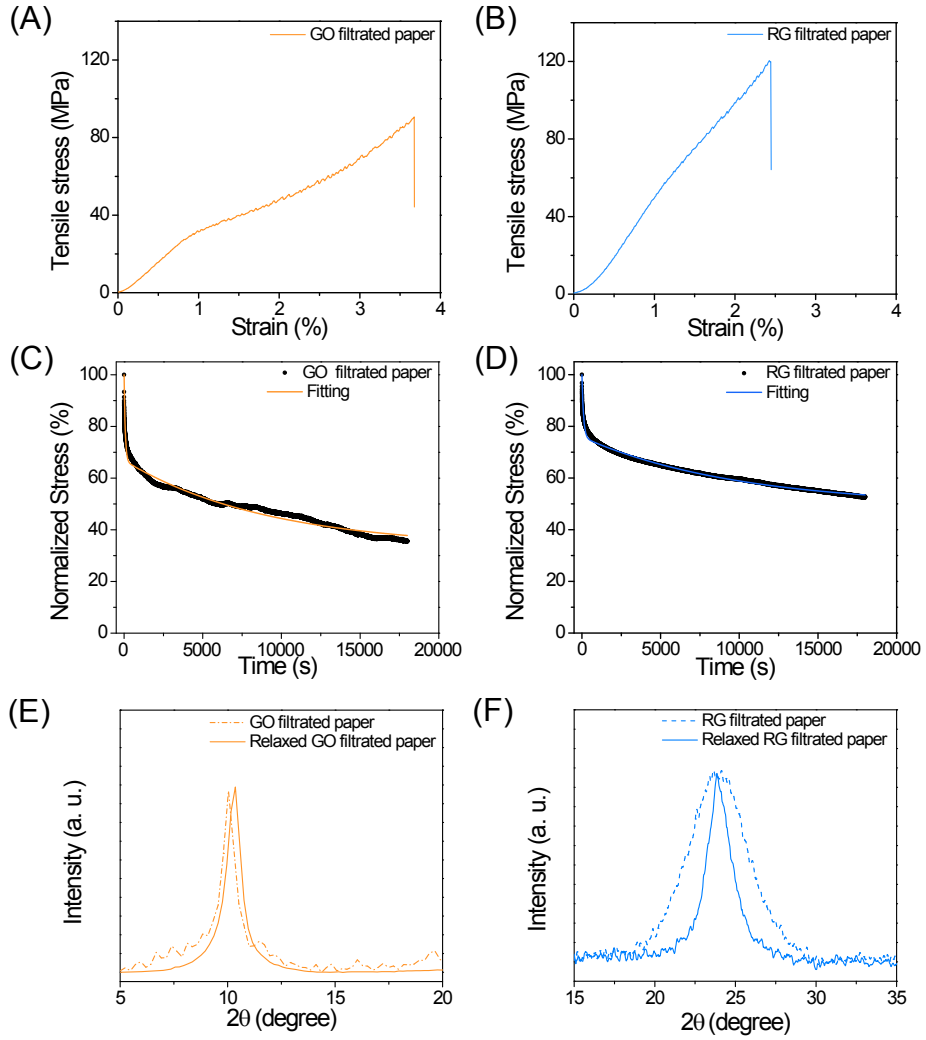

Fig. S4 (A, B) Tensile strength, (C, D) stress relaxation curves and (E, F) XRD spectra of filtrated GO and RG papers. Left column (A, C, E) is filtrated GO paper and right one (B, D, F) is filtrated RG paper.

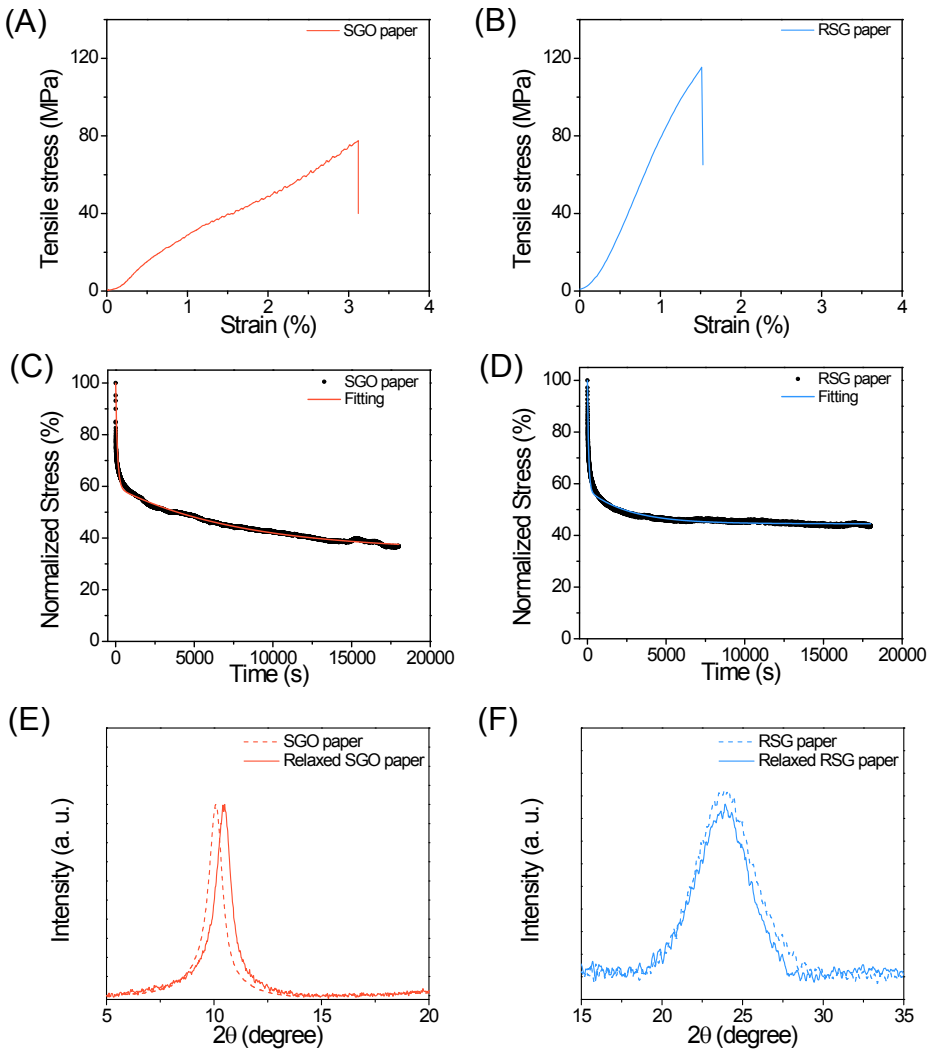

Fig. S5 (A, B) Tensile strength, (C, D) stress relaxation curves and (E, F) XRD spectra of SGO and SRG papers. Left column (A, C, E) is SGO paper and right one (B, D, F) is SRG paper.

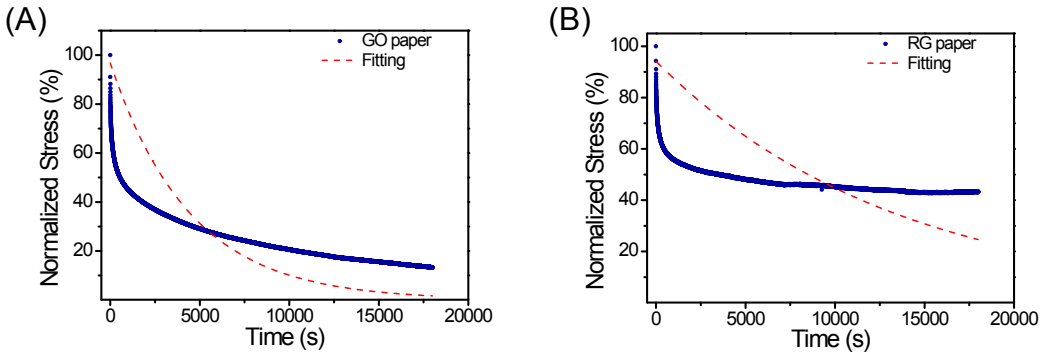

Fig. S6 Stress relaxation curves fitted by simple Maxwell model. (A) GO and (B) RG papers.

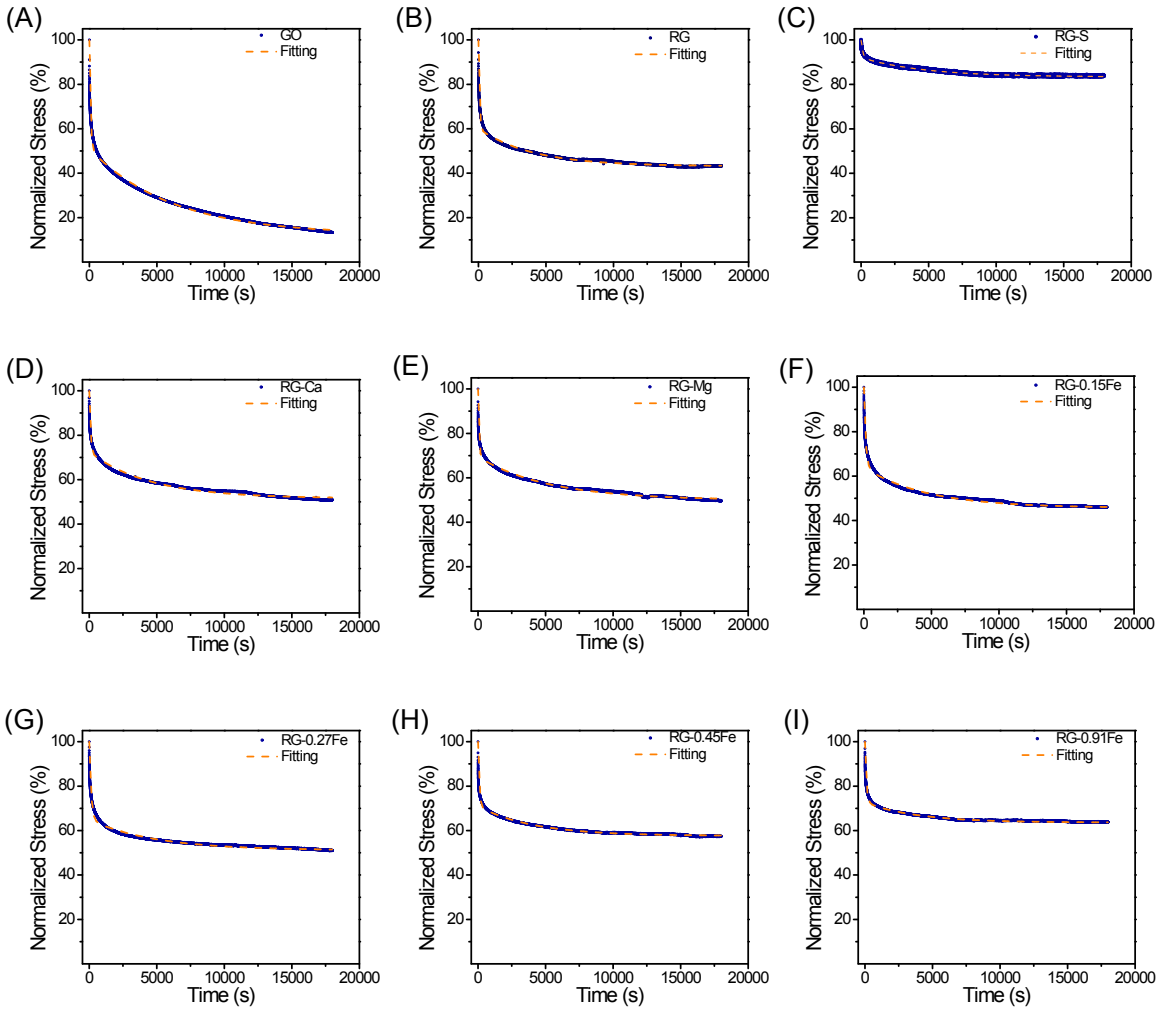

Fig. S7 Fitting curves of stress relaxation data to the ternary Maxwell-Wiechert model. (A) GO, (A) RG, (C) RG-S, (D) RG-Ca, (E) RG-Mg, (F) RG-0.15Fe, (G) RG-0.27Fe, (H) RG-0.45Fe and (I) RG-0.91Fe papers.

Table S1 Fit parameters for stress relaxation data by eqn 3 (main text).

Paper types	A_∞ (%) (mean± s.d.)	A_1 (%) (mean± s.d.)	A_2 (%) (mean± s.d.)	τ_1 (s) (mean± s.d.)	τ_2 (s) (mean± s.d.)	Adj. R^2
GO(1.2%)	12.43±0.032	48.85±0.042	38.66±0.033	81.7±0.40	6061±16.5	0.9996
GO-0.4%	48.81±0.085	15.67±0.037	35.51±0.033	226.3±1.48	3580±24.4	0.9992
GO-2.0%	17.01±0.058	65.20±0.065	17.75±0.048	235.1±1.10	7380±76.0	0.9992
RG(1.2%)	43.15±0.023	39.90±0.032	16.92±0.028	67.0±0.35	4018±17.7	0.9995
RG-0.8%	50.69±0.100	26.78±0.022	22.40±0.087	137.3±0.68	12350±116	0.9993
RG-0.4%	50.79±0.043	18.01±0.021	31.27±0.032	213.8±1.13	10037±32.3	0.9998
RG-Mg	49.99±0.019	30.55±0.030	19.44±0.026	67.8±0.48	5273±19.8	0.9993
RG-Ca	46.97±0.021	26.06±0.027	26.95±0.026	94.6±0.66	4911±20.7	0.9995
RG-0.15Fe	45.71±0.018	36.06±0.031	18.20±0.031	129.76±0.61	4669±10.1	0.9994
RG-0.27Fe	51.13±0.013	34.99±0.031	13.85±0.030	134.39±0.63	4860±13.6	0.9993
RG-0.45Fe	57.62±0.016	29.01±0.021	13.33±0.021	86.17±0.91	4107±8.8	0.9995
RG-0.91Fe	63.61±0.009	27.27±0.019	9.10±0.019	102.50±0.79	3883±11.2	0.9994
RG-S	83.34±0.008	7.85±0.021	8.72±0.009	120.9±0.79	4713±14.7	0.9982
Filtrated GO	33.10±0.025	33.12±0.034	33.76±0.051	73.3±0.85	9109±48.2	0.9989
Filtrated RG	49.13±0.049	24.11±0.023	26.75±0.039	102.2±0.65	9900±41.3	0.9992
SGO	35.10±0.054	40.21±0.038	24.65±0.044	113.57±0.91	7832±45.6	0.9990

RSG	44.51±0.015	42.36±0.029	13.11±0.032	76.49±0.39	2648±36.1	0.9982

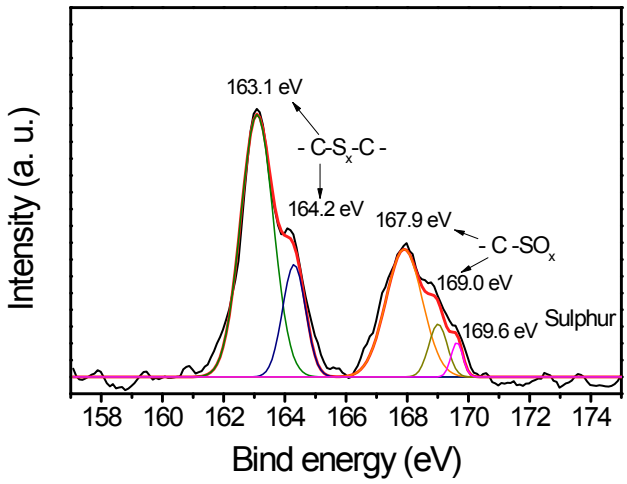

Fig. S8 XPS data of RG-S paper.

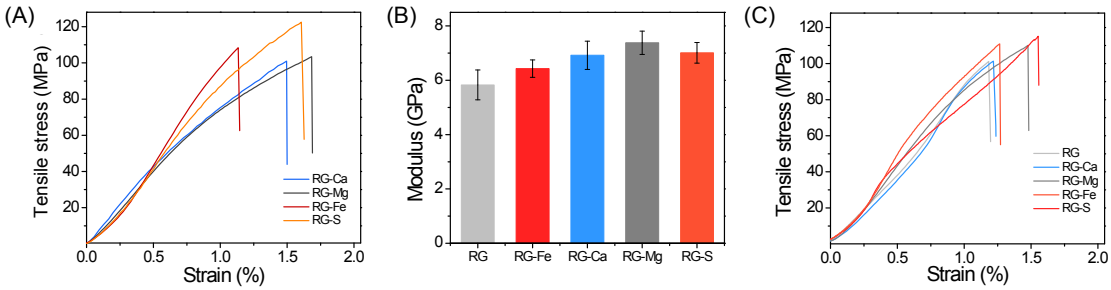

Fig. S9 (A) Tensile tests and (B) modulus of crosslinked RG papers without relaxation. (C) Tensile tests of crosslinked RG papers after relaxation process.

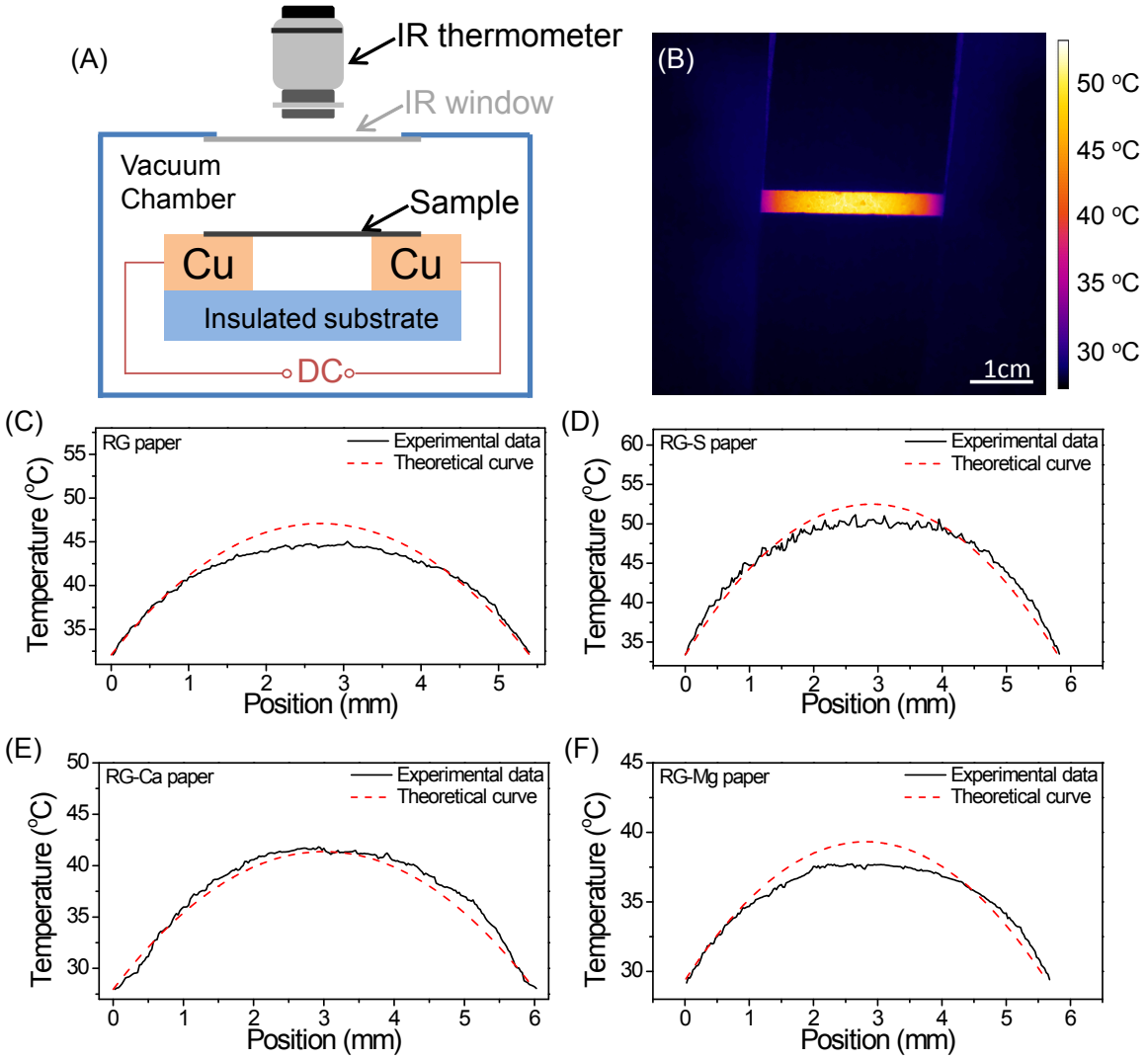

Fig. S10 Thermal conductivity of RG papers with or without crosslinking. (A) Schematic sketch of thermal conductivity test. (B) IR image of RG-S paper at 1V. (C-F) Temperature distributions of RG and crosslinked RG papers at 1V.

Table S2 Experimental data of thermal conductivity.

Sample type	U (V)	I (mA)	ΔT (°C)	K ($W \cdot m^{-1} \cdot K^{-1}$)
RG paper	1	1.6	15	45.28
RG-Mg	1	1.2	9.3	52.82
RG-Ca	1	1.8	13.4	50.63
RG-S	1	3.1	19.1	65.37

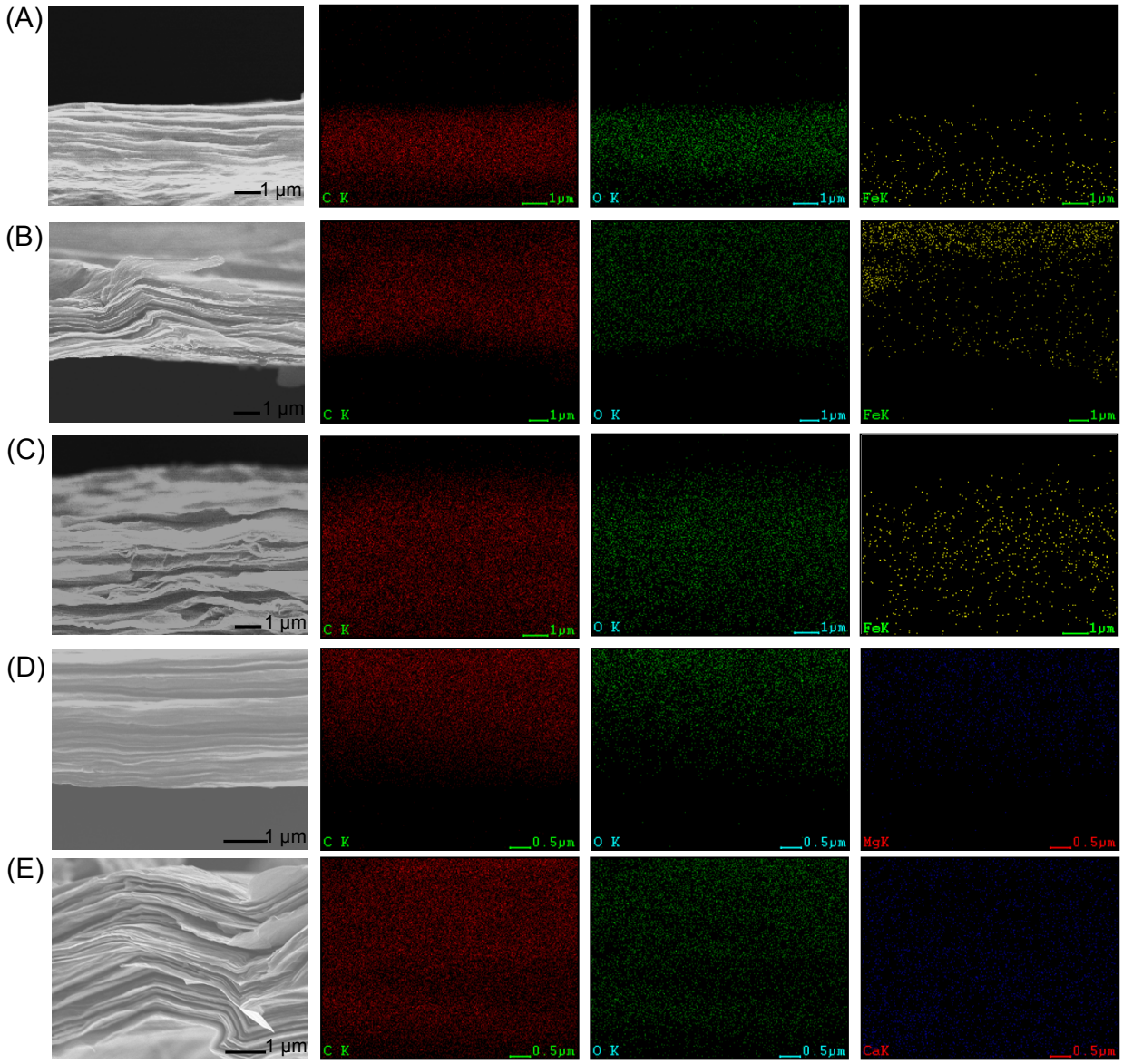

Fig. S11 EDS mapping of ion crosslinked RG papers. (A) RG-0.15Fe, (B) RG-0.27Fe, (C) RG-0.91Fe, (D) RG-Mg and (E) RG-Ca papers.

Table S3 Element analysis of crosslinked RG papers

Elements	C At.%	O At.%	Fe At.%	Mg At.%	Ca At.%	S At.%
RG-0.15Fe	72.13	27.60	0.15	-	-	-
RG-0.27Fe	71.31	27.92	0.27	-	-	-
RG-0.45Fe	77.30	22.08	0.45	-	-	-
RG-0.91Fe	69.31	28.97	0.91	-	-	-
RG-Mg	76.07	23.33	-	0.60	-	-
RG-Ca	75.57	23.91	-	-	0.52	-
RG-S	87.88	11.78	-	-	-	0.34

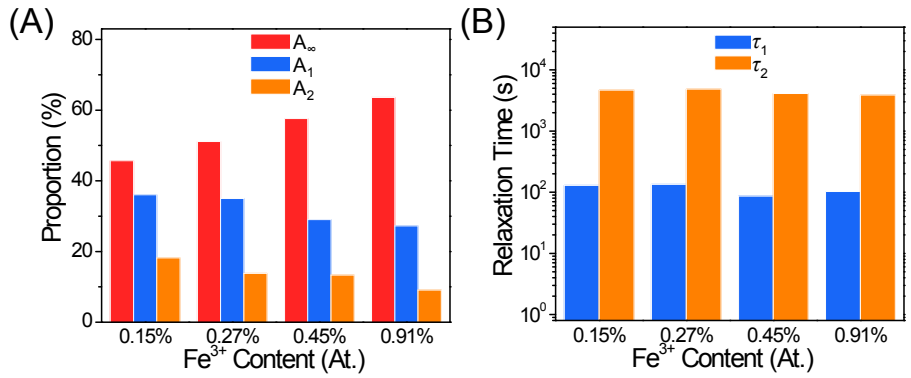

Fig. S12 (A) Proportions of each unit and (B) characteristic relaxation times (τ) of Fe^{3+} crosslinked RG papers measured by eqn. 3.

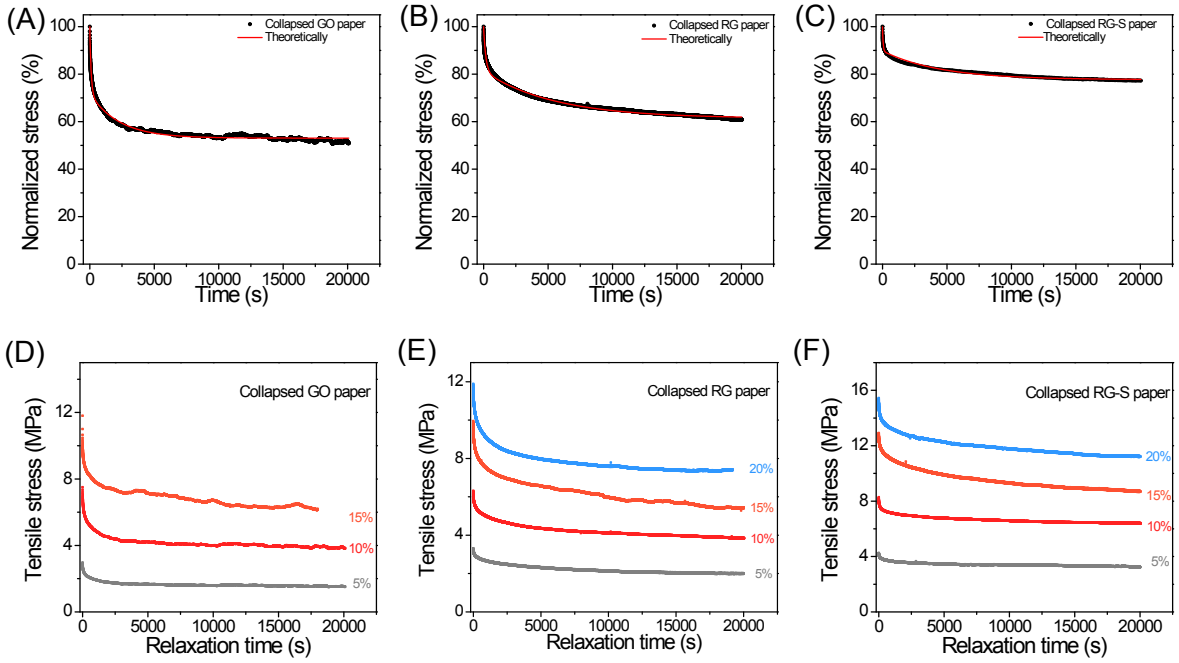

Fig. S13 Stress relaxation curves of collapsed graphene papers. (A) GO, (B) RG and (C) RG-S are all measured at strain of 10%. (D), (E) and (F) are stress relaxation behaviours of collapsed graphene papers at different strains (from 5% to 20%).

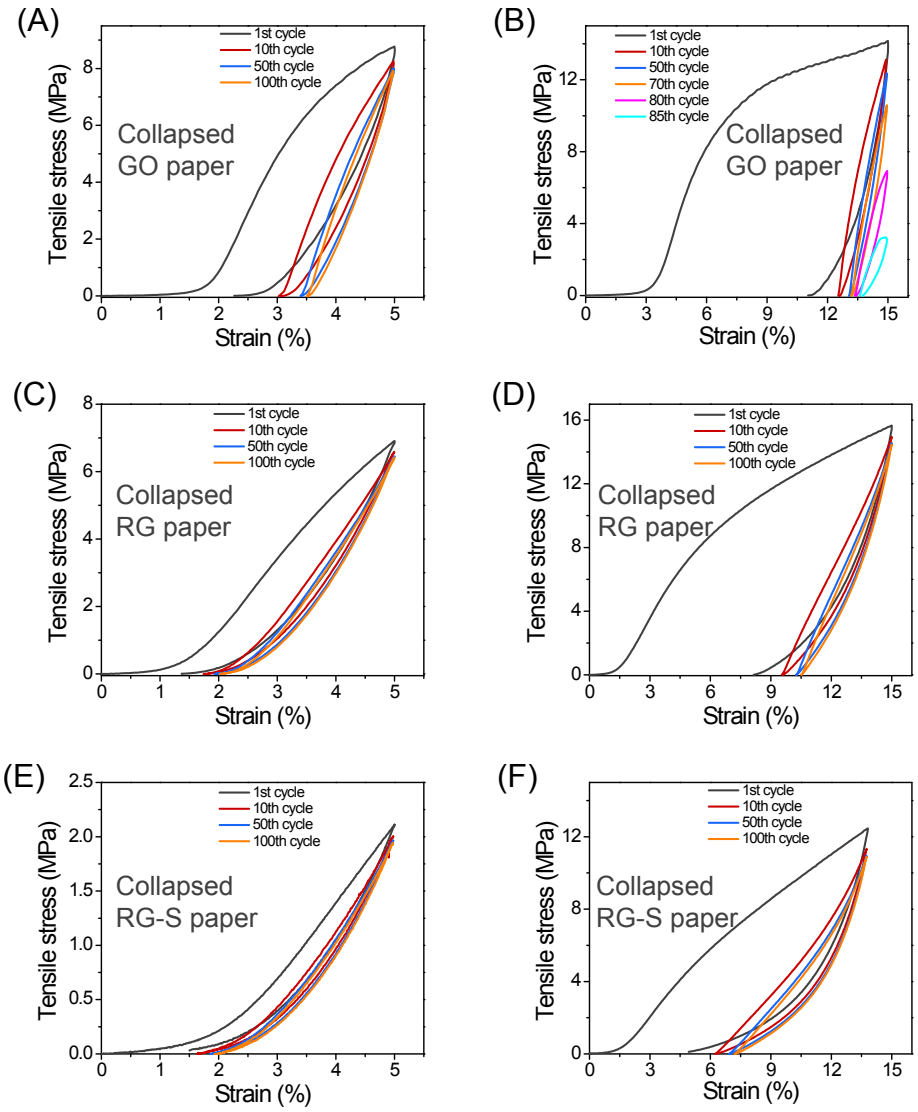

Fig. S14 Collapsed graphene papers at different strains (5% and 15%).

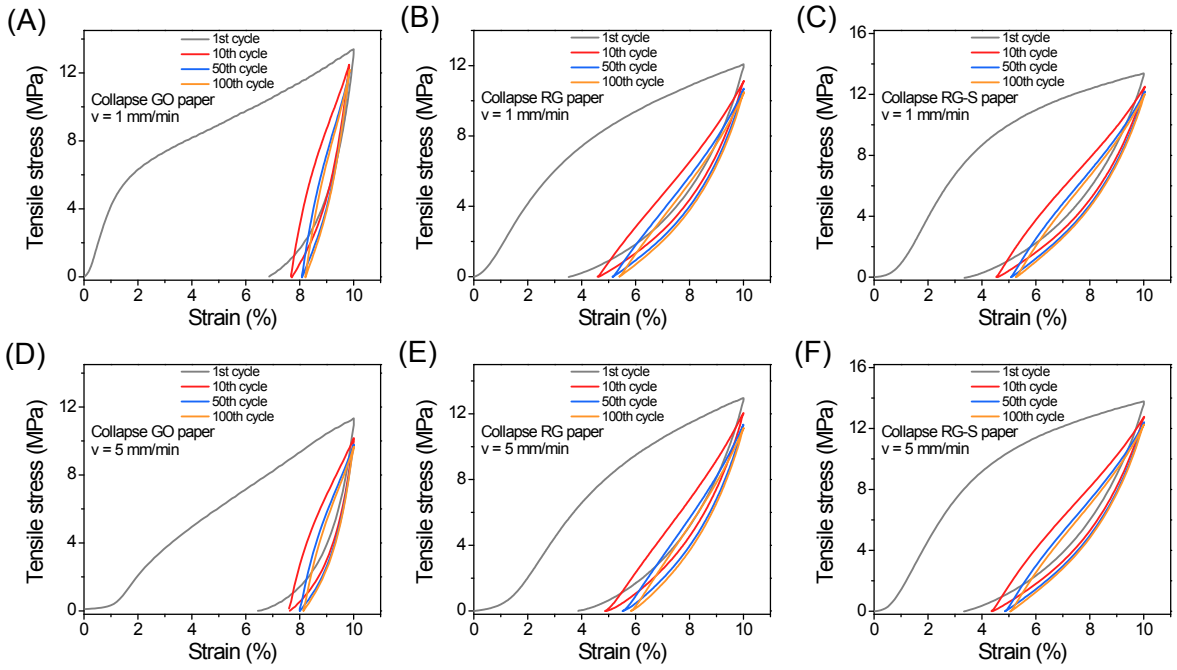

Fig. S15 Collapsed graphene papers at strain of 10% with different strain rates. (A) GO, (B) RG and (C) RG-S are tested at rate of 1 mm/min; (D) GO, (E) RG and (F) RG-S are tested at rate of 5 mm/min.

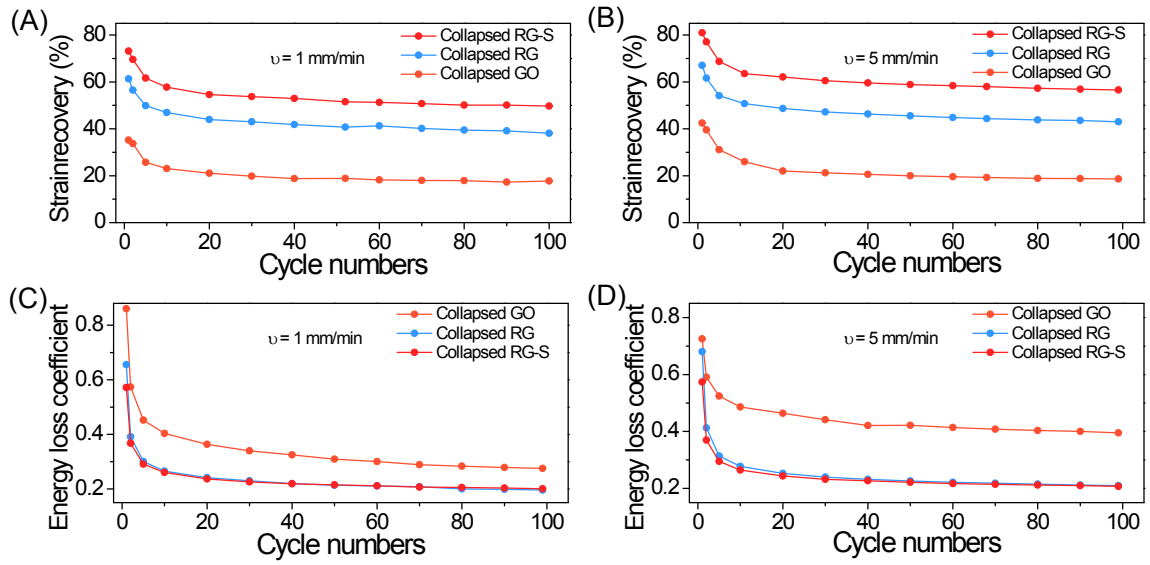

Fig. S16 Strain recovery (A, B) and energy loss coefficient (C, D) of collapsed graphene papers at rate of 1 and 5 mm/min, respectively.

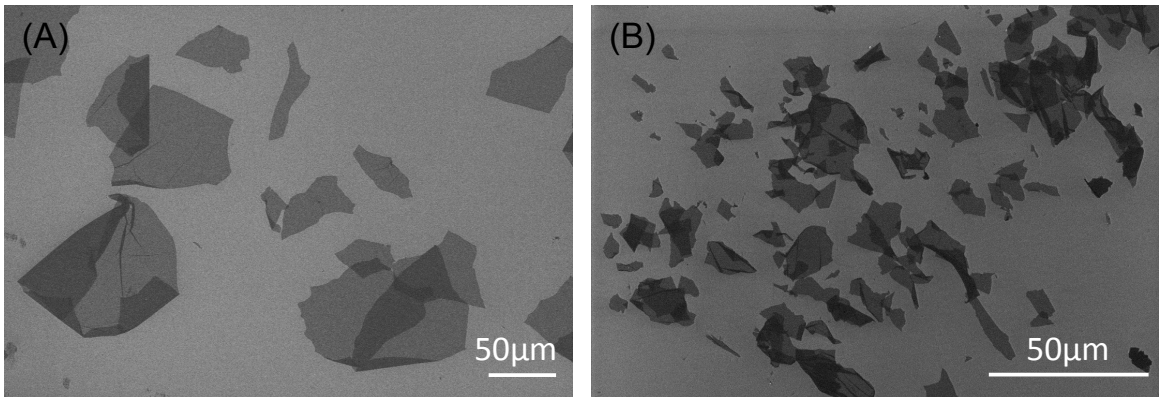

Fig. S17 SEM images of (A) GO sheets and (B) SGO sheets.