Electronic Supplementary Information

Exceptionally Tough and Notch-Insensitive Magnetic Hydrogels

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Movie S1: Movement of a magnetic hydrogel in a plastic plate.

A rod-shaped magnetic hydrogel fast rolled in a plastic plate, (8.5 cm in diameter and 1.2 cm in height) containing deionized water under the remote guidance of a magnet.

Movie S2: The elongation of notch insensitive magnetic hydrogel with single edge notch.

A sheet of magnetic hydrogel (length 35 mm, width 30 mm, thickness 2 mm) was cut at one center edge of the sample (notch length 1 cm). The sample was stretched at a velocity of 2 mm per minute. The stretch direction was perpendicular to the notch.

Movie S3: The elongation of notch insensitive magnetic hydrogel with multi notches at side edge.

A sheet of magnetic hydrogel was symmetrically cut at the two edges. Each edge has three notches (notch length 0.5 cm). The distance between every two notches was 2 mm. The sample was stretched at a velocity of 20 mm per minute. The stretch direction was perpendicular to the notches.

Movie S4: Magnetic catheter navigation.

A long flexible Fe_3O_4@Fe-alginate/PAAm hydrogel (40 mm in length, 5 mm in diameter) with 20 wt% Fe_3O_4 nanoparticles was navigated in a calcium ions crosslinked alginate/PAAm hydrogel tube (length 250 mm, inner diameter 7 mm) under the remote magnetic attraction of a NdFeB magnet.

Preparation of poly(2-acrylamido-2-methyl propyl sulfonic acid (PAMPS) hydrogel

We prepared 23.0 wt% solution of 2-acrylamido-2-methyl propyl sulfonic acid AMPS in water. Then added MBAA (4 wt %) and 2-oxoglutaric acid (0.9 wt %) with respect to the weight of AMPS. This transparent solution was transferred into glass mould and placed under UV light at 365 nm for 6 h for gelation. After that, PAMPS hydrogel was separeted from the glass mould.

Determination of fracture energy

The fracture energy of Fe_3O_4@Fe-alginate/PAAm hydrogels was measured and calculated using a method introduced by Rivlin and Thomas.[1,2,] In brief, two samples (unnotched samples and notched sample) with same width a_0, thickness b_0 were selected. First the unnotched sample was stretched to obtain the force displacement curve. The area beneath the curve gave the work done by the force, $W(L)$. Then, the notched sample was stretched to measure the critical distance between the clamps, L_c, which was determined as the distance when the notch turned into a running crack and the sample ruptured. Finally, the fracture energy was calculated as $W(L_c)/a_0b_0$ with the dimension of J/m^2.

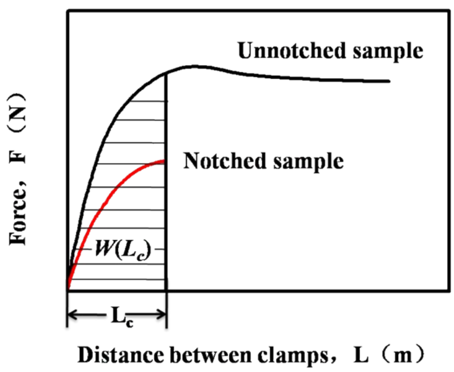

Figure S1: Determination of fracture energy.

Fourier transform infrared spectroscopy (FTIR)

To investigate the presence of alginate to Fe_3O_4 nanoparticles, the dried Fe_3O_4 nanoparticles, alginate and alginate-coated Fe_3O_4 samples were tested using an infrared spectrometer (BRUKER TENSOR 27, Bruker). All the samples were subjected to analysis with a range of 4000-400 cm^{-1}.

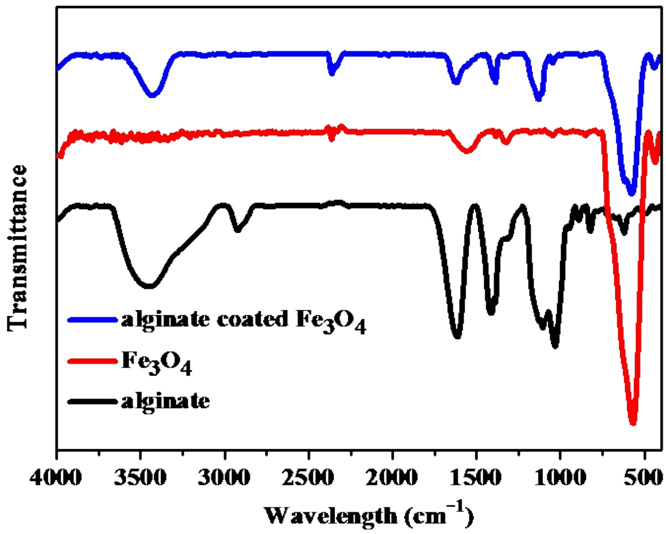

Figure S2: FTIR of alginate-coated Fe_3O_4 nanoparticles, Fe_3O_4 nanoparticles, and alginate. The Fe_3O_4 nanoparticles possess the strong band at 569.20 cm^{-1} in the low frequency region of (1000–500 cm^{-1}) which is due to iron oxide skeleton. In the spectrum of alginate-coated Fe_3O_4, two bands were observed at 1626 cm^{-1} and 1387 cm^{-1}.

Characterization of Fe_3O_4 nanoparticles

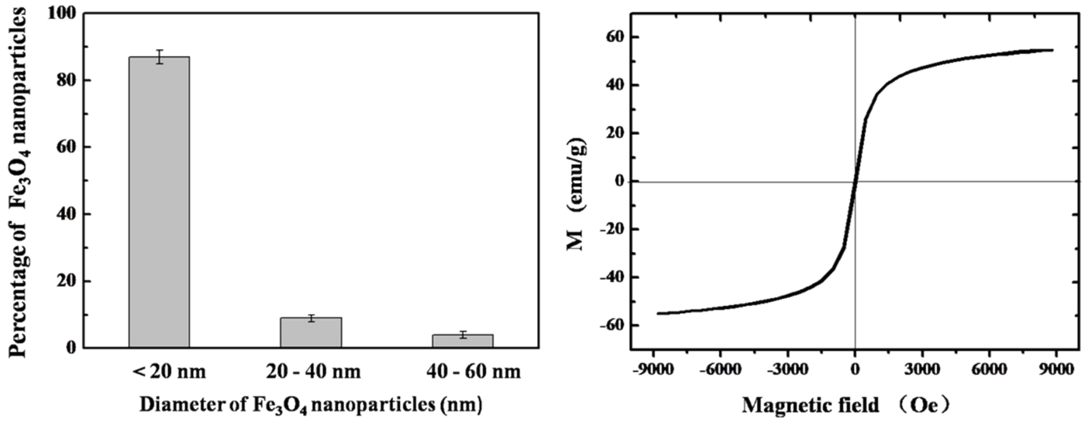

Figure S3: Size distribution of alginate-coated Fe_3O_4 nanoparticles of 1.0 wt% Fe_3O_4@Fe-alginate/PAAm hydrogel (left), and magnetic hysteresis loop of pure magnetic nanoparticles(right). TEM image shows that the diameter of over 80% nanoparticles is below 20 nm. Only less than 10% fell in the range of 21−40 nm or 41−60 nm respectively. 200 Fe_3O_4 nanoparticles were selected from each TEM images for the statistics. Error bars denote the standard deviation from at least three TEM images.

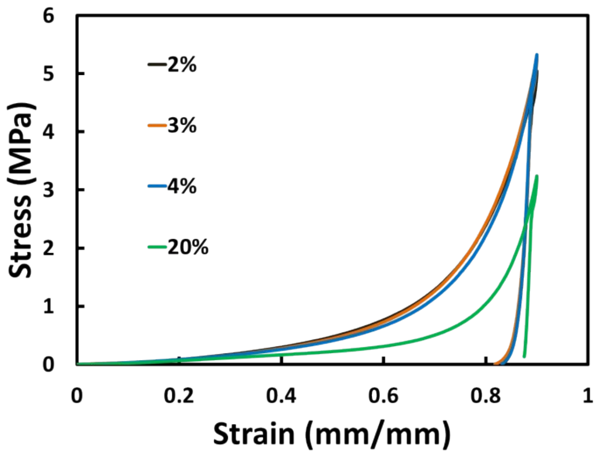

Figure S4: Compressive hysteresis curves of the tough magnetic Fe_3O_4@Fe-alginate/PAAm hydrogels when $W_{Fe_3O_4}$ = 2.0, 3.0, 4.0, 20.0 wt%.

Table S1: Mechanical properties of tough magnetic hydrogels with various contents of Fe_3O_4 nanoparticles (wt %).

Magnetic Hydrogel [wt%]	Tensile Strength [kPa]	Tensile Strain [mm mm^{-1}]	Energy Dissipation [kJ m^{-3}]	Tensile Modulus [kPa]	Fracture Energy [J m^{-2}]	Compressive Stress (Strain = 0.9) [MPa]
1.0	916.9 ± 53.3	11.4 ± 1.5	3445.0±140.3	199.5 ± 28.1	2814.0 ± 69.6	4.9 ± 0.2
2.0	827.6±197.3	11.3 ± 2.7	2649.4 ± 57.7	201.0 ± 36.5	2782.1 ± 69.6	5.1 ± 0.1
3.0	712.1±136.8	9.9 ± 0.5	2598.2 ± 31.4	203.1 ± 25.4	2613.1±227.5	5.2 ± 0.3
4.0	571.4 ± 21.1	9.8 ± 0.5	2282.9 ± 62.7	205.9 ± 36.9	2572.7 ± 44.2	5.6 ± 1.6
5.0	567.1 ± 50.5	8.8 ± 1.5	2013.0 ± 12.0	223.6 ± 15.6	2521.8±116.5	5.6 ± 0.6
10.0	334.7 ± 9.0	8.0 ± 0.4	1024.0 ± 43.2	215.9 ± 8.1	2279.1 ± 40.8	3.2 ± 0.4
20.0	201.0 ± 9.1	2.7 ± 0.4	329.1 ± 10.0	191.7 ± 5.8	1550.5±194.9	3.1 ± 0.2

References

1 R. S. Rivlin and A. G. Thomas, *J. Polym. Sci.*, 1953, **10**, 291.

2 J. Y. Sun, X. Zhao, W. R. Illeperuma, O. Chaudhuri, K. H. Oh, D. J. Mooney, J. J. Vlassak and Z. Suo, *Nature* 2012, **489**, 133.