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Electronic Supplementary Information

Self-healable, super tough graphene oxide/poly(acrylic acid) nanocomposite hydrogels facilitated by dual cross-linking effects through dynamic ionic interactions

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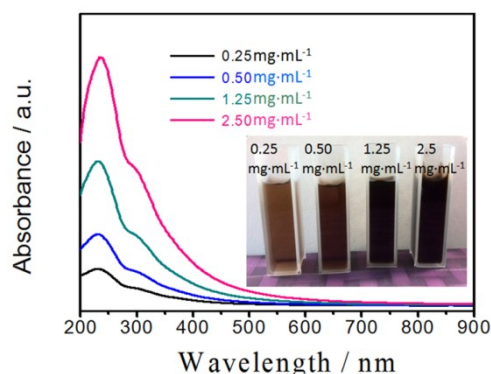


Fig.S 1 UV-vis spectra of GO aqueous solutions at various GO concentrations, *i.e.*, 0.25, 0.50, 1.25 and 2.5 mg mL⁻¹, respectively. The corresponding photographs are presented in the inset. All GO aqueous solutions are diluted by 10 times for the UV-vis measurements.

UV-vis spectra show two characteristic peaks at 239 nm and 302 nm attributed to the $\pi \rightarrow \pi^*$ transition of aromatic C-C bonds and $n \rightarrow \pi^*$ transition of C=O bonds, respectively. These spectra also indicate that homogeneous GO aqueous solutions are successfully obtained.

S2 Supplementary Tables

Table S1

Table S1 Influence of the Fe³⁺ ion content on the mechanical properties of GO/PAA nanocomposite hydrogels

samples	Tensile strength / kPa	Elongation at break / %	Modulus / kPa	Work of extension / MJ m ⁻³
0.1 mol % Fe ³⁺	272.6	2185.6	32.0	3.4
0.2 mol % Fe ³⁺	531.5	2296.2	35.6	5.9
0.5 mol % Fe ³⁺	777.3	2980.7	50.7	11.9
1.0 mol % Fe ³⁺	471.3	1661.8	35.9	3.7

The content of GO and total of water are fixed at 0.5 wt % and 80%.

Table S2

Table S2 Influence of the GO content on the mechanical properties of the GO/PAA nanocomposite hydrogels

samples	Tensile strength / kPa	Elongation at break / %	Modulus / kPa	Work of extension / MJ m ⁻³
Neat	411.8	2024.6	31.6	3.4
0.10 wt% GO	577.2	2451.1	36.3	5.8
0.25 wt % GO	660.6	2813.8	47.0	8.4
0.50 wt% GO	777.3	2980.7	50.7	11.9
0.75 wt% GO	729.3	2512.9	74.8	9.3
1.00 wt% GO	711.0	2074.7	105.5	7.5

The content of Fe³⁺ and total of water are fixed at 0.5 mol % and 80%.**Table S3**

Table S3 Influence of the water content on the mechanical properties of the GO/PAA nanocomposite hydrogels

Samples / H ₂ O	Tensile strength / kPa	Elongation at break / %	Modulus / kPa	Work of extension / MJ m ⁻³
90 %	75.6	1740.1	5.7	0.6
85 %	274.3	1819.3	24.2	2.2
80 %	777.3	2980.7	50.7	11.9
75 %	844.1	2290.5	65.3	10.2
70 %	862.5	2056.6	90.8	9.2

The content of GO and Fe³⁺ are fixed at 0.5 wt % and 0.5 mol %.

S3 Supplementary Videos

Supplementary Video 1

The GO/PAA nanocomposite hydrogels (GO = 0.5 wt%, Fe³⁺ = 0.5 mol%, H₂O = 80 wt%) tube sample can be repeatedly blown up and immediately blown off. This movie shows the large deformation, recoverable behavior of the GO/PAA nanocomposite hydrogels.

Supplementary Video 2

The balloon can maintain the configuration under high deformation, and can also be quickly recovered after being slightly compressed. This movie shows the super tough and highly stretchable properties of the GO/PAA nanocomposite hydrogels.

Supplementary Video 3

This movie shows that the GO/PAA nanocomposite hydrogels (GO = 0.5 wt%, Fe³⁺ = 0.5 mol%, H₂O = 80 wt%) have good self-healing properties. The cut-off samples, after being treated at 45 °C for 48 h, can be stretched over 20 times compared to the original length.