

Stretchable, Adhesive, and Biocompatible Hydrogel Based on Iron–Dopamine Complexes

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Synthesis of methacrylic-modified dopamine (DA)

Methacrylic-modified dopamine was synthesized following the protocol outlined in the literature [S1]. The synthesis involved the reaction of 3,4-dihydroxyphenethylamine hydrochloride with methacrylate anhydride in an aqueous solution of sodium borate and sodium bicarbonate under moderately basic conditions. The presence of aqueous sodium borate solution, serving as the reaction medium, played a role in protecting the dopamine moiety by forming a borate ester. The synthesized DA monomer was gained as a solid pale-gray powder with a yield of 14%. ¹H NMR (400 MHz, D₂O): δ 1.71 (s, 3H), 2.59 (t, J = 6.8 Hz, 2H), 3.31 (t, J = 6.8 Hz, 2H), 5.24 (s, 1H), 5.41 (s, 1H), 6.57 (dd, J = 2.0, 8.0 Hz, 1H), 6.65 (d, J = 2.0 Hz, 1H), 6.72 (d, J = 8.0 Hz, 1H).

Table S1 Physical properties of PAID hydrogels.^a

	AAM ^b	MBA ^b	FeCl ₃ · 6H ₂ O ^b	DA ^b	Appr. ^c	G', G'' (Pa)
PAID-0	10	0.06	-	0.18	TG	2.51×10 ² , 1.05×10 ²
PAID-1	10	0.06	0.06	0.18	YTG	1.39×10 ³ , 2.75×10 ²
PAID-2	10	0.06	0.06	0.36	YTG	6.99×10 ² , 1.53×10 ²
PAID-3	10	0.06	0.06	0.54	YTG	3.52×10 ² , 0.83×10 ²

^aIrgacure 2959 (2 mol % to AAM monomer), acrylamide is AAM, *N,N'*-methylene- bis-acrylamide is MBA, and methacrylic-modified dopamine is DA. ^bunit: % w/v; ^cTG: transparent gel; YTG: yellow transparent gel.

Table S2 Compares the tensile stress-strain performance between this work and previously reported hydrogels.

Materials	Strain (%)	Stress (kPa)	Ref.
AAM/MAH/Zn ²⁺	400	6000	S2
AAM/MBAA/Cu ²⁺	360	550	S3
Chitosan/PACG/Fe ³⁺	550	900	S4
PMA/Ca ²⁺	95	700	S5
MAA/QHPMA/Zn ²⁺	160	580	S6
PVA/EGaIn	350	115	S7
AAM/MBA/DA/Fe ³⁺	1011	8	This work (PAID-3)

Table S3 Compares the adhesive strength between this work and previously reported hydrogels. (substrate: glass)

Materials	Adhesive strength (kPa)	Ref
Aa(Ta)/HMA/AAM	0.09	S8
PAAM/ PAA/ GR/ PEDOT:PSS	2.9	S9
AAM/SA/DA	5.9	S10
PVA/FSWCNT/PDA	9.2	S11
PVA/PAA/PEDOT:PSS	10.97	S12
AA/ ZnCl ₂ /AR	10.4	S13
HACC/PAAM	14.4	S14
AAM/MBA/DA/Fe ³⁺	15.2	This work (PAID-3)

Table S4 Summarized t-test table of p-values for cell viability.

T-test	p-value (DAY 1)	p-value (DAY 3)	p-value (DAY 7)
PAID-0 vs Control	0.030	0.150	0.286
PAID-1 vs Control	0.129	0.103	0.048
PAID-2 vs Control	0.232	0.436	0.073
PAID-3 vs Control	0.194	0.098	0.064

P-values of <0.05 mean that there is a significant difference between the groups.

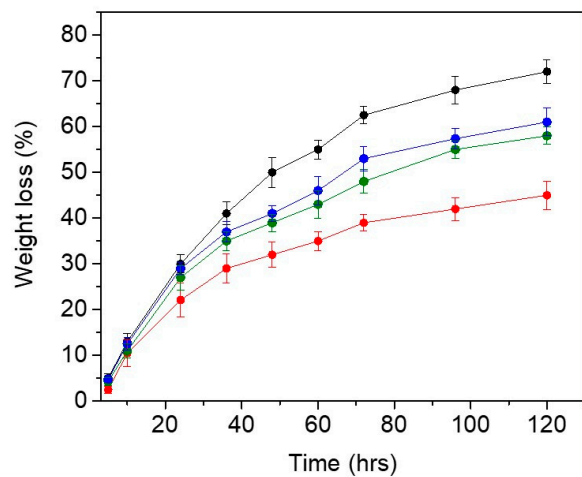


Figure S1: Degradation curves of PAID hydrogels. (Black for **PAID-0**, red for **PAID-1**, olive for **PAID-2** and blue for **PAID-3**)

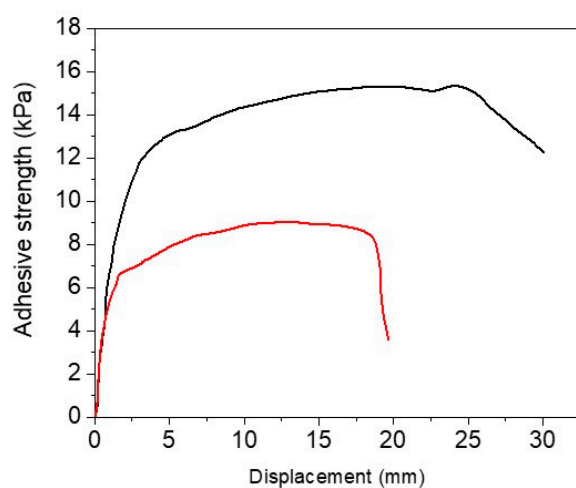


Figure S2: Lap-shear strength tests of the **PAID-3** hydrogel on different substrates. (Black: glass; Red: Al)

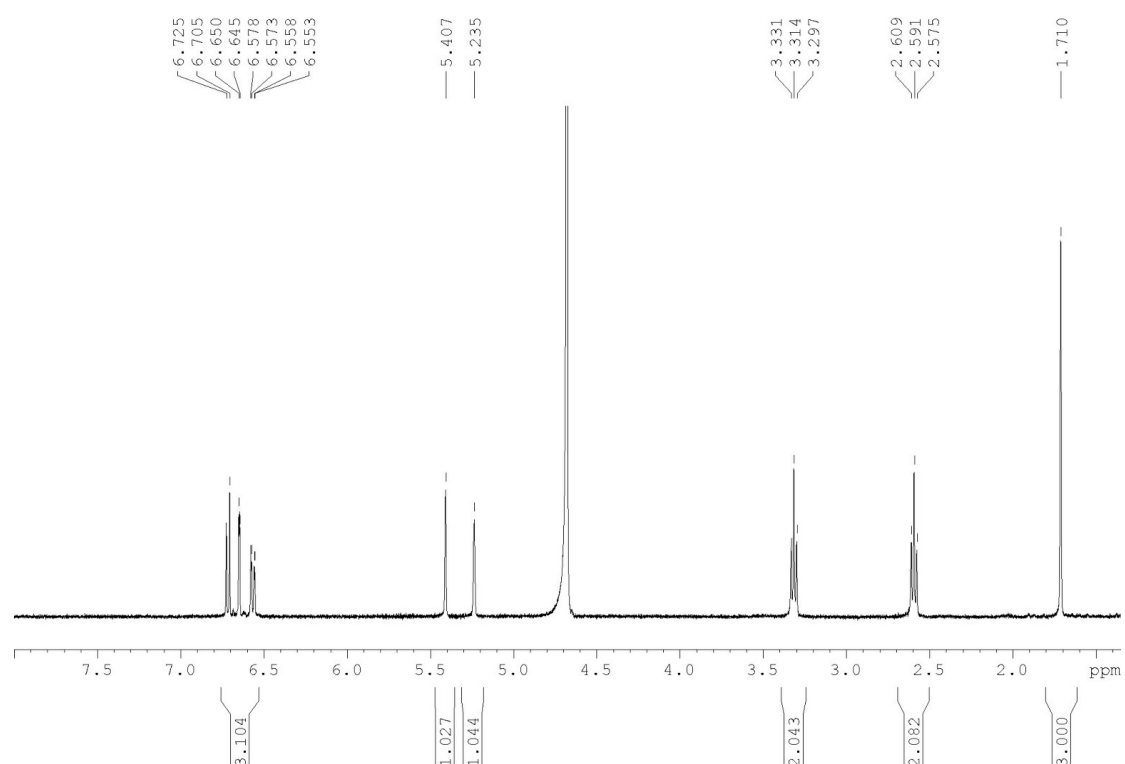


Figure S3: ¹H NMR spectrum of methacrylic-modified dopamine in D₂O.

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