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Supporting information to

Engineering the Interface in Mechanically Responsive Graphene-Based Films

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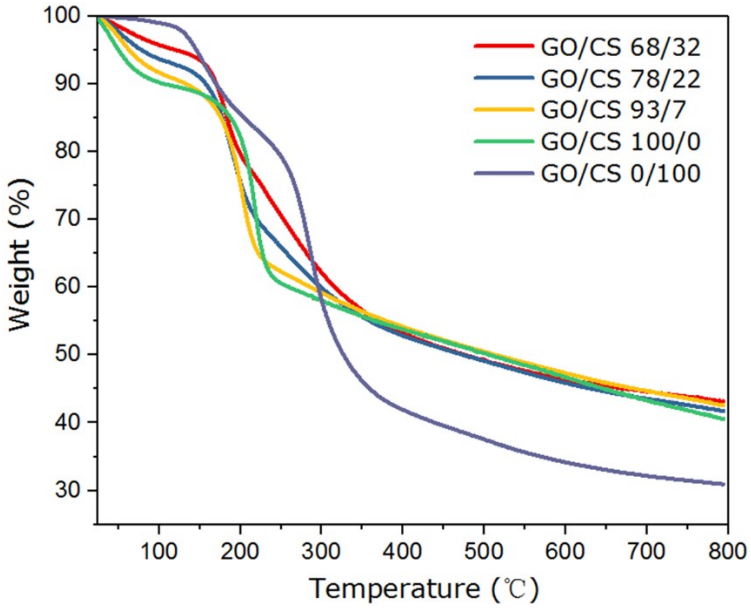

Fig. S1 TGA curves of GO/CS nanocomposte films with a heating rate of 10 °C/min under nitrogen atmosphere. Considering the weight loss before 105 °C is mainly due to water evaporating and both GO and CS have all degradation peaks before 500 °C, temperature ranging from 105 °C to 500 °C is chosen to calculate the GO weight fraction of GO/CS nanocompsoites.

The calculation of GO weight percent (w) in GO/CS nanocomposites is decieded by equation[1] S1:

$$w = \frac{M - M_{CS}}{M_{GO} - M_{CS}} \times 100\% \qquad \text{(S1)}$$

w represents the GO weight percent; M, M_{GO} and M_{CS} are weight losses of GO/CS composites between 105 °C to 500 °C, respectively.

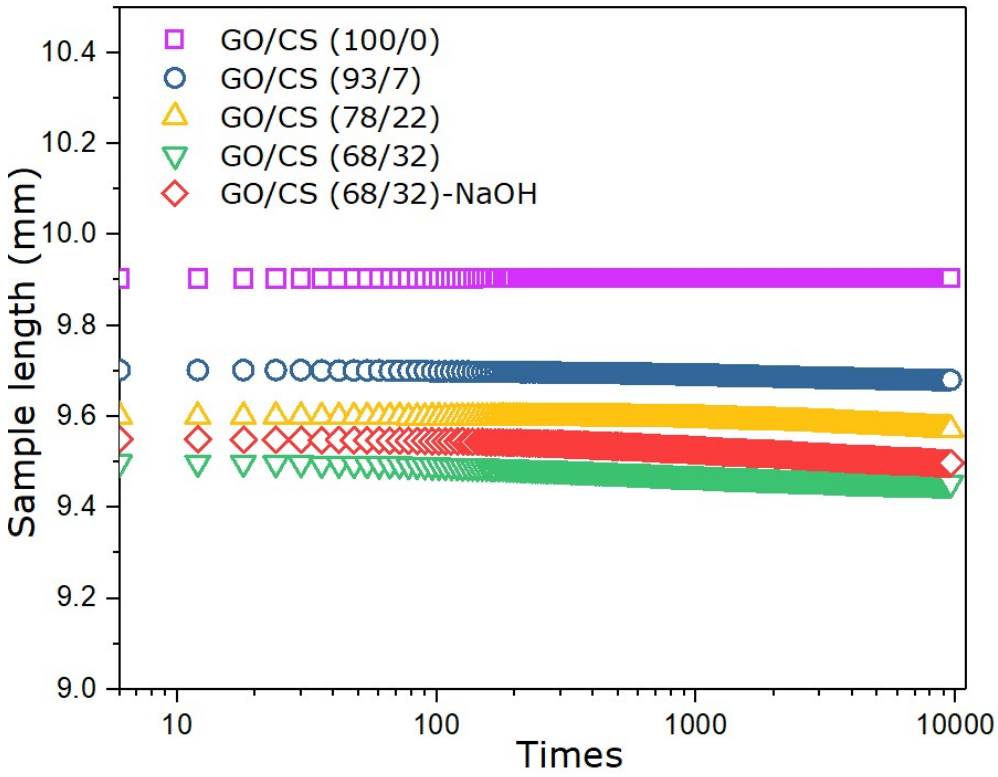

Fig. S2 Length changes ofa series of GO/CS nanocomposite films with different weight ratios (100/0, 93/7, 78/22, 68/32) andalkali treated GO/CS (68/32) films during dynamic tension.

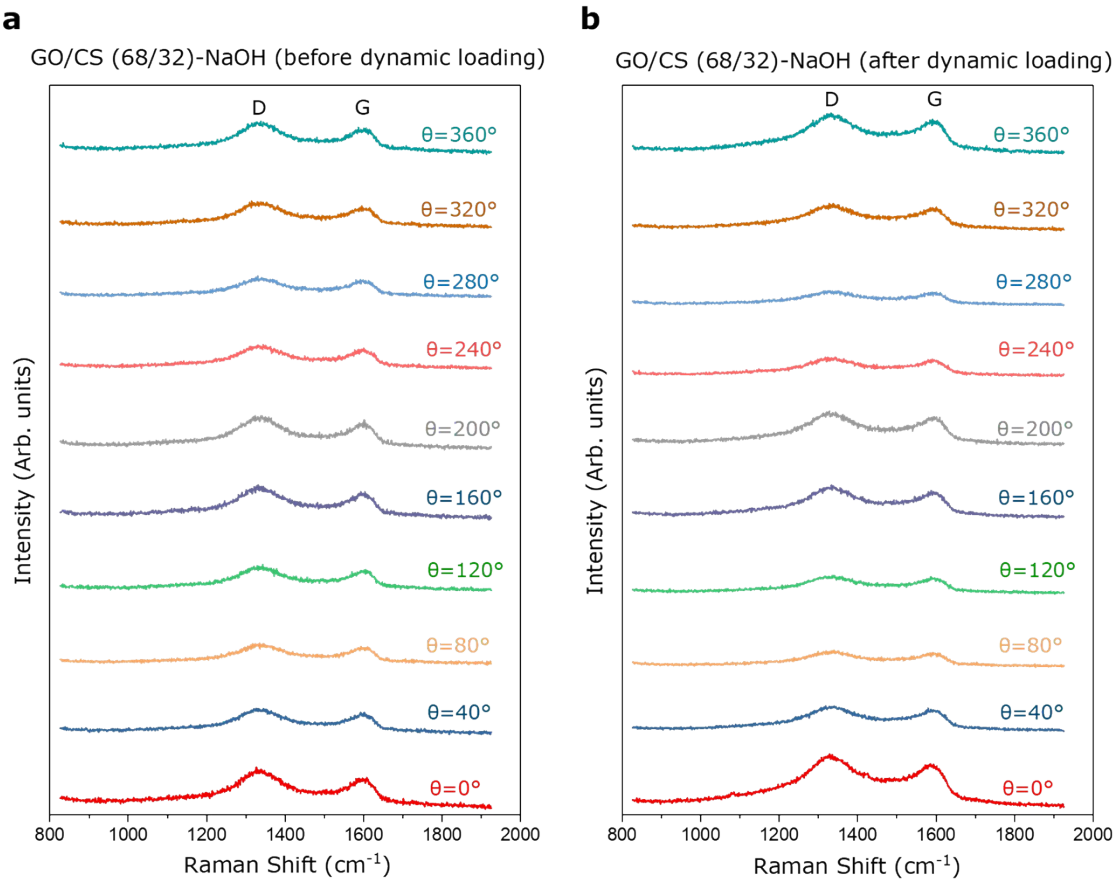

Fig. S3 Raman spectra of alkali treated GO/CS (68/32) films with varying polarization angles (θ) of incident laser for samples (a) before dynamic loading and (b) after dynamic loading. It is acknowledged that[13]

$$I_G(\alpha) = \frac{1}{2} c^2 \cos^2\alpha \left\{2 + \cos\left[2(\alpha - \theta)\right] + \cos\left[2(\alpha + \theta)\right]\right\} \quad \text{(S2)}$$

I_G reaches a maximum value I_G (∥) when the electric field vector is parallel to the base plane of the GO/CS (68/32)-NaOH film (θ=0° and 180°), while arrives at a minimum value I_G (⊥) when perpendicular to the base plane (θ=90° and 270°). I_G (∥)/ I_G (⊥) is equal to $\cot^2\alpha$, indicating a decreasing α with an increasing ration of I_G (∥)/ I_G (⊥).

Table S1. The detailed peak information from XPS spectra of untreated and alkali treated GO and GO/CS films.

	C1s					N1s		
Functional groups	C in graphite (eV)	C-N (eV)	C-O (eV)	C=O (eV)	C(O)O (eV)	NH_2 (eV)	O=C-N (eV)	NH^{3+} (eV)
GO	284.7 (52.7%)	——	286.8 (25.5%)	287.3 (10.6%)	288.6 (11.2%)	——	——	——
GO-NaOH	284.7 (72.6%)	——	286.4 (17.2%)	288.2 (10.2%)	——	——	——	——
GO/CS (68/32)	284.8 (39.6%)	286.2 (18.1%)	286.8 (20.4)	287.8 (15.3%)	288.3 (6.6%)	399.3 (53.6%)	399.9 (30.6%)	401.7 (15.8%)
GO/CS (68/32)-NaOH	284.7 (52.0%)	285.8 (14.6%)	286.4 (27.1%)	288.2 (6.3%)	——	399.7 (92.1%)	400.1 (7.9%)	——

Table S2. The mechanical properties of alkli treated GO-based films in our work and other GO-based nanocomposites reported in literatures.[1-12]

Materials	Modulus (GPa)	Stress (MPa)	Toughness (MJ/m^3)	Reference
rGO-CS	6.5	526.7	17.7	1
GO-GA	30.0	101.0	0.3	2
GO-Ca^{2+}	28.1	125.8	0.3	3
GO-Mg^{2+}	27.9	80.6	0.2	3
GO-Al^{3+}	26.2	100.5	0.2	4
GO-PMMA	7.5	148.3	2.4	5
GO-PVA	10.4	188.9	2.5	5
rGO-PAPB	7.17	382.0	7.5	7
rGO-MoS_2-TPU	10.0	235.0	6.9	8
rGO-SL	26.0	300.0	0.8	9
rGO-MMT-PVA	2.5.0	356.0	7.4	10
GO-MMT-PVA	16.0	263.0	7.5	10
rGO-DWNTs-PVA	6.0	375.8	11.3	11
rGO-PCDO-NFC	2.5	314.6	9.8	12
GO	9.4±0.8	67.0±9.8	0.3±0.1	This work
GO/CS	12.7±1.3	170.7±15.2	2.8±0.3	This work
GO/CS-NaOH	20.9±3.5	316.0±31.7	6.0±0.9	This work
GO/CS-NaOH stressed	35.1±4.2	614.0±36.6	9.3±1.7	This work

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