Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.


# Supporting Information

# Hydrogels from natural egg white with extraordinary stretchability, direct-writing 3D printability and self-healing for fabrication of electronic sensor and actuator

*Qiang Chang,[1,2], Mohammad Ali Darabi[1,6], Yuqing Liu[1], Yunfan He[2], Wen Zhong[3], Kibret Mequanin[4], Bingyun Li[5], Feng Lu[2]*, Malcolm MQ Xing[1]**

1 Department of Mechanical Engineering, University of Manitoba, Children's Hospital Research Institute of Manitoba, Winnipeg, Manitoba, R3T 2N2, Canada.

2 Department of Plastic Surgery, Southern Medical University, Guangzhou, Guangdong, 510515, China.

3 Department of Biosystems Engineering, University of Manitoba, Winnipeg, Manitoba, R3T 2N2, Canada.

4 Department of Chemical and Biochemical Engineering, University of Western Ontario, London, Ontario, N6A 5B9, Canada

5 Department of Orthopedics, School of Medicine, West Virginia University, Morgantown, WV, 26506, USA.

6 Department of Bioengineering, University of California - Los Angeles, Los Angeles, California, 90095, USA

*Corresponding authors: malcolm.xing@umanitoba.ca, doctorlufeng@hotmail.com

## Results

| Amino Acids | Asp | Thr | Ser | Glu | Gly | Ala | Cys | Val | Met | Ile | Leu | Tyr | Phe | Lys | His | Arg | Pro |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Content g/100g | 1.20 | 0.53 | 0.81 | 1.61 | 1.11 | 0.68 | 0.25 | 0.76 | 0.66 | 0.59 | 0.97 | 0.45 | 0.69 | 0.80 | 0.26 | 0.69 | 0.43 |
| COOH $10^{-4}$mol | 1.8 | 0.44 | 0.77 | 2.19 | 0.54 | 0.77 | 0.21 | 0.65 | 0.44 | 0.45 | 0.74 | 0.25 | 0.42 | 0.55 | 0.17 | 0.40 | 0.37 |
| $NH_2$ $10^{-4}$mol | 0.90 | 0.44 | 0.77 | 1.09 | 0.54 | 0.77 | 0.21 | 0.65 | 0.44 | 0.45 | 0.74 | 0.25 | 0.42 | 1.10 | 0.17 | 0.80 | N/A |

Table S1. The total content of different amino acid and estimated molar mass of COOH and $NH_2$ groups in EW.

| Group | 1 | 2 | 3 | 4 | 5 |
|---|---|---|---|---|---|
| EW(mg/ml) | 50 | 62.5 | 70 | 62.5 | 62.5 |
| NaOH(mg/ml) | 7.5 | 7.5 | 7.5 | 10 | 12.5 |
| pH | 13.26 | 13.26 | 13.26 | 13.41 | 13.49 |

Table S2. The varied concentration of raw EW and NaOH used for EW hydrogel gelation.

| EW (mg/ml) | 62.5 | 62.5 | 62.5 | 62.5 | 62.5 | 62.5 | 62.5 |
|---|---|---|---|---|---|---|---|
| NaOH (mg/ml) | 2.81 | 3.75 | 5.63 | 7.50 | 9.38 | 12.38 | 14.06 |
| Gelation Time(s) | N/A | N/A | 500 | 100 | 25 | 23 | 19 |
| pH | 12.81 | 12.92 | 13.15 | 13.26 | 13.36 | 13.48 | 13.54 |

Table S3. Hydrogel based on fixed EW concentration and varied NaOH concentration for further clarifying the NaOH effect on the gelation.

| Center (Raw EW) | Assignment | Area (%) |
|---|---|---|
| 1624.33 | β-sheet | 8.42 |
| 1632.45 | β-sheet | 10.35 |
| 1637.22 | β-sheet | 13.99 |
| 1646.50 | random coils | 7.36 |
| 1652.12 | α-helix | 13.38 |
| 1660.64 | α-helix | 11.50 |
| 1670.94 | β-turns | 11.78 |
| 1682.56 | β-turns | 12.78 |
| 1692.65 | anti-parallel β-sheet | 10.45 |

Table S4. Peak center, assignments and relative area percentages of secondary structure of raw.

| Center (EW hydrogel) | Assignment | Area (%) |
|---|---|---|
| 1617.47 | β-sheet | 2.33 |
| 1627.13 | β-sheet | 7.32 |
| 1635.92 | β-sheet | 17.67 |
| 1644.92 | random coils | 15.16 |
| 1653.51 | α-helix | 18.02 |
| 1662.38 | α-helix | 13.77 |
| 1671.79 | β-turns | 11.53 |
| 1682.74 | β-turns | 9.62 |
| 1692.80 | anti-parallel β-sheet | 4.57 |

Table S5. Peak center, assignments and relative area percentages of secondary structure of EW hydrogel.

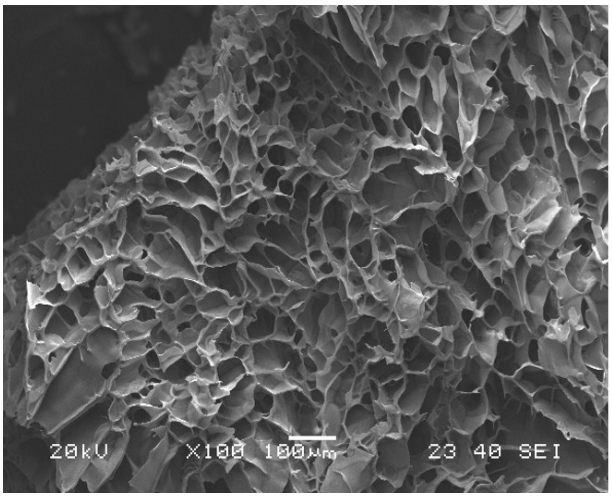


Figure S1. SEM images of EW hydrogel showed porous network structure.

Figure S2. The healed EW hydrogel could be separated into two parts quickly in urea solution.

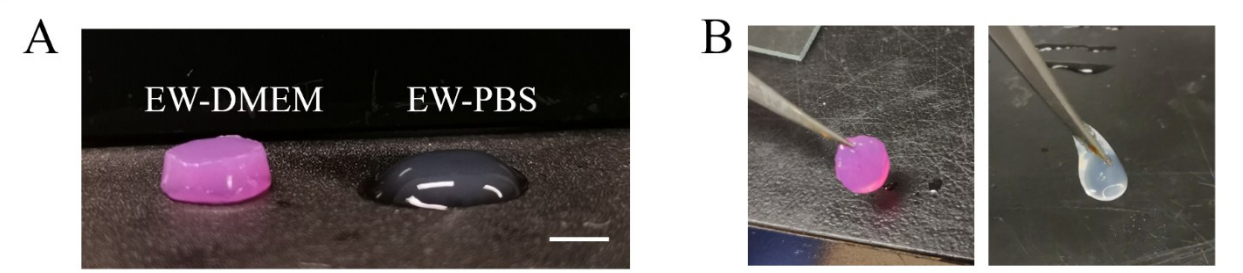


Figure S3. Representative photos of primary EW hydrogel treated with DMEM and PBS after 48 h. A) The front view of samples, hydrogels after both treatments could achieve shape integrity. B) The hydrogels could maintain the shape under gravity with DMEM treatment, however, the PBS-treated sample was stretched to drop shape under gravity.

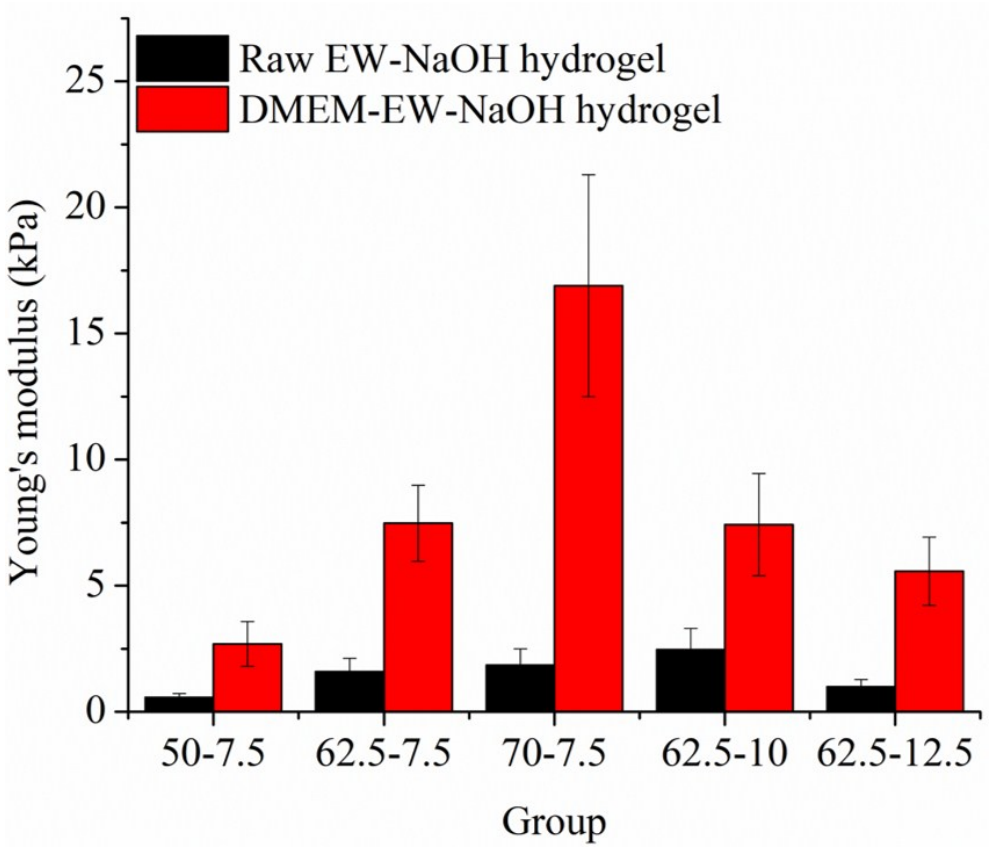


Figure S4. The histogram represented the Young's modulus of primary EW hydrogel and DMEM-treated hydrogel.

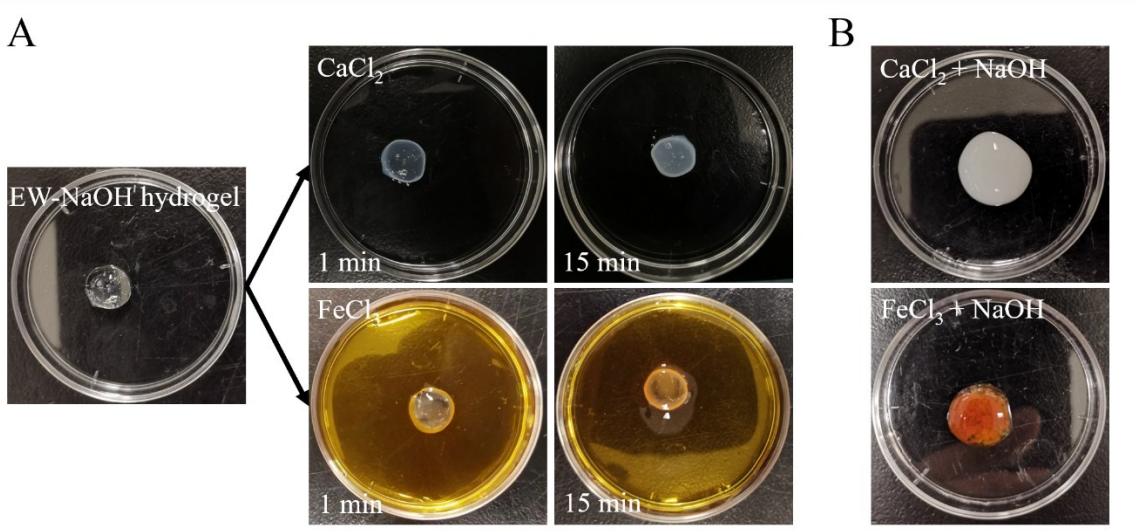


Figure S5. The morphology changes of secondary crosslinking of primary EW hydrogel with $CaCl_2$ and $FeCl_3$. A) The color changes of EW hydrogel after soaking in $CaCl_2$ and $FeCl_3$ (0.5 M) for 1 and 15 min, no visible precipitation was observed in either solution, which was totally different from the precipitation of $Ca(OH)_2$ and $Fe(OH)_3$ when mixed $CaCl_2$ and $FeCl_3$ directly with NaOH (B).

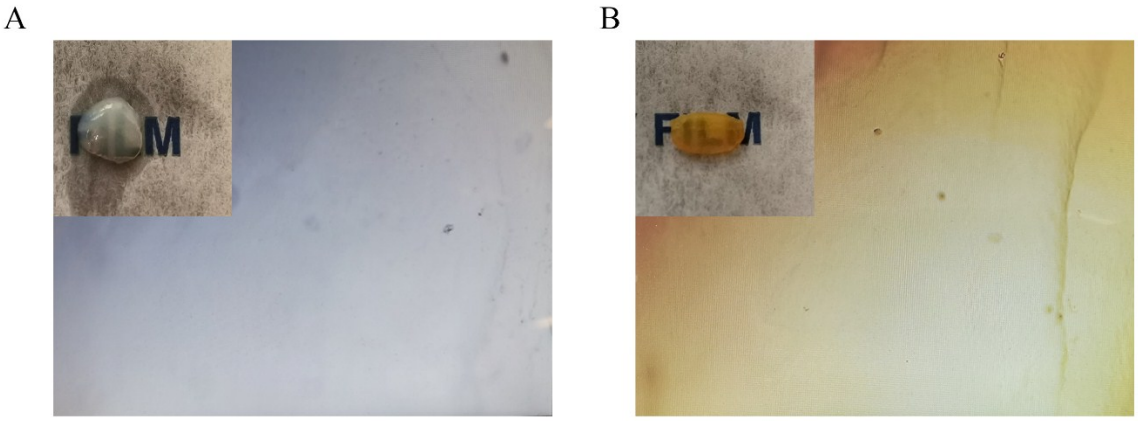


Figure S6. The microscopic view and photography demonstrated semi-transparent property of hydrogels treated after $CaCl_2$ (A) and $FeCl_3$ (B), the insets shown the hydrogel slice placed over the printed word.

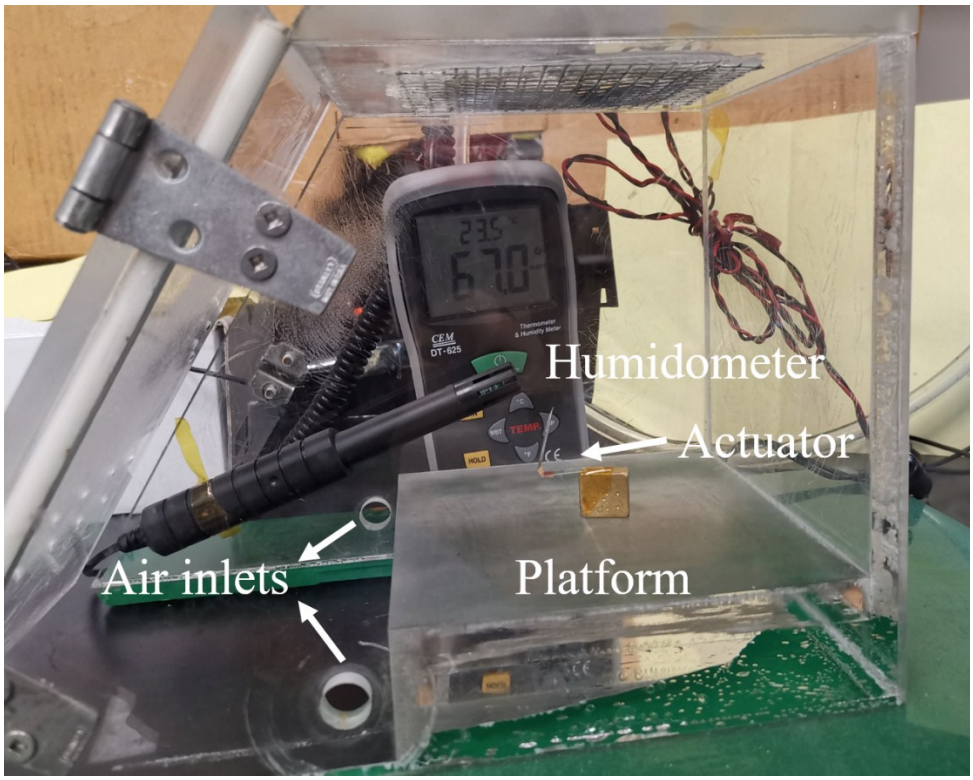


Figure S7. The customized humidity chamber for humidify-responsive EW actuator.

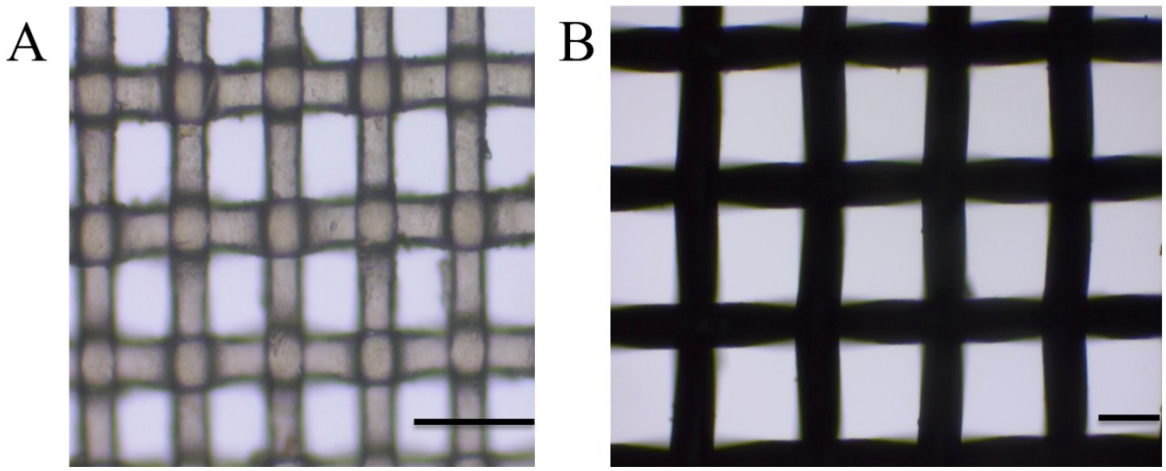


Figure S8. Optical microscope images of sieves with different size used for artificial hydrogel fiber manufacture, A) with a pore size of 80μm and B) with a pore size of 250μm, respectively. Scale bar=200μm.

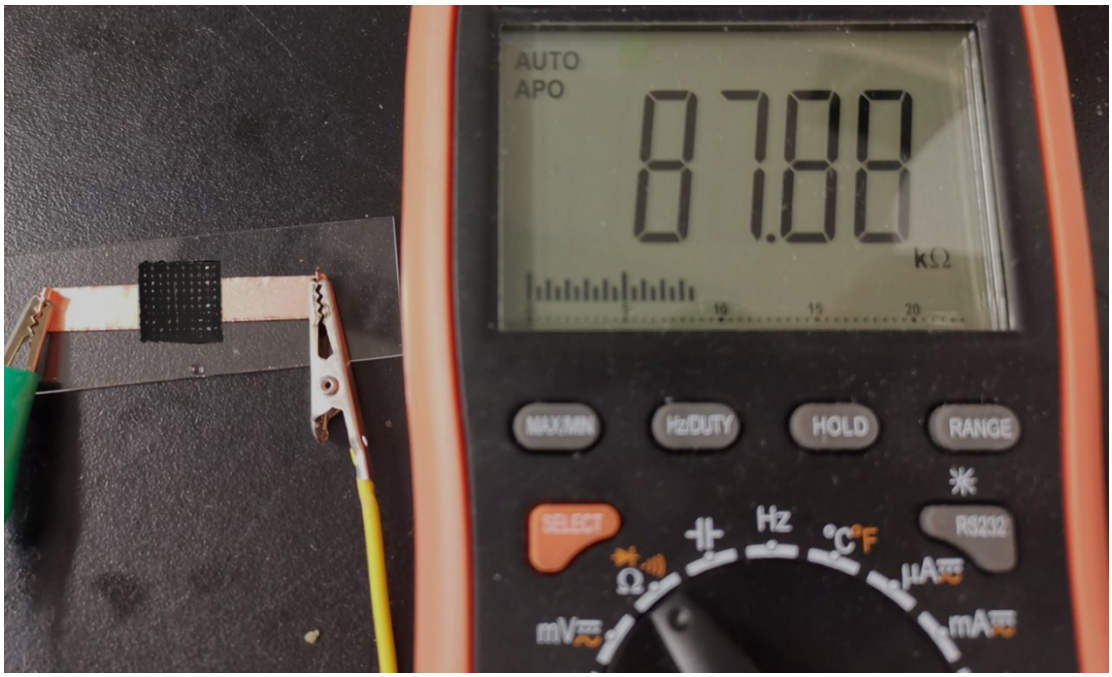


Figure S9. Resistance measurement of EW-CNT sensor.