

## Supplementary Information

### Injectable chondroitin sulfate grafted self-antioxidant hydrogel ameliorates nucleus pulposus degeneration against overactive inflammation

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Table S1. Primers sequences used in RT-qPCR

Gene	Primer sequence (5'-3')
TNF- $\alpha$	F: GACAAGCCTGTAGCCCATGT
	R: CTCTGATGGCACCACCAACT
IL-6	F: TTCGGTCCAGTTGCCTTCTC
	R: GCCTCTTTGCTGCTTTCACA
MMP3	F: CACTCACAGACCTGACTCGG
	R: AGTCAGGGGGAGGTCCATAG
MMP13	F: TCCTGATGTGGGTGAATACAATG
	R: GCCATCGTGAAGTCTGGTAAAAT
ADAMTS5	F: GCCTCTCCCATGACGATTCC
	R: TCGTGGTAGGTCCAGCAAAC
COL II	GGTAAGTGGGGCAAGACTGTTA
	TGTTGTTTCTGGGTTCAGGTTT
ACAN	GTCAGATACCCCATCCCACTC
	CATAAAAGACCTCACCTCCAT
GAPDH	F: GAAGGTCGGAGTCAACGG
	R: GGAAGATGGTGATGGGATT

Table S2. Histological grades of IVD

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Histological grades IVD
I. Cellularity of the annulus fibrosus
Grade:
1. Fibroblasts comprise >75% of the cells
2. Neither fibroblasts nor chondrocytes comprise >75% of the cells
3. Chondrocytes comprise >75% of the cells
II. Morphology of the annulus fibrosus
Grade:
1. Well-organized collagen lamellae without ruptured or serpentine fibers
2. Inward bulging, ruptured or serpentine fibers in less than one third of the annulus
3. Inward bulging, ruptured or serpentine fibers in more than one third of the annulus
III. Border between the annulus fibrosus and nucleus pulposus
Grade:
1. Normal, without any interruption
2. Minimal interruption
3. Moderate or severe interruption
IV. Cellularity of the nucleus pulposus
Grade:
1. Normal cellularity with stellate shaped nuclear cells evenly distributed throughout the nucleus
2. Slight decrease in the number of cells with some clustering
3. Moderate or severe decrease (>50%) in the number of cells with all the remaining cells clustered and separated by dense areas of proteoglycans
V. Morphology of the nucleus pulposus
Grade:
1. Round, comprising at least half of the disc area in midsagittal sections
2. Rounded or irregularly shaped, comprising one quarter to half of the disc area in midsagittal sections
3. Irregularly shaped, comprising less than one quarter of the disc area in midsagittal sections

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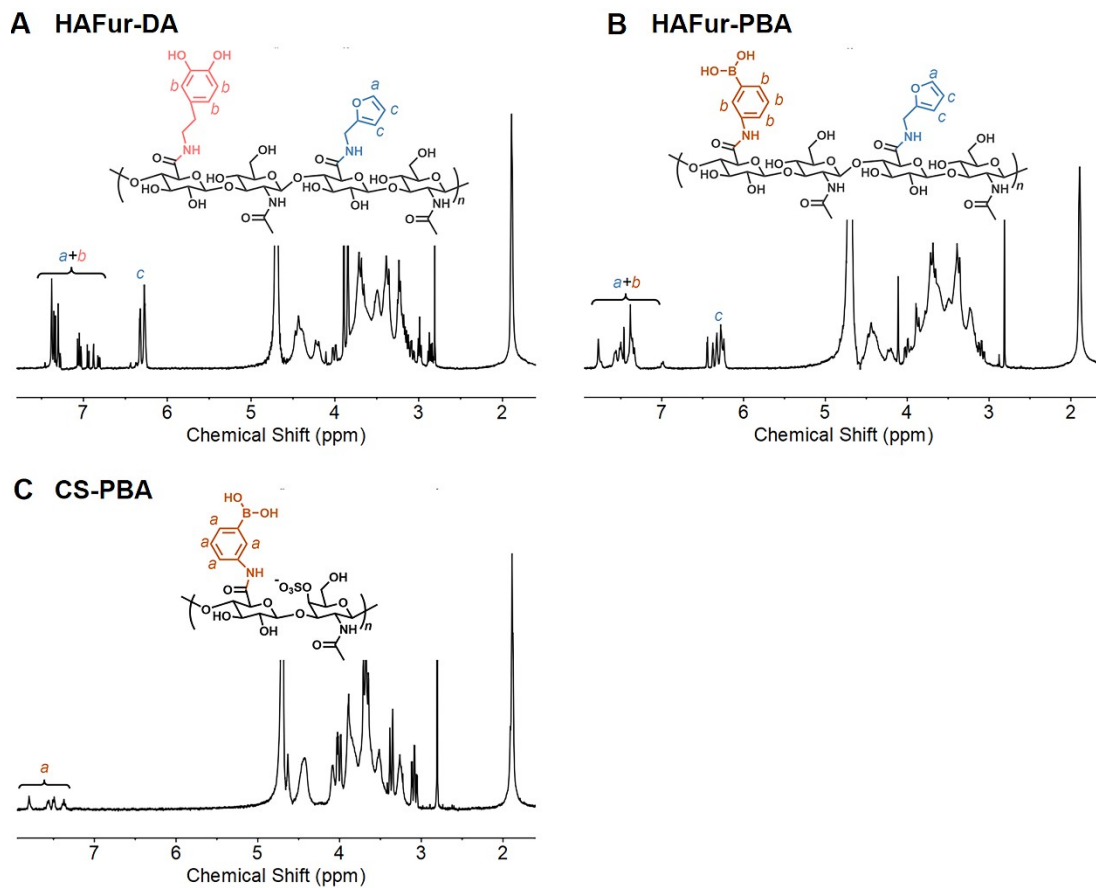


Fig. S1  $^1\text{H}$  NMR spectra of (A) HAFur-DA, (B) HAFur-PBA and (C) CS-PBA.

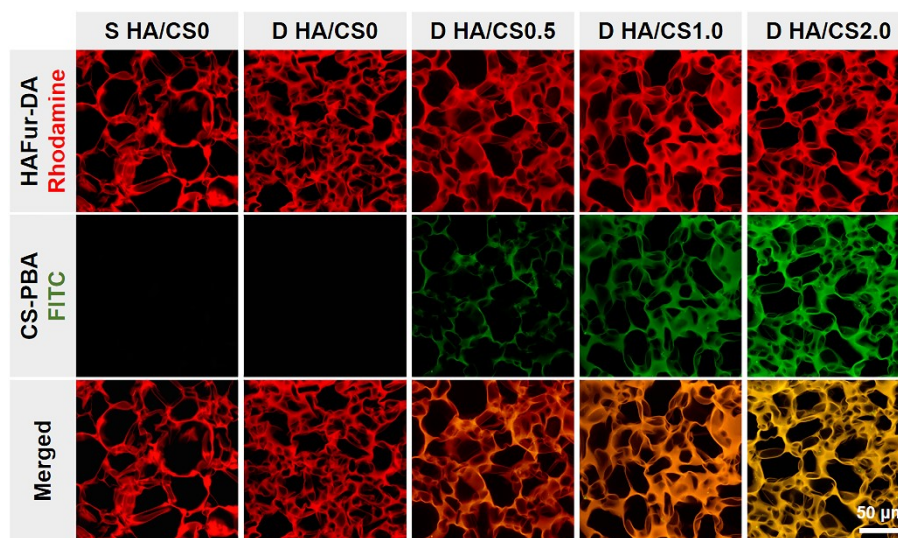


Fig. S2 CLSM images of single crosslinked and double crosslinked hydrogels (HAFur-DA is labeled by Rhodamine and CS-PBA is labeled by FITC).

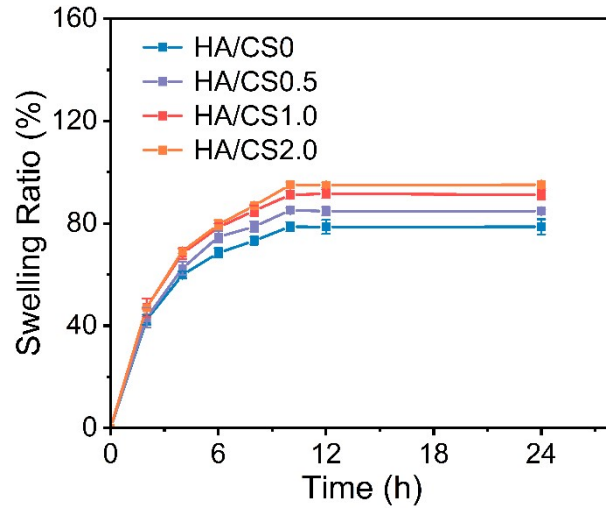


Fig. S3 Swelling ratio of double crosslinked hydrogels.

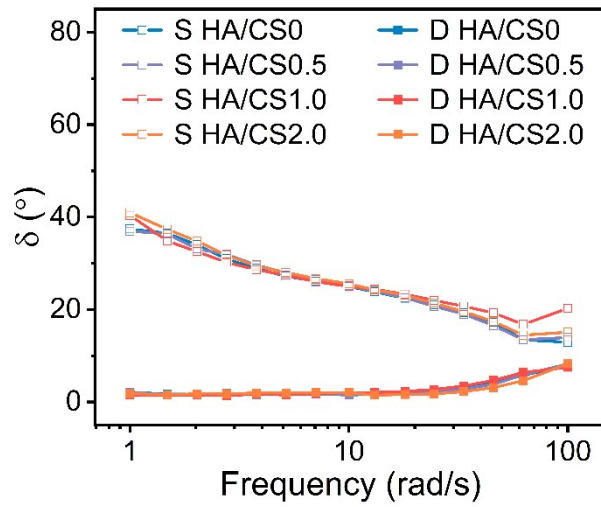


Fig. S4 Phase shift angle ( $\delta$ ) of different hydrogels under frequency sweep (1-100 rad/s) with constant strain ( $\gamma = 1\%$ ).

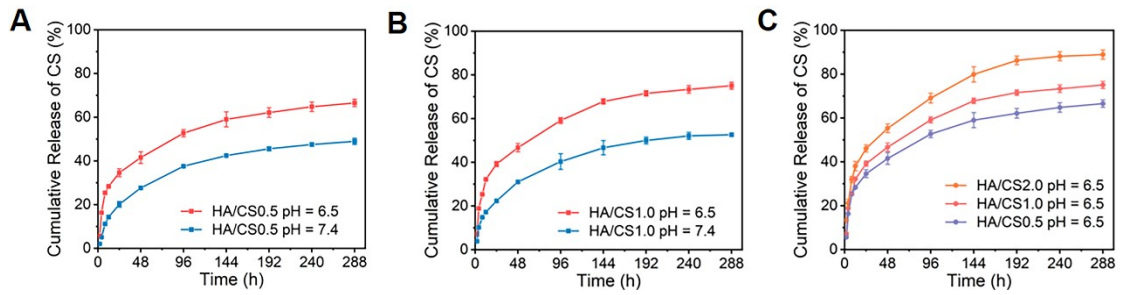


Fig. S5 CS release behavior of double crosslinked HA/CS hydrogels.

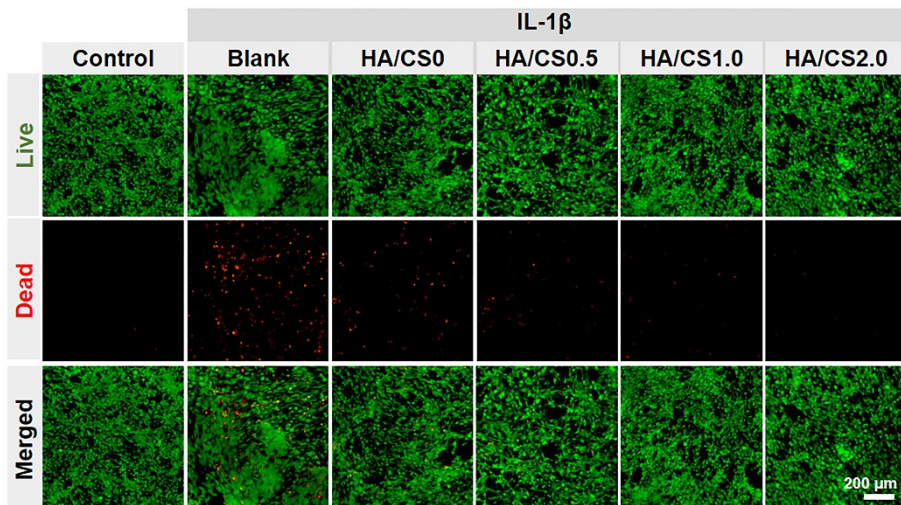


Fig. S6 Live/Dead staining of NPCs exposed to IL-1 $\beta$  with or without hydrogel treatment.

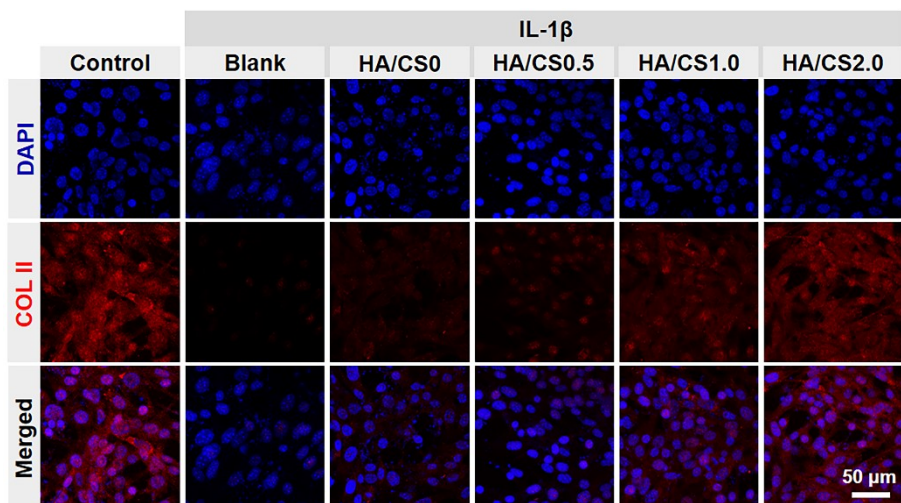


Fig. S7 Immunofluorescence staining of COL II in NPCs with different treatments.