Supporting Information

Double Network Hydrogels that Mimic the Modulus, Strength and Lubricity of Cartilage

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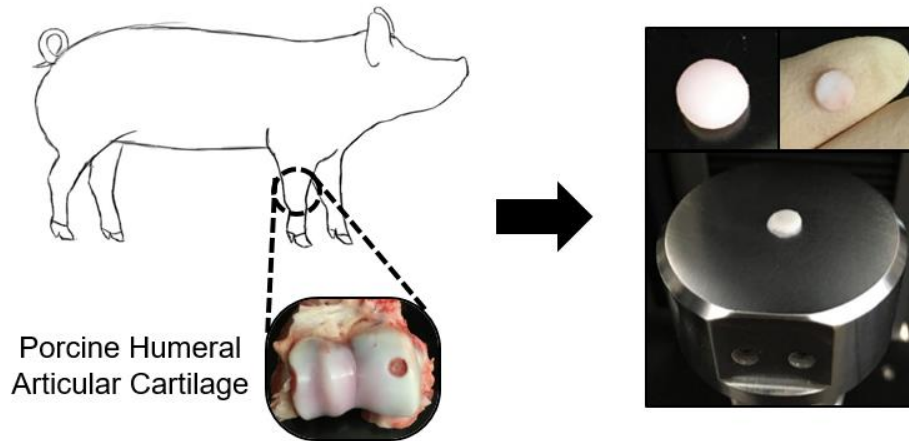KEYWORDS: hydrogels, double network, cartilage, ultra-strong, load-bearing

Figure S1. Compression of harvested porcine articular cartilage specimen.
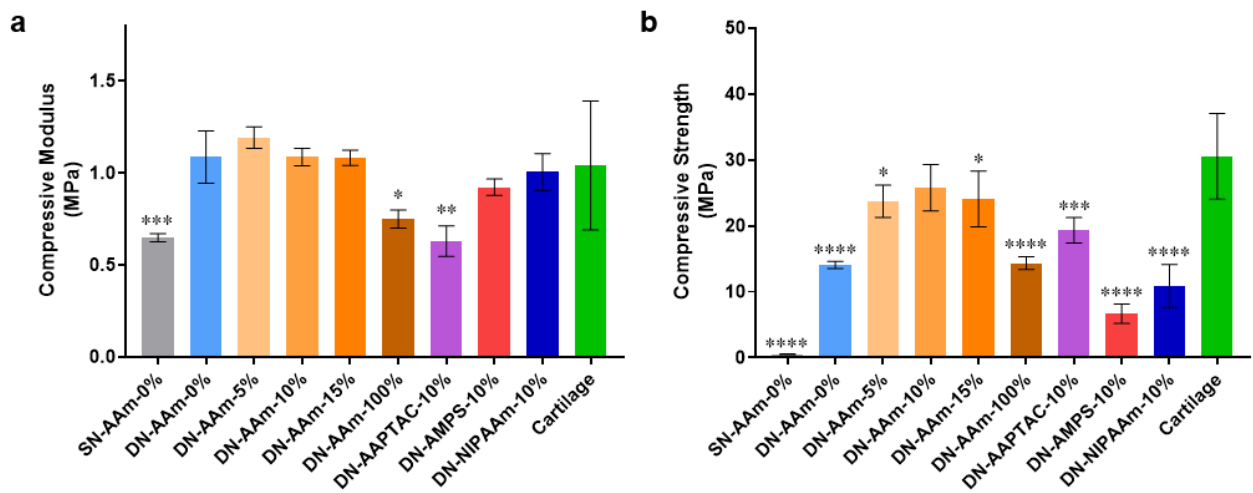

Figure S2. Compressive (a) modulus and (b) strength of additional controls compared to porcine articular cartilage. All *'s indicate statistical significance from cartilage, in which "*" represents $p < 0.05$, "**" represents $p < 0.01$, "***" represents $p < 0.001$ and "****" represents $p < 0.0001$.
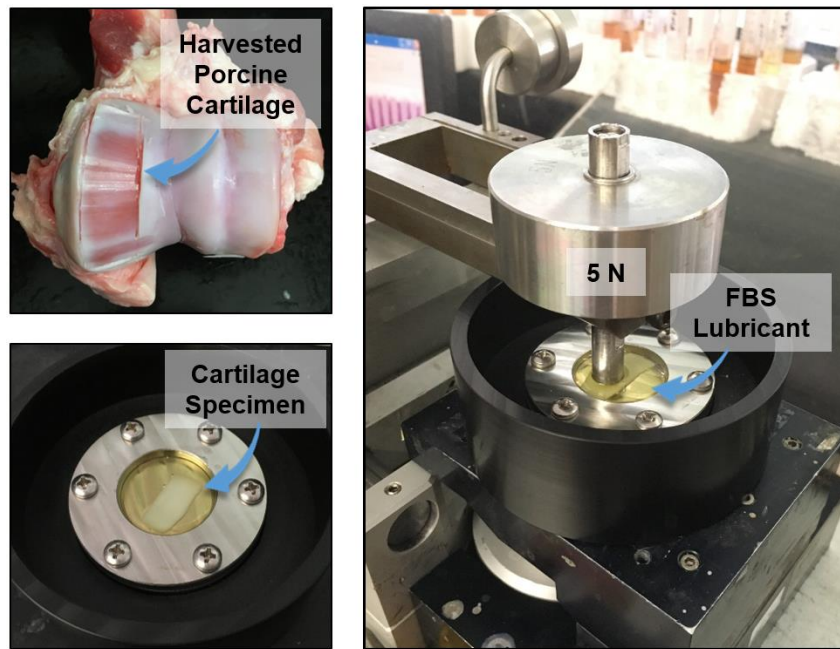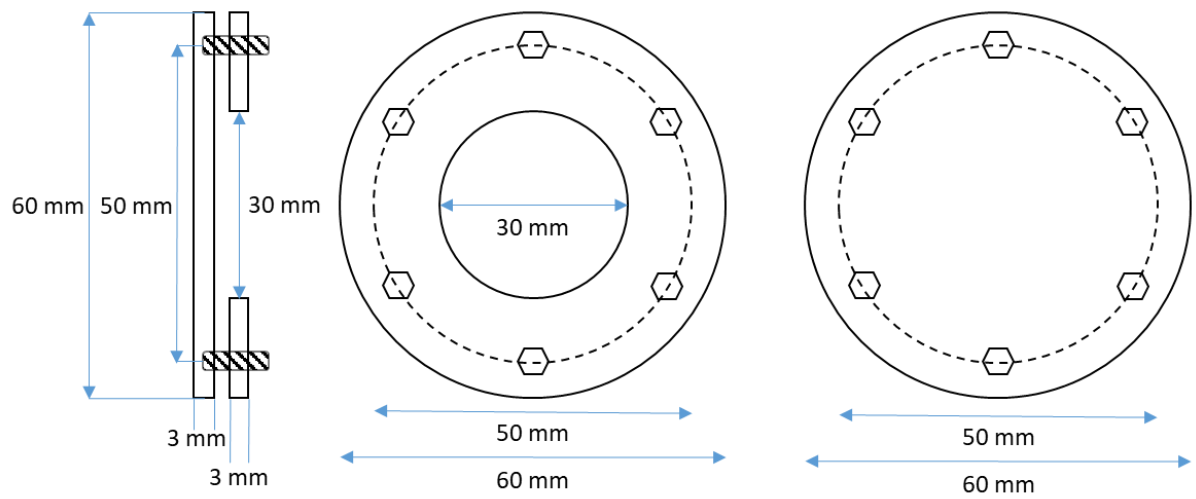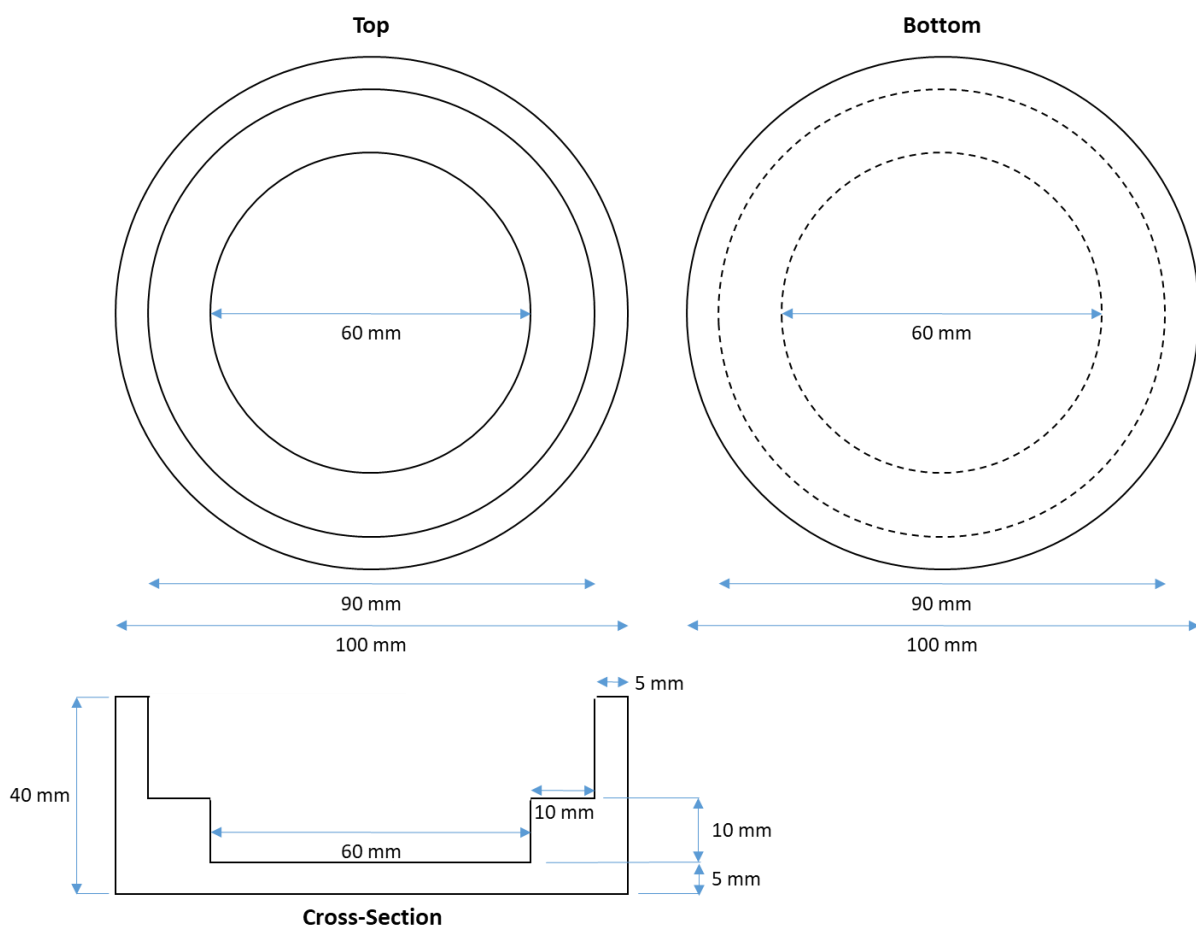
Figure S3. Friction testing (tribology) of harvested porcine articular cartilage.

Scheme S1. Friction testing (tribology) specimen clamp drawing.

Scheme S2. Friction testing (tribology) submersion chamber drawing.

Table S1. DN hydrogels detailing 1st network and 2nd network compositions as well as their thermal transitions (VPTTs).

Hydrogel Type	Composition			VPTT	
	1st Network*	2nd Network+		T_o(°C)	T_{max}(°C)
	AMPS (X)	NIPAAm	AAm (Y) (wt% of NIPAAm)		
PAMPS SN:					
SN-AAm-0%	1.5M	-	-	-	-
PAMPS/PNIPAAm DN:					
DN-AAm-0%	1.5 M	2.0 M	0 wt%	32.8 ± 0.17	35.4 ± 0.22
PAMPS/(PNIPAAm-*co*-AAm) DNs:					
DN-AAm-5%	1.5 M	2.0 M	5 wt%	37.4 ± 0.33	41.6 ± 0.07
DN-AAm-10%	1.5 M	2.0 M	10 wt%	41.4 ± 0.68	48.0 ± 0.19
DN-AAm-15%	1.5 M	2.0 M	15 wt%	45.5 ± 0.25	52.6 ± 0.77

*4 mol% BIS crosslinker, 0.1 mol% 2-oxoglutaric acid initiator
+0.1 mol% BIS crosslinker, 0.1 mol% 2-oxoglutaric acid initiator

Table S2. Overall mechanical properties of the PAMPS/PNIPAAm-*co*-AAm) hydrogel series, including equilibrium water content; tensile modulus, strength and fracture strain; and compressive modulus, strength, fracture strain and toughness.

	EWC	Tensile Properties			Compressive Properties			
	(%)	E (MPa)	σ (MPa)	ε (%)	E (MPa)	σ (MPa)	ε (%)	U_t (MJ m-3)
PAMPS SN:								
SN-AAm-0%	96.55 ± 0.00	0.33 ± 0.046	0.04 ± 0.02	10.8 ± 4.1	0.65 ± 0.02	0.54 ± 0.1	34.4 ± 0.8	0.07 ± 0.01
PAMPS/PNIPAAm DN:								
DN-AAm-0%	86.3 ± 0.17	1.00 ± 0.04	1.61 ± 0.04	147.8 ± 9.2	1.09 ± 0.14	14.1 ± 0.5	78.8 ± 1.8	2.63 ± 0.07
PAMPS/(PNIPAAm-*co*-AAm) DNs:								
DN-AAm-5%	83.9 ± 0.11	1.04 ± 0.05	1.57 ± 0.06	139.4 ± 13.7	1.19 ± 0.06	23.7 ± 2.4	86.9 ± 1.9	4.31 ± 0.48
DN-AAm-10%	83.9 ± 0.21	1.13 ± 0.06	1.54 ± 0.12	128.0 ± 20.5	1.09 ± 0.05	25.8 ± 3.5	90.8 ± 2.6	4.69 ± 0.68
DN-AAm-15%	83.5 ± 0.16	1.15 ± 0.09	1.53 ± 0.04	131.0 ± 13.1	1.08 ± 0.04	24.1 ± 4.2	89.9 ± 3.1	4.39 ± 0.80
Cartilage								
Porcine Cartilage	76.5 ± 2.12	-	-	-	1.04 ± 0.35	30.6 ± 6.5	77.8 ± 9.9	7.03 ± 2.37

EWC = equilibrium water content, E = elastic modulus, σ = stress, ε = strain, U_t = compressive toughness (deformation energy)