**Electronic Supplementary Information**

**Hydroxyethyl Cellulose-based Electrically Conductive Mechanically Resistant Strain-Sensitive Self-Healing Hydrogels**

Imtiaz Hussain1\*, Xiaofeng Ma1, Linlin Wu2, and Zhenyang Luo1\*

1Department of Chemistry and Materials Science, College of Science, Nanjing Forestry

University, 210037, P.R. China.

2College of Materials Science and Engineering, Nanjing Tech University, Nanjing 211816, P. R.

China

* To whom the correspondence should be addressed:
* Tel: +86-25-85427080
* E-mail: imtiazhussain@njfu.edu.cn

# Composition of hydrogels and their mechanical strength

Table S1. Comparative tensile strength in terms of fracture stress, strain and toughness as a function of various constituent’s composition of original (O) and healed (H) hydrogels

|  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| Composition of Hydrogel | | | | | Stress (MPa) | | Strain (%) | | Toughness (MJm-3) | |
| S.No | **HEC**  **(%)** | **PVA**  **(%)** | **AA**  **(mL)** | **Fe3+**  **(M)** | **O** | **H** | **O** | **H** | **O** | **H** |
| 1 | 2 | 10 | 3 | 0.20 | 0.34 | 0.30 | 880 | 830 | 1.31 | 1.07 |
| 2 | 3 | 10 | 3 | 0.20 | 0.38 | 0.35 | 1100 | 1040 | 1.95 | 1.67 |
| 3 | **4** | **10** | **3** | **0.20** | **0.51** | **0.49** | **1250** | **1235** | **2.98** | **2.78** |
| 4 | 5 | 10 | 3 | 0.2 | 0.45 | 0.38 | 1010 | 930 | 2.12 | 1.61 |
| 5 | 4 | 5 | 3 | 0.20 | 0.43 | 0.37 | 1350 | 1300 | 2.56 | 2.15 |
| 6 | 4 | 15 | 3 | 0.20 | 0.50 | 0.42 | 1040 | 920 | 2.28 | 1.68 |
| 7 | 4 | 10 | 3 | 0.05 | 0.28 | 0.25 | 1290 | 1200 | 1.64 | 1.36 |
| 8 | 4 | 10 | 3 | 0.10 | 0.42 | 0.38 | 1350 | 1240 | 2.56 | 2.14 |
| 9 | 4 | 10 | 3 | 0.40 | 0.40 | 0.36 | 730 | 680 | 1.28 | 1.06 |
| 10 | 4 | 10 | 3 | 0.60 | N/A | N/A | N/A | N/A | N/A | N/A |
| 11 | 4 | 10 | 3 | 0.80 | N/A | N/A | N/A | N/A | N/A | N/A |

Table S2. Comparative percent growth rates as a function of various constituent’s composition of original and healed hydrogels

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| Variation in Composition | % G.R.  in stress | | % G.R.  in strain | | % G.R.  in toughness | |
| **Original** | **Healed** | **Original** | **Healed** | **Original** | **Healed** |
| HEC % Concentration | | | | | | |
| 2-3% | 11.76 | 16.67 | 25.00 | 25.30 | 49.15 | 55.58 |
| 2-4% | 50.00 | 44.12 | 42.05 | 48.81 | 128.45 | 158.89 |
| 2-5% | 32.35 | 11.76 | 14.77 | 12.05 | 62.71 | 49.83 |
| 3-4% | 34.21 | 40.00 | 13.64 | 18.75 | 53.17 | 66.41 |
| 4-5% | -11.76 | -22.45 | -19.20 | -24.29 | -28.78 | -42.10 |
| Fe3+ Molar Concentration | | | | | | |
| 0.05-0.10 M | 50.00 | 52.00 | 4.65 | 3.33 | 55.94 | 57.09 |
| 0.05-0.20 M | 82.14 | 96.00 | -3.10 | 2.92 | 81.66 | 104.35 |
| 0.05-0.40 M | 42.86 | 55.00 | -43.41 | -43.33 | -21.95 | -21.95 |
| 0.10-0.20 M | 21.43 | 11.00 | -7.41 | -0.40 | 16.49 | 30.00 |
| 0.20-0.40 M | -21.56 | -26.53 | -41.60 | -44.93 | -57.03 | -61.80 |
| PVA % Concentration | | | | | | |
| 5-10% | 18.60 | 32.43 | -7.41 | -5.00 | 16.49 | 29.27 |
| 5-15% | 16.27 | 13.51 | -22.96 | -29.23 | -10.76 | -22.54 |
| 10-15% | -1.96 | -14.29 | -16.00 | -25.51 | -23.41 | -40.08 |

# Measurement of the swelling property

The hydrogel samples with identified weight were dried in a vacuum oven at 60⁰C for 24 hours. The dry hydrogel samples were immersed in DI water after measuring their weight on an analytical balance. The hydrogel samples were left in DI water to fully swell and the equilibrium water content (EWC) and the swelling ratio (SR) of the hydrogels at different time intervals were calculated by the following equations.

EWC = Mwet – Mdry/Mwet × 100 (1)

SR = Msw – Mdry/Mdry× 100 (2)

Where “Mwet”is the mass of as prepared hydrogel sample, “Mdry” is the mass of dry hydrogel sample and “Msw” is the mass of swollen hydrogel samples.

# Tensile and compression tests

A universal testing machine (CMT4501) were used for both compression and tensile stress-strain tests for original and healed samples. Cylindrical shape samples with a 15 mm in diameter and 10 mm in height were used for measuring the compression strength with a fixed cross-head speed at 2.0 mm/min. Rectangular shape samples with a specific dimension (3 × 6 × 35 mm) were used for tensile stretching tests, and the cross-head speed was fixed at 60 mm/min. All the tests were recorded at room temperature without any external interventions. The tensile stress (σ) of the original and healed samples were calculated by using the formula:

σ = F/A

Where “F” is the load, and “A” is the sectional area

While the tensile elongation strain of the original and healed samples was calculated by using

ε = Δl/lo

Here, “Δl” is the change in length, and “lo” is the original length of the tested sample.

Similarly, the compressive stress σc were recorded by using the formula:

σc = load/πr2

The strain εc under compression was recorded by using the formula:

εc = (ho – h)/ ho,

Here, “r” represents the radius, “h” change in height and “ho” is the initial height of the tested sample.

Toughness was calculated by the area under the stress-strain curves.

# The percent growth ratio (% GR)

The percent healing growth was calculated by the following equations:

% GR = T2 – T1/T1 × 100

Here “T1” and “T2” represent the tensile strength of the initial and final hydrogel samples, respectively.

# Self-healing efficiency

Mechanical self-healing efficiency (SHE) was quantified as the proportion strength restored relative to the original strength of hydrogel. We calculate the SHE in stress, strain and toughness by using the following formula.

SHE = TS(H)/TS(O) × 100

Here “TSH” and “TSO” represent the tensile strength of the self-healed and original hydrogels, respectively.

# FTIR analysis

FTIR spectra were obtained using Nicolet 5700 spectrometer to confirm the formation of the hydrogel network as well as to confirm metal coordinated bonds. FTIR spectra of Gly and Gly/P(AA-co-AAm)-Fe3+ dry hydrogels were all taken at spectral scanning in the range of 4000-500 cm-1 with KBr.

# Electrical measurements tests:

The conductivity of the hydrogels was investigated with an electrochemical workstation (CH1660E) AC impedance spectrum over frequency ranging from 0.1 Hz to 100 Hz. The conductivity (σ, S/cm) of the hydrogel samples were calculated from the following formula.

σ = L/Rπr2

where L is the distance between adjacent electrodes, R is the resistance of hydrogels and r is the radius of the hydrogel samples.

The resistance changes of the hydrogels were measured by the same electrochemical instrument. The relative change of the resistance was calculated by the following formula:

ΔR/R₀ = (R- R₀)/R₀

Where R and R₀ are the resistance with and without applied strain respectively.

Electrochemical workstation CH1660E and CMT2103 electrochemical tensile testing machines were used to record the real-time electrical signals at different strains. A constant voltage of 2.0 V was used during the whole process and the real-time signal was recorded by using the amperometric i-t curve program. The hydrogel based sensor was connected to the electrochemical workstation through copper wires. For human motion detection, the hydrogels based sensor was directly attached on the volunteer’s finger, wrist, etc. to evaluate the sensing performance. The relative changes in resistance (ΔR = R-R₀/R₀), based on fixed applied voltage to the strain sensors and variation in the electrical signal under various strains were calculated by the Ohm’s Law (R = U/I).