Electronic Supplementary Material (ESI) for *Soft Matter*

Self-Healable, Tough and Highly Stretchable Ionic Nanocomposite Physical Hydrogels

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Characterization of VSNP

Transmission electron microscopy (TEM) images were obtained using a Hitachi H7700 electron microscope operating at an accelerating voltage of 100 kV. Samples for imaging were prepared by placing drops of a diluted VSNP aqueous suspension onto a carbon coated copper grid, which was then dried under ambient conditions prior to being introduced into the TEM chamber.

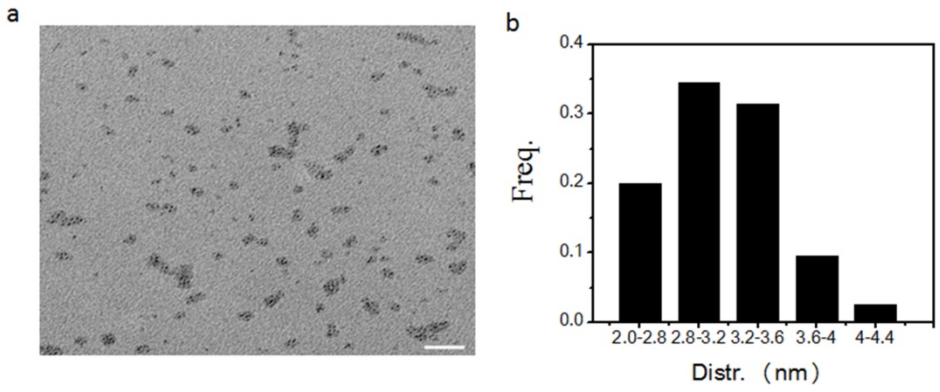

Fig. S1 TEM image of the vinyl hybrid silica nanoparticles (a) and the diameter distribution of the VSNPs (b). Scale bar is 50 nm.

It was found that the VSNPs have a uniform diameter of 3 nm and was well dispersed within the solution.

Self-healing efficiency of Ionic-NCP gels

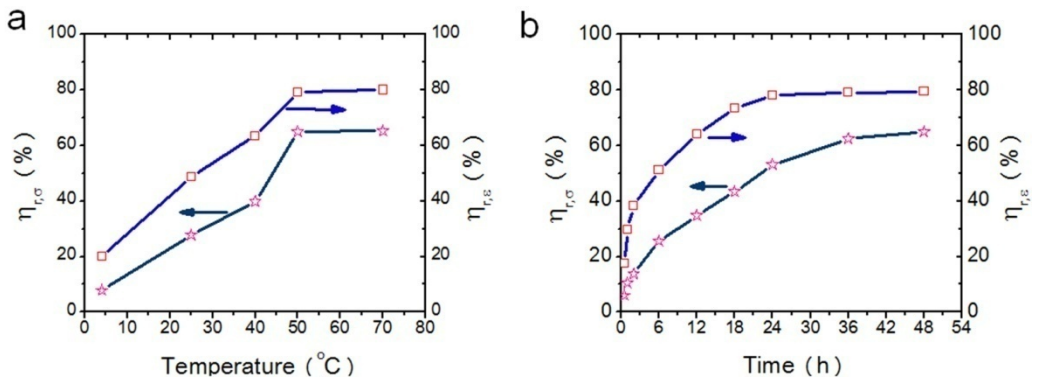

Fig. S2 The mechanical properties recovery efficiency of self-healing of the Ionic-NCP gel(VSNP = 0.50 wt%, Fe^{3+} = 0.50 mol%, H_2O = 80 wt%) preset for 48 h at different temperatures (a), and treated at 50 °C for different time(b). □indicates the values measured during the application of strain and ☆indicates those obtained during removal of strain.

The ratio of the tensile strength ($\eta_{r,\sigma}$) and elongation at break ($\eta_{r,\varepsilon}$) of the healed gel to that of the original gel, to define the healing efficiency of the gels.

Supporting Tables

Table S1 Effect of ferric ions content on the mechanical properties of the Ionic-NCP gels

Samples Fe^{3+}/ mol%	Tensile strength / kPa	Elongation at break / %	Modulus / kPa	Fracture energy / MJ m^{-3}
0	49.2 ± 3.3	1383.5 ± 75.2	15.1 ± 0.3	0.3
0.10	152.4 ± 9.6	1609.7 ± 95.8	22.0 ± 0.8	1.0 ± 0.1
0.20	375.3 ± 8.3	2036.1 ± 105.3	28.8 ± 1.4	2.9 ± 0.1
0.50	862.3 ± 10.2	2367.2 ± 152.7	36.8 ± 2.0	8.3 ± 0.2
1.00	665.0 ± 14.4	2011.6 ± 86.6	47.5 ± 2.5	5.8 ± 0.1

The total water content and VSNP content were fixed at 80 wt% and 0.50 wt%, respectively.

Table S2 Effect of VSNP content on the mechanical properties of the Ionic-NCP gels

Samples VSNP / wt%	Tensile strength / kPa	Elongation at break / %	Modulus / kPa	Fracture energy / MJ m^{-3}
0.10	589.4 ± 12.5	2451.3 ± 125.3	27.7 ± 1.2	5.7 ± 0.1
0.20	746.0 ± 15.3	2595.2 ± 102.5	30.2 ± 1.6	7.8 ± 0.2
0.50	862.3 ± 10.2	2367.2 ± 152.7	36.8 ± 2.0	8.3 ± 0.2
1.00	676.7 ± 8.6	1795.6 ± 132.1	46.7 ± 3.2	4.5 ± 0.1

The total water content and ferric ions content were fixed at 80 wt% and 0.50 mol%, respectively.

Table S3 Effect of total water content on the mechanical properties of the Ionic-NCP gels

Samples H_2O / wt%	Tensile strength / kPa	Elongation at break / %	Modulus / kPa	Fracture energy / MJ m^{-3}
90	54.6 ± 2.7	1038.4 ± 65.6	5.7 ± 0.4	0.2
85	196.2 ± 8.5	1755.1± 102.5	15.2 ± 1.3	1.5 ± 0.1
80	862.3 ± 10.2	2367.2 ± 152.7	36.8 ± 2.0	8.3 ± 0.2
75	1095.9 ± 18.3	2175.3 ± 85.2	53.8 ± 2.6	10.4 ± 0.3
70	1288.9 ± 20.1	1878.8 ± 65.4	88.6 ± 3.1	9.9 ± 0.3

The VSNP and ferric ions content were fixed at 0.50 wt% and 0.50 mol%, respectively.

Supporting Videos

Supporting Video 1

The Ionic-NCP hydrogel was repeatedly inflated and deflated. After repeated deformations, the rough surface of the tubal Ionic-NCP gel becomes more and more smooth. This movie shows the ability of the Ionic-NCP gel to undergo large deformation from which it can recover. It also indicated the deformation-triggered uniform network behavior of the Ionic-NCP gel (VSNP = 0.50 wt%, Fe^{3+} = 0.50 mol%, H_2O = 80 wt%).

Supporting Video 2

This movie shows the highly tough and stretchable behavior of the tubular Ionic-NCP gel. The highly stretched Ionic-NCP gel, in the form of a balloon can be compressed, and once the stress relax, it recovers. It also indicated the highly tough and stretchable properties of the Ionic-NCP gel (VSNP = 0.50 wt%, Fe^{3+} = 0.50 mol%, H_2O = 80 wt%).

Supporting Video 3

The healed hydrogels can be stretched more than 18 times the initial length. This movie indicates the Ionic-NCP gel (VSNP = 0.50 wt%, Fe^{3+} = 0.50 mol%, H_2O = 80 wt%) has good self-healing properties.