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# Supporting Information

# Magnetic Double-Network Hydrogels for Tissue Hyperthermia and Drug Release

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## Table S1 Specific Characteristics of DN and M-DN Gels

| Concentration $C_{NaOH}$ (M) | Fracture stretch $\lambda_b$ | Fracture stress $\sigma_b$ (MPa) | Modulus $E$ (MPa) | Toughness $\Gamma$ (J/m$^2$) | Dissipated energy $W_d$ (MJ/m$^3$) | Saturation magnetization $M_s$ (emu/g) |
|---|---|---|---|---|---|---|
| 0[a)] | 11.28 | 1.23 | 0.38 | 2727.74 | 0.94 | / |
| 0.1 | 8.38 | 0.81 | 0.43 | 1531.00 | 0.86 | 2.61 |
| 0.2 | 6.59 | 0.50 | 0.41 | 743.35 | 0.63 | 3.50 |
| 0.5 | 4.92 | 0.25 | 0.21 | 214.39 | 0.35 | 4.16 |
| 1 | 4.96 | 0.22 | 0.19 | 285.43 | 0.32 | 4.98 |

a) 0M denotes the conventional DN hydrogel without magnetic properties.

## Comparison of the mechanical properties of magnetic DN gels with other materials

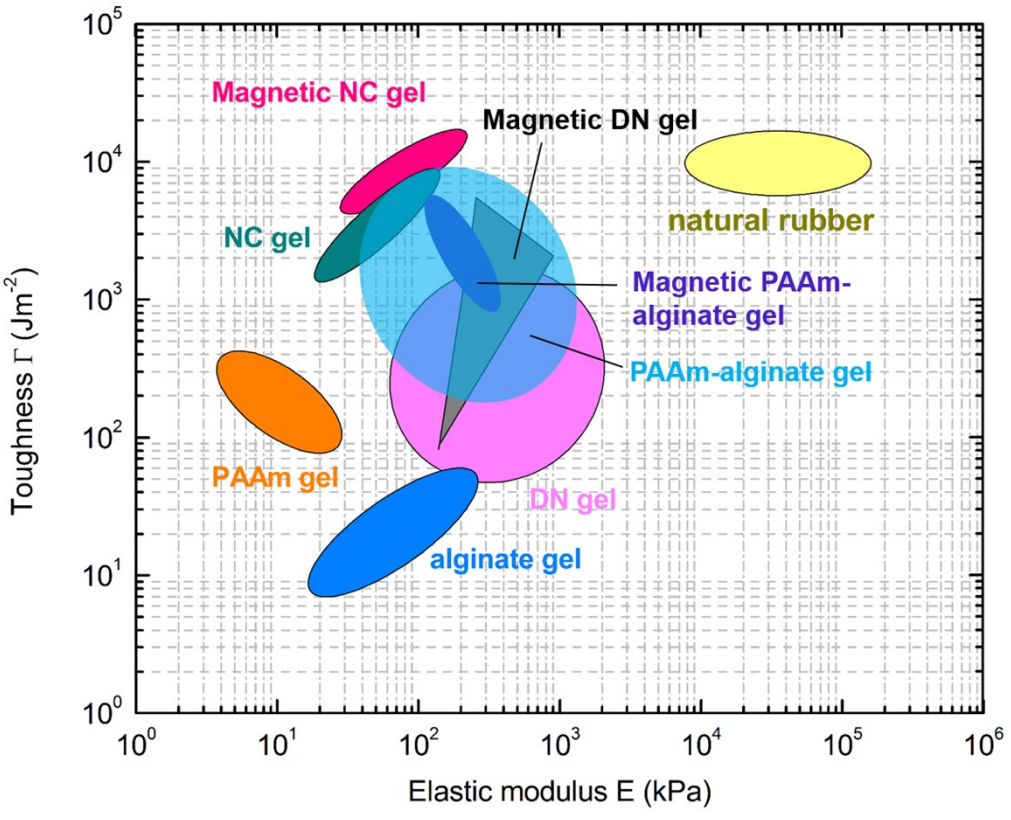


**Figure S1.** Ashby plot for various soft materials. Magnetic DN hydrogels developed in this paper are compared to other well-known hydrogels (summarized in our previous paper, Ref [5] of the main text) in terms of modulus and toughness.

**Hydrolysis of PAAm network of DN hydrogel in NaOH solutions**

The hydrolysis of the amide groups will introduce negative charges into the polymer and yield an acrylamide-acrylic acid random copolymer as the second network. This makes both the two networks polyelectrolyte (Fig. S2,a) and induces additional swelling of DN hydrogels (Fig. S2,b). M-DN hydrogels also suffer hydrolysis in NaOH solutions and the swelling is similar to those non-magnetic counterparts (Fig. S2,c).

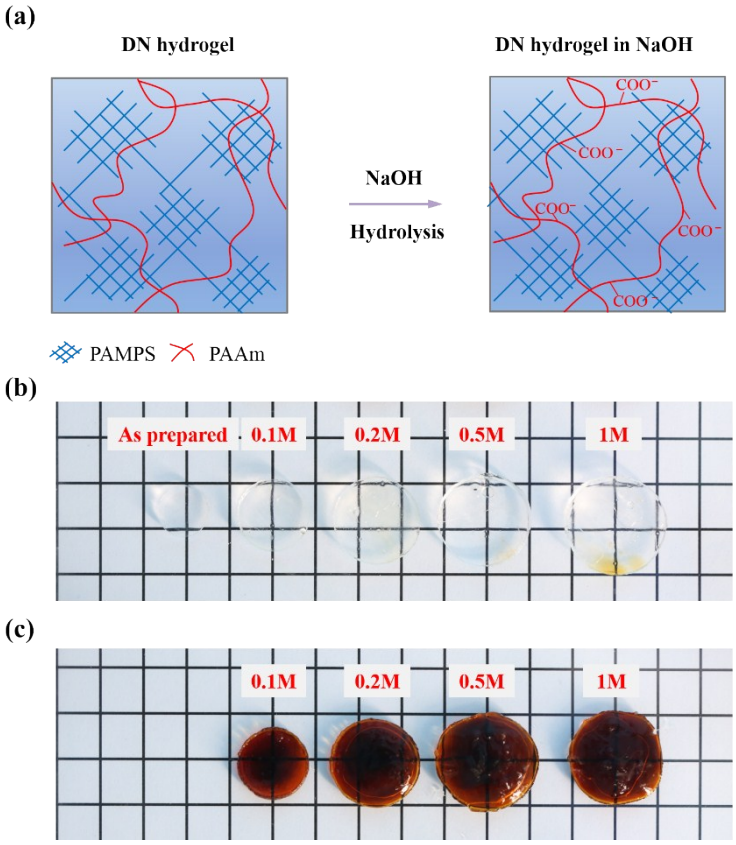


**Figure S2.** (a) The second polymer network, polyacrylamide, would suffer hydrolysis in NaOH solutions and will introduce negative charges to afford an acrylamide-acrylic acid radom copolymer. (b) The additional swelling of DN hydrogels due to the hydrolysis of polyacrylamide increases with $C_{NaOH}$. (c) The additional swelling of M-DN hydrogels prepared from NaOH solutions of different concentrations. The square mesh size in the photos is 8mm.

**Stability of M-DN hydrogels in PBS compared to M-alginate/PAAm**

Both the two polymer networks of M-DN hydrogels are chemically crosslinked, thus they are inert to the salt ions. In contrast, for M-alginate/PAAm hydrogels, the alginate chains are crosslinked by $Ca^{2+}$ ions. The $Na^{+}$ ions in PBS solutions can exchange with $Ca^{2+}$ ions in the hydrogels as shown in Fig. S3. This will cause the dissociation of the alginate polymer network and the toughening mechanism fails for this hydrogel. This will greatly deteroirate the mechanical performace of the M-alginate/PAAm hydrogels.

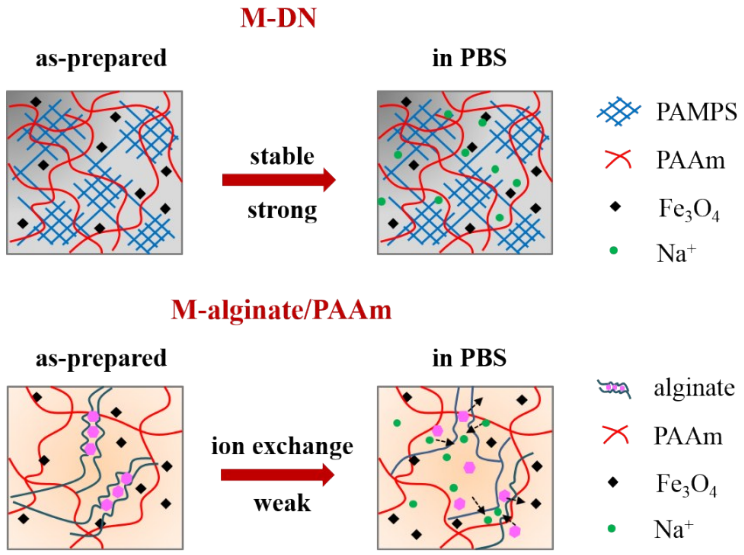


**Figure S3.** Schematic of the polymer network of M-DN and M-PAAm/alginate hydrogels upon immersion of PBS.

**Adhesion between M-DN hydrogels and tissues**

The adhesion between M-DN hydrogels and tissues were characterized by the peeling test. Figure S4,a shows the schematic of the sample for the peeling test. The M-DN hydrogel and the tissue were glued by a tissue adhesive Histoacryl®. A backing layer (PET film) was glued ontop of the M-DN hydrogel to constrain the stretching of the peeling arm. Figure S4,b shows the sample composed of M-DN hydrogel and porcine tenderloin. Figure S4,c and Figure S4,d show the force-displacement curves and the adhesion energy between the M-DN hydrogel and various tissues, respectively. The

adhesion between the tissues and magnetic hydrogels is satisfactory for the application we concerned. The adhesion energy is on the order of 100J/m$^2$ for all the tissues we studied.

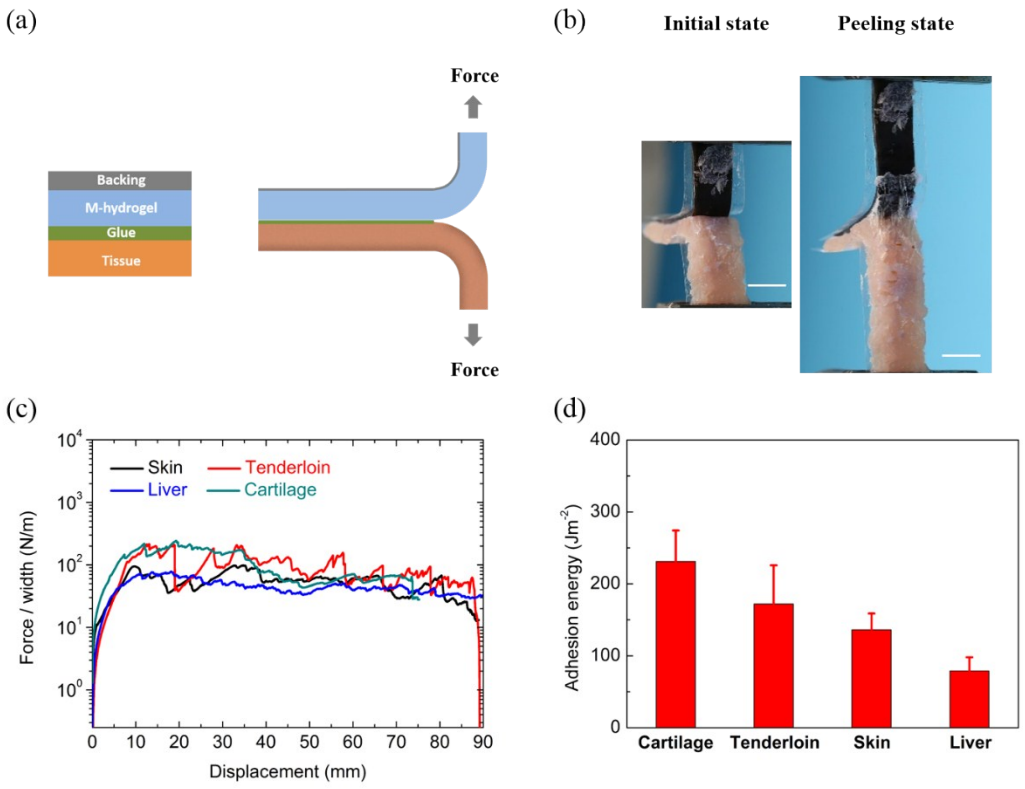


**Figure S4.** Bonding M-DN gels to biological tissues. Schematic (a) and picture (b) of the peeling test for the M-DN gel and tissue (porcine tenderloin). Scale bar: 10mm. (c) The force-displacement curves in the peeling test. (d) The adhesion energy between the M-DN hydrogel and various tissues.

**The standard absorbance-concentration curve of the vitamin $B_{12}$ solution**

We calibrated the concentration of the vitamin $B_{12}$ solution through the standard absorbance-concentration curve. The vitamin $B_{12}$ solution shows a characteristic peak at 361nm. We prepared the vitamin $B_{12}$ solutions with concentration of 5, 10, 25, 50, 75, 100 μg/mL. We plotted the intensity versus concentration curve on Fig. S5 and a linear relation was observed. For the drug release experiment, we measured the absorbance of the released drug solution at 361nm and determined its concentration through this linear relation.

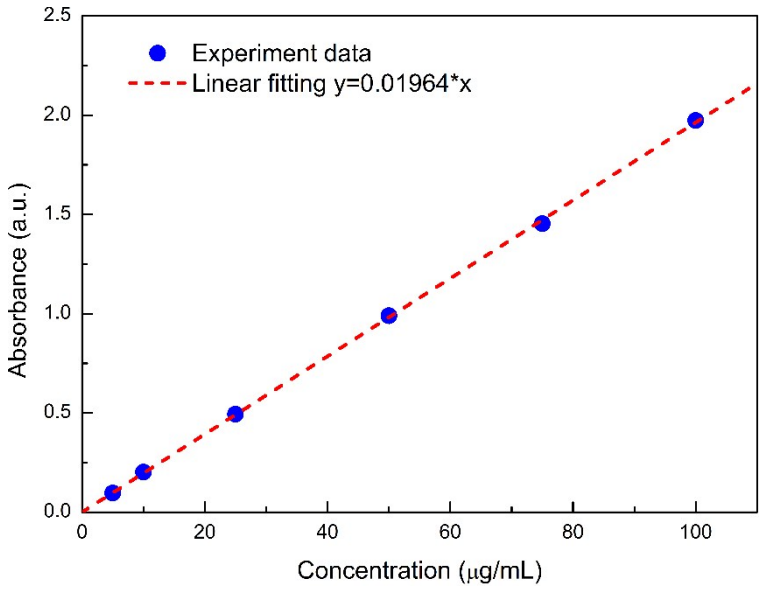


**Figure S5.** The standard absorbance-concentration curve of the vitamin $B_{12}$ solution.