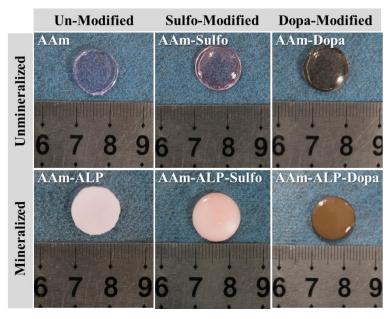
## Dopamine enhances the mechanical and biological properties of the

## enzyme-induced mineralized hydrogel

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### **Supplemental Data 1:**



#### Photographs of different hydrogels

Fig. S1 Photographs of different hydrogels.

Fig. S1 shows the photographs of the obtained hydrogels in the absence and presence of enzymatic mineralization. The hydrogels appeared transparent before biomineralization (top) and became white and opaque after biomineralization (bottom), indicating the occurrence of biomineralization. The hydrogels biofunctionalized with dopamine solution turned to deep brown in color. Compared to the AAm-ALP mineralized sample, it was worth noting that the AAm-ALP-Dopa mineralized hydrogel had a small shrinkage after dopamine biofunctionalization, while the AAm-ALP-Sulfo mineralized hydrogel remained the almost same size after Sulfo-SANPAH biofunctionalization.

# Supplemental Data 2: High magnification SEM micrographs of dopamine biofunctionalized hydrogels.

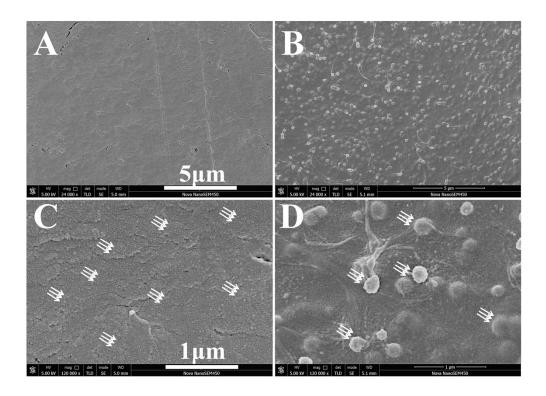


Fig.S2 High magnification SEM micrographs of AAm-Dopa (A, C) and AAm-ALP-Dopa (B, D)

Fig. S2 shows the high magnification SEM micrographs of dopamine biofunctionalized hydrogels. The unmineralized hydrogel shows smooth surface (Fig.S2A) and dopamine nanoparticles with diameter around tens of nanometers were observed on the surface (Fig.S2C, white arrow). The mineralized hydrogel shows mineral particles distributed on the hydrogel (Fig.S2B) and dopamine nanoparticles covered not only the surface of hydrogel but also mineralized particles (Fig.S2D, white arrow). The high magnification SEM micrographs of dopamine biofunctionalized hydrogels indicating that the dopamine was successful grafted to the hydrogels.

#### Supplemental Data 3: High resolution XPS Ca2p and P2p spectra of different hydrogels

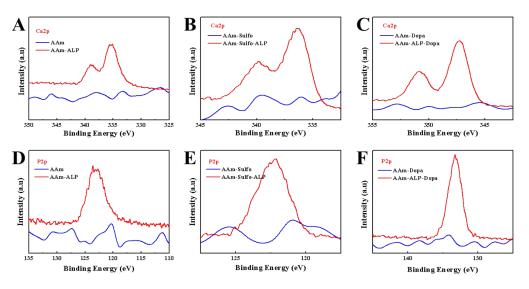


Fig. S3 Ca2p and P2p high resolution spectra of different hydrogels.

Fig. S3 shows the Ca2p and P2p high resolution spectra of different hydrogels. The mineralized samples showed clear presence of Ca and P elements, indicating the occurrence of the mineralization.

#### Supplemental Data 4: The chemical bond ratios of different hydrogels

Table 51. The chemical bond ratios of unferent hydrogets						
Ratio (%)	C=C	С-С/С-Н	C-N/C- NH/C-NH <sub>2</sub>	C- <b>S</b>	C-OH/C- O-C	C=O
AAm	22.59	58.78	8.11	-	-	10.49
AAm-Sulfo	17.35	36.42	<b>30.8</b> 7	5.91	-	9.44
AAm-Dopa	20.56	30.13	<b>19.1</b> 7	-	17.97	12.14

Table S1. The chemical bond ratios of different hydrogels

Table S1 shows the chemical bond ratios of different hydrogels. Compared with the pristine AAm hydrogel, additional C-S bond was observed in the AAm-Sulfo hydrogel due to the grafted Sulfo-SANPAH. The dopamine modified hydrogel showed the new peaks of C-OH and C-O-C bonds, as a result of dopamine self-polymerization.

#### Supplemental Data 5: The swelling ratios of AAm and AAm-ALP hydrogels

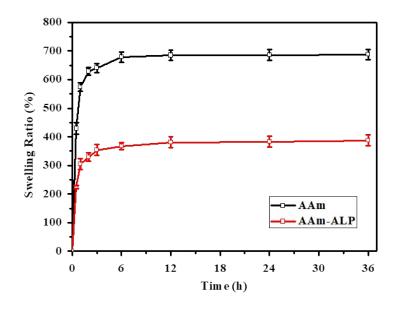


Fig. S4 The swelling ratios of AAm and AAm-ALP hydrogels

Fig. S4 shows the swelling ratios of the AAm and AAm-ALP hydrogels. Both AAm and AAm-AlP hydrogels almost reached maximum water absorption within 6 hours. The AAm-ALP hydrogel displayed a lower swelling ratio than AAm hydrogel. The AAm hydrogels reached maximum water absorption  $\approx 688\%$  of their initial weight after 36h. The lower water absorption of about 387% was observed for AAm-ALP hydrogel.

#### Supplemental Data 6: The statistical results of cell number

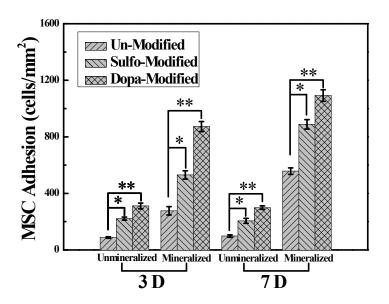


Fig. S5 Cell counting after 3 and 7 days of cell cultured on the different hydrogels Fig. S5 shows the cell counting results after 3 and 7 days of cell culture. Significant

improvements in cell number were observed for mineralized hydrogels at 3<sup>rd</sup> day and 7<sup>th</sup> day. In particular, the dopamine modified mineralized hydrogel showed the highest cell number among all hydrogel samples.

### Supplemental Data 7: The statistical results of cell spreading areas

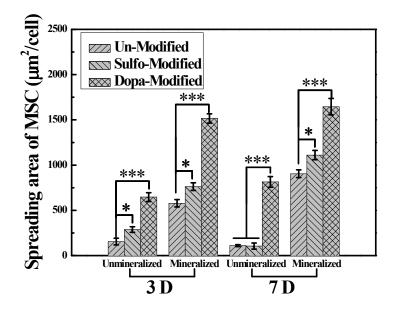


Fig. S6 The statistical results of cell spreading area after 3 and 7 days of cell cultured on the different hydrogels

Fig. S6 shows the statistical results of cell spreading area after 3 and 7 days of cell culture. The MSCs on the dopa-modified hydrogels showed increased spreading area compared with the cells on the un-modified and Sulfo-modified hydrogels in the absence and presence of mineralization. Overall, the dopamine biofunctionalized hydrogel showed the best spreading among all samples.