

Supporting Information

Tunable enzymatically degradable hydrogels for controlled cargo release with dynamic mechanical properties

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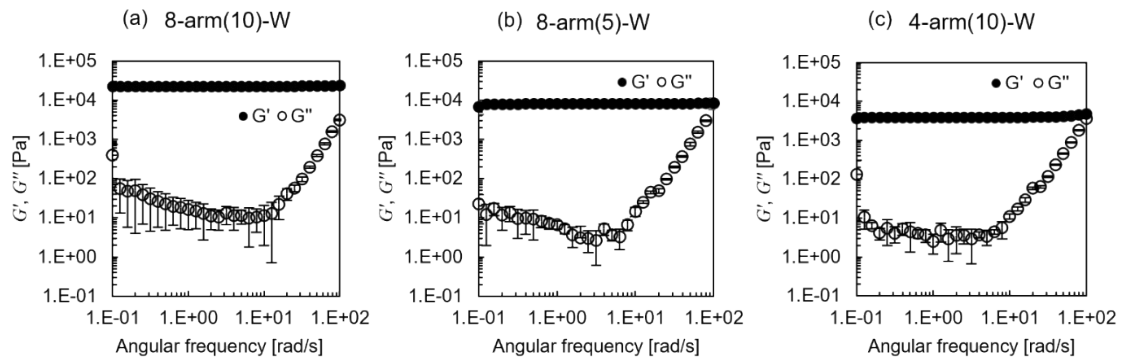


Figure S1 □ Frequency sweep conducted at 1% strain for swollen gels (before degradation) ($n=3$). Since G'' was negligible relative to G' in the low frequency region, most of the mechanical structure of the hydrogel is supported by G' . The value of G' was almost constant independent of frequency.

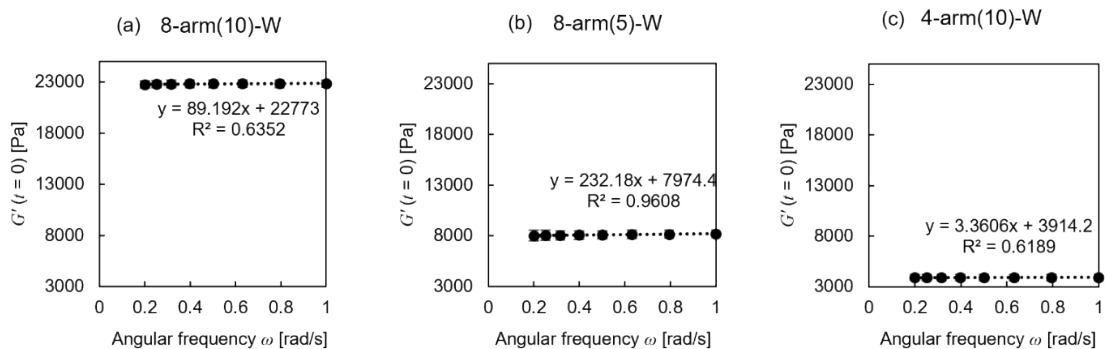
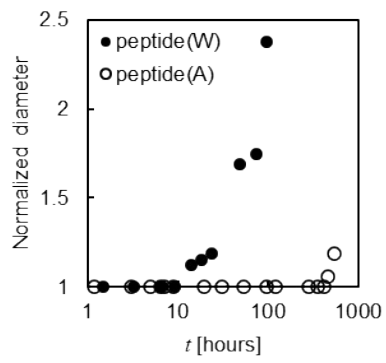


Figure S2 □ A linear fit of the storage modulus plateau ($n = 3$) was performed for each gel to calculate the zero-frequency storage modulus ($G'(t = 0, \omega = 0)$). The equation from the linear fit is displayed on each graph. The y-intercept from the equation was used as the $G'(t = 0, \omega = 0)$ to estimate the hydrogel mesh size.

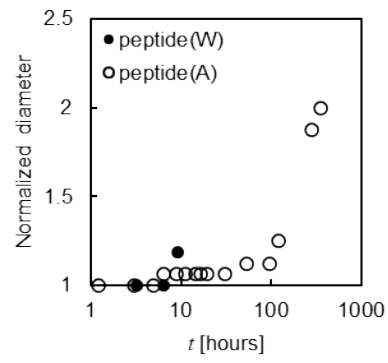
Table S1 □ Summary of mesh size for swollen initial hydrogels (ζ), and a kinetic constant of hydrogel's enzymatic degradation (k^*), and the kinetic constant (K_m) and diffusion or release exponent (n) of FD released because of hydrogel degradation.

	$\zeta(t=0)$ estimated from $G'(t=0, \omega=0)$ [nm]	$\zeta(t=0)$ estimated from Q_v [nm]	$k^* [M^{-1} \cdot s^{-1}]$	$K_m(t \geq 0)$	$n(t \geq 0)$
8-arm (10)-W	5.73±0.03	11.27±0.57	1.76±0.14	0.02±0.02	1.78±0.18
8-arm (10)-A		12.17±0.89		0.13±0.08	1.97±0.27
8-arm (5)-W	8.13±0.18	13.94±0.54	4.04±0.68	0.05±0.02	1.67±0.18
8-arm (5)-A		14.08±0.14		0.13±0.07	2.07±0.16
4-arm (10)-W	10.31±0.25	17.39±1.20	23.32±3.38	0.77±1.00	1.20±0.39
4-arm (10)-A	10.25±0.23	18.23±0.22	151.71±52.55	0.73±0.30	1.38±0.16
$2r_H$ (a hydrodynamic diameter of FD ($M_w = 150k$))	13.09-21.08	13.09-21.08			
$2r_H$ (a hydrodynamic diameter of collagenase ($M_r = 70-120 k$))	6.0	6.0			

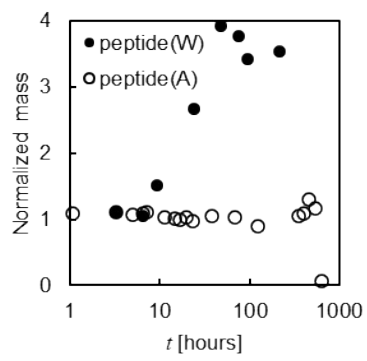
(a) 8-arm(5), 0.05 $\mu\text{g/mL}$ collagenase



(b) 8-arm(5), 0.5 $\mu\text{g/mL}$ collagenase



(c) 8-arm(5), 0.05 $\mu\text{g/mL}$ collagenase



(d) 8-arm(5), 0.5 $\mu\text{g/mL}$ collagenase

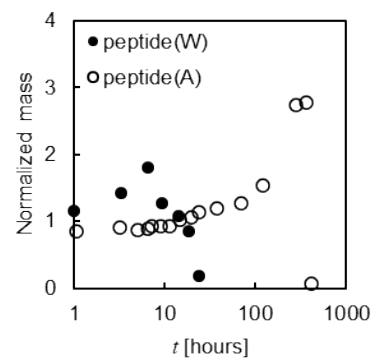


Figure S3. (a), (b) Mass change and (c), (d) diameter change of hydrogels in degradation process ($n = 1$).