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


Electronic Supplementary Material

# Role of high calcium ions in extending the properties of alginate dual-crosslinked hydrogels

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## Section 1: the control samples in the preparation of CaAG hydrogel

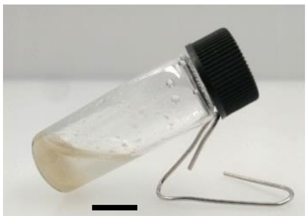

**Fig. S1** Mixture of alginate, borax, gelatin, and $CaCl_2$. Scale bar: 1 cm.

**Fig. S2** Mixture of ald-alginate, borax, gelatin, and $CaCl_2$. Scale bar: 1 cm.

## Section 2: the swelling and degradation of AG hydrogel

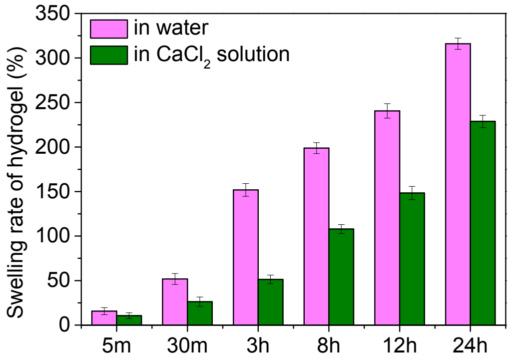


**Fig. S3** Swelling rate of AG hydrogel immersed in water or $CaCl_2$ solution for different times.

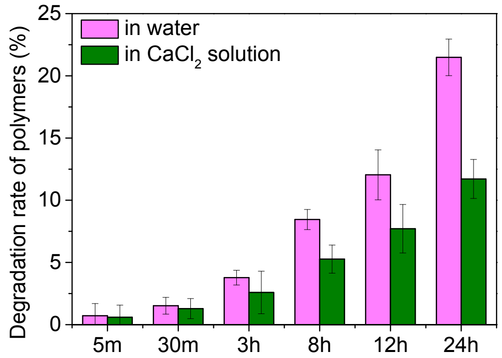


**Fig. S4** Degradation rate of AG hydrogel immersed in water or $CaCl_2$ solution for different times.

## Section 3: the tensile test of hydrogels

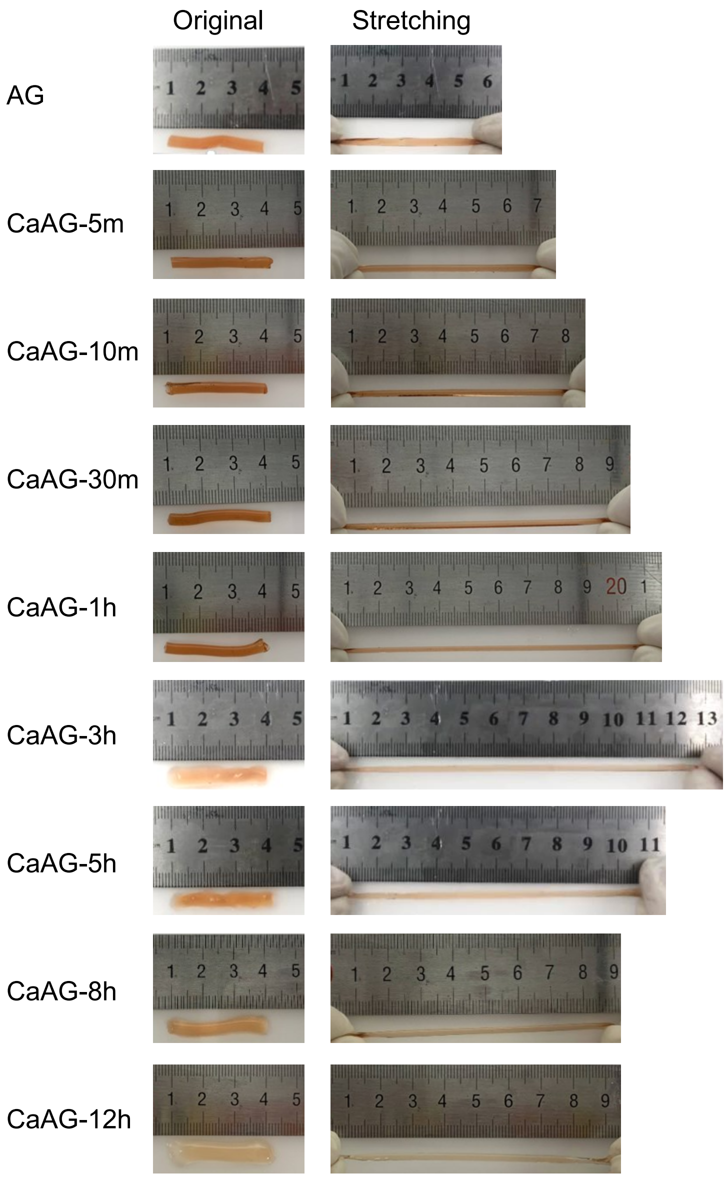


**Fig. S5** Images of hydrogels immersed in $CaCl_2$ solution for different times and their tensile test.

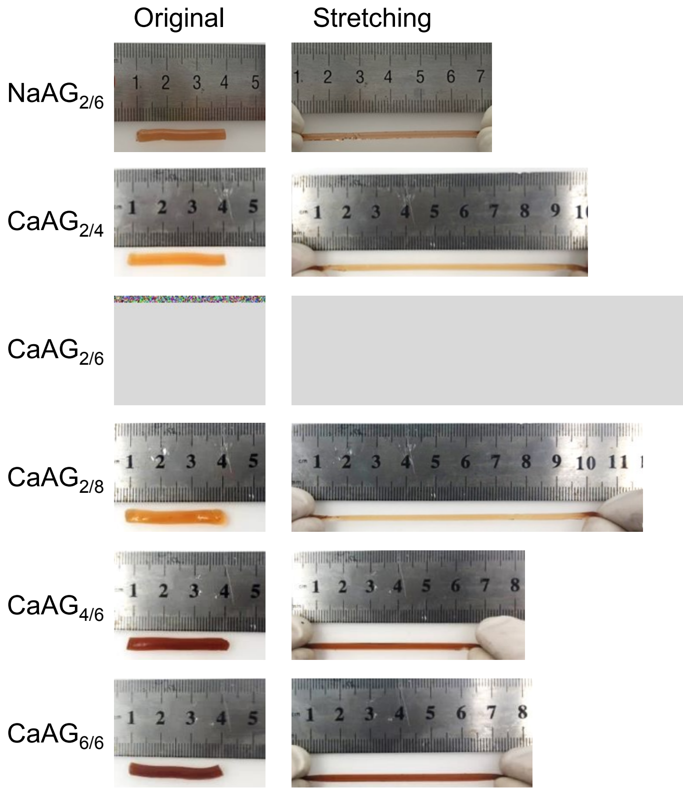


**Fig. S6** Images of hydrogels with different compositions immersed in $CaCl_2$ solution for 3 h and their tensile test.

## Section 4: self-healing test of hydrogels

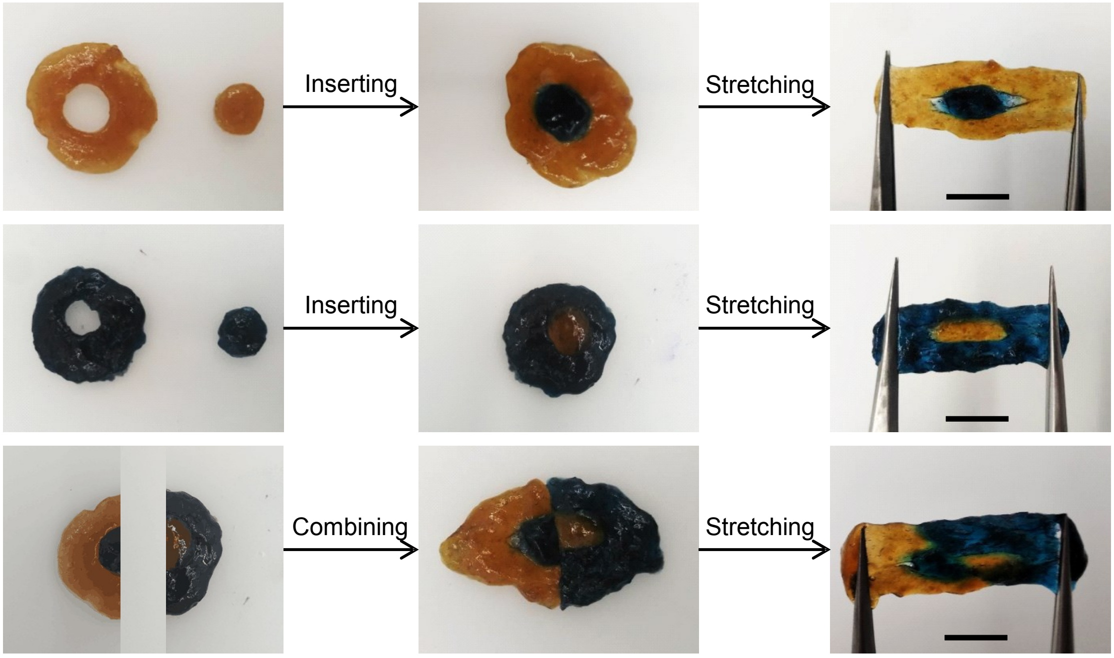


**Fig. S7** Self-healing of CaAG hydrogel. Scale bar: 1 cm.


**Fig. S8** Self-healing of CaAG hydrogel pieces. Scale bar: 1 cm.

## Section 5: the adhesion test of hydrogels

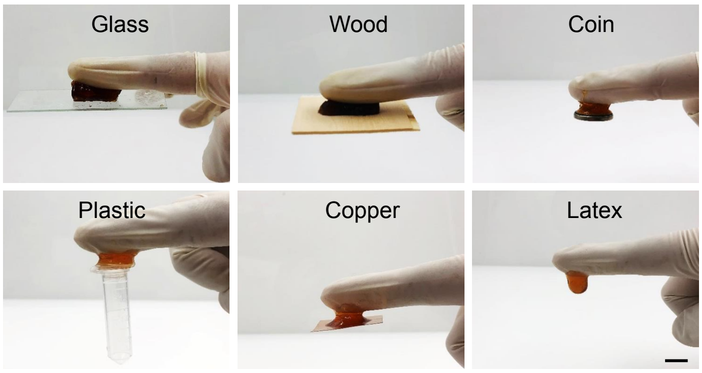


**Fig. S9** Hydrogel adhesion onto different substrates.

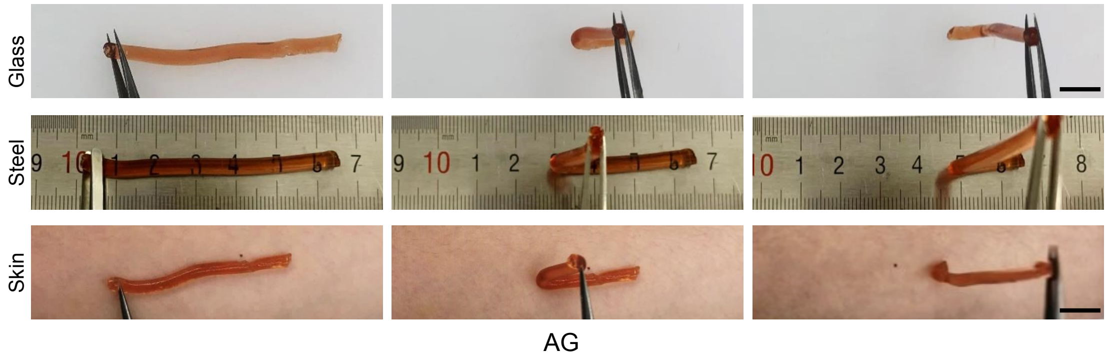


**Fig. S10** Peeling process of AG hydrogel onto various substrates (glass, steel, and skin).

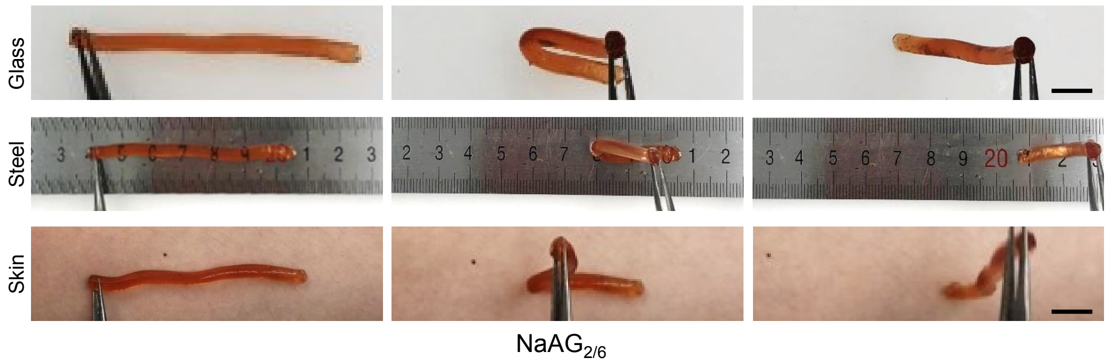


**Fig. S11** Peeling process of NaAG hydrogel onto various substrates (glass, steel, and skin).

## Section 6: the cytotoxicity assay of hydrogels

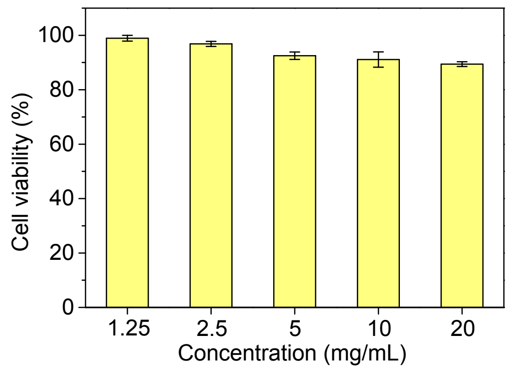


**Fig. S12** Relative cell viability of MCF-7 cultured with ald-alginate for 24 h as demonstrated by MTT assay (average of four measurements).

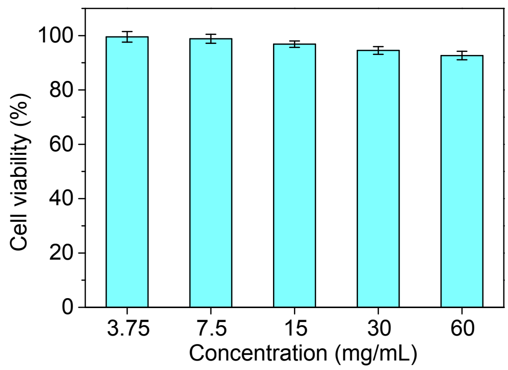


**Fig. S13** Relative cell viability of MCF-7 cultured with gelatin for 24 h as demonstrated by MTT assay (average of four measurements).

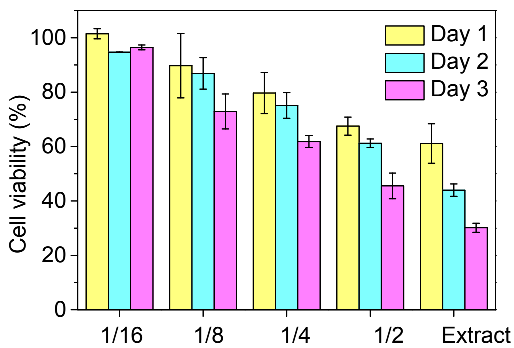


**Fig. S14** Relative cell viability of MCF-7 cultured with CaAG hydrogel extract for 24 h as demonstrated by MTT assay (average of four measurements).

## Section 7: gauge factor of CaAG hydrogel as a strain sensor

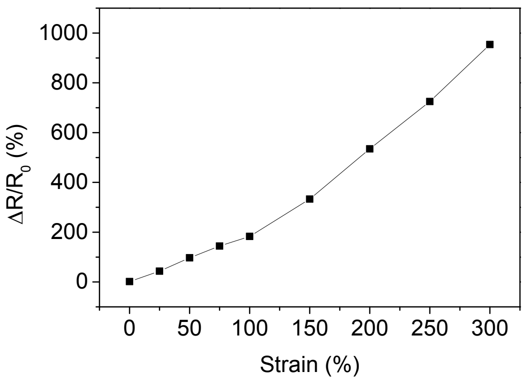


**Fig. S15** Gauge factor of CaAG hydrogel as a strain sensor.

## Section 8: self-healing and conductivity of CaAG hydrogels at −20 ℃

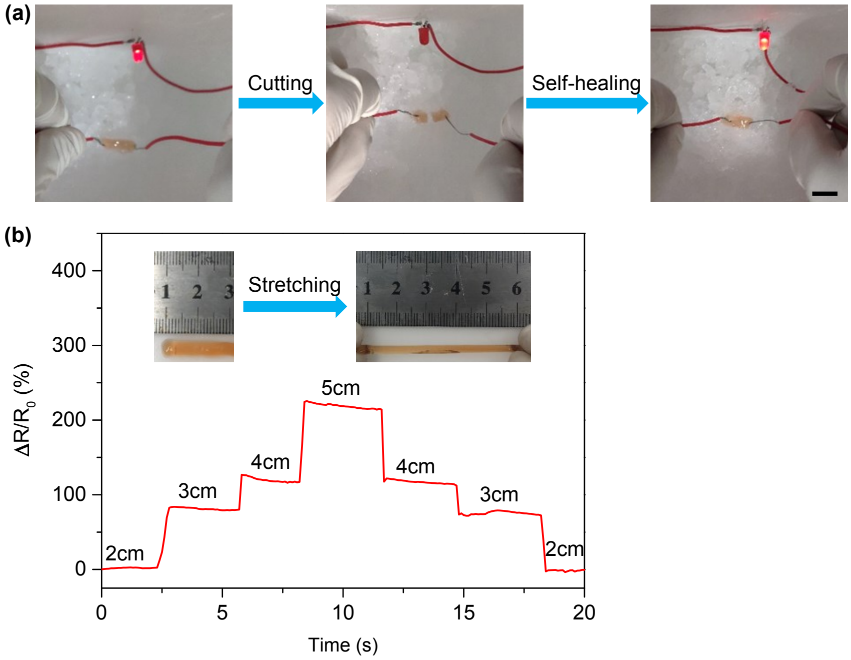


**Fig. S16** Self-healing and conductivity of CaAG hydrogels at −20 ℃. (a) LED light switching device at −20 ℃. LED lights up when CaAG hydrogel is connected. LED goes out when CaAG hydrogel is cut off. LED lights up again when CaAG hydrogel self-heals. (b) Relative resistance of CaAG hydrogel under different tensile conditions at −20 ℃.

## Section 9: the composition of hydrogels

**Table S1.** Hydrogels with different immersion times.

| Samples | Ald-alginate (wt/vol) | Gelatin (wt/vol) | Immersion time (37ºC) |
|---|---|---|---|
| AG | 2% | 6% | 0 min |
| CaAG-5m | 2% | 6% | 5 min |
| CaAG-10m | 2% | 6% | 10 min |
| CaAG-30m | 2% | 6% | 30 min |
| CaAG-1h | 2% | 6% | 1 h |
| CaAG-3h | 2% | 6% | 3 h |
| CaAG-5h | 2% | 6% | 5 h |
| CaAG-8h | 2% | 6% | 8 h |
| CaAG-12h | 2% | 6% | 12 h |

**Table S2**. Hydrogels with different ratios of ald-alginate and gelatin.

| Samples | Ald-alginate (wt/vol) | Gelatin (wt/vol) | Immersion time (37ºC) |
|---|---|---|---|
| $NaAG_{2/6}$ | 2% | 6% | 3 h |
| $CaAG_{2/4}$ | 2% | 4% | 3 h |
| $CaAG_{2/6}$ | 2% | 6% | 3 h |
| $CaAG_{2/8}$ | 2% | 8% | 3 h |
| $CaAG_{4/6}$ | 4% | 6% | 3 h |
| $CaAG_{6/6}$ | 6% | 6% | 3 h |