Measurement of Interfacial Strength of Ultra-soft Materials Using Needle-Induced Cavitation

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S1. Micro-indentation to measure the ultra-soft hydrogel modulus

Young's moduli of hydrogels in vials are determined with the flat probe indentation test. The resultant force (F) and displacement (δ) are collected with the Texture Analyzer (model name: TA-XT plus) manufactured by Stable Micro Systems. The utilized flat steel probe has a diameter of 2 mm. The Young's modulus is calculated with the following algorithm[1–3]:

$$E = \frac{3}{8\left(\frac{d\delta}{dF}\right)a}\left(1 + 1.33\left(\frac{a}{h}\right) + 1.33\left(\frac{a}{h}\right)^3\right)^{-1}. \tag{S1.1}$$

Here, we adopt the Poisson's ratio ν to be 0.5 by assuming PAM hydrogels to be incompressible. The shear modulus can be determined by $\mu = \frac{E}{2(1+\upsilon)} = \frac{E}{3}$ accordingly. The parameter $\frac{d\delta}{dF}$ is the compliance at the small strain regime, specifically around 0.1 mm after contact. The contact radius (a) is 1 mm; and the hydrogel thickness in vials (h) is approximately 35 mm. The average Young's modulus of hydrogels is determined with the moduli of five hydrogel samples with different separator thicknesses, and the averaged Young's modulus is subsequently utilized to compute G_c in Equation 10. The results of hydrogel shear moduli are listed in Table S1.

Table S 1 Shear moduli of polyacrylamide hydrogels

Hydrogel formulation	Separator thickness (mm)	Shear modulus (kPa)	Average ± Standard deviation (kPa)
5 wt.% AM aqueous solution, 1:100 (w:w) bis-AM:AM	0.13	0.680	0.783±0.077
	0.26	0.723	
	0.39	0.840	
	0.78	0.817	
	1.08	0.853	

S2. Finite Element Modeling (FEM) for normal stress at the interface

We use FEM to calculate the normal stress at the interface with respect to the separator thickness (t). A two-dimensional model is utilized, depicting a solid circle containing a rectangular void at its center. The diameter of the circle ($D = 30$) and the width of the void ($w = 15$) are predetermined based on the experimental geometries. The neo-Hookean model was applied, and we set Young's modulus to be 1 kPa and Poisson's ratio to be 0.495 by setting parameters C10 to be 0.166 and D1 to be 0.066. To simulate the normal stress in the deformed state, displacement boundary conditions are applied at the entire width edges of the rectangle. The displacement of each side equals half of the void thickness, resulting in a zero-gap distance between the two widths in the deformed state, resembling the closed state. The outer surface of the model is pinned. The thickness of the rectangular void ranges from 0.01 to 3, enabling the investigation of the normal stress relationship related to the thickness. Normal stress is extracted from the field output function S_{22}. The amplitude of normal stress at the rectangular void center is the representative σ_{re} to be compared with the critical pressure decrease in Figure 4(c).

S3 Strain energy density of internally inflated cylinder, u_2

We utilize a two-dimensional cylinder tube model to elucidate the deformation occurring at the needle nozzle end. The pressure is applied at the inner wall to inflate the void and the stretch ratio in the azimuth direction is denoted as $\lambda_\theta = \lambda = a/A$, where a and A are the deformed and undeformed inner radii, respectively. Assuming the solid to be incompressible, the stretch ratio in the radial direction and axial direction is denoted as $\lambda_r = 1/\lambda$ and $\lambda_z = 1$, respectively. In accordance with the neo-Hookean constitutive model, the strain energy density (u_2) is expressed as:

$$u_2 = \frac{\mu}{2}(I_1 - 3) = \frac{\mu}{2}\left(\frac{1}{\lambda^2} + \lambda^2 - 2\right). \tag{S3.1}$$

Here, $\mu = E/3$ is the shear modulus, and I_1 is first invariant of the right Cauchy-Green deformation tensor. By integrating the energy density in the whole body, the total strain energy per unit length (L_0) in the axial direction is:

$$U_2 = \int_A^{R_o \to \infty} u_2 * d(\pi R^2) = \frac{\pi}{2}\mu A^2(\lambda^2 - 1)\ln(\lambda^2). \tag{S3.2}$$

As the radial expansion is balanced by hydrostatic pressure, non-negligible surface tension, the total energy (U_{cyt}) of an inflated cylinder is the sum of strain energy (U_2), surface energy (U_{2s}), and potential energy ($-PV_{cy}$), expressed as:

$$U_{cyt} = U_2 + U_{2s} - PV_{cy} = \frac{\pi}{2}\mu A^2(\lambda^2 - 1)\ln(\lambda^2) + 2\pi A\lambda\gamma - \pi A^2 P\lambda^2. \tag{S3.3}$$

$U_{s,cy}$is the surface energy; $\gamma = 0.07 J/m^2$ is the surface energy of water; $V_{cy} = \pi A^2 P\lambda^2$ is the volume of cylindrical air per unit length at the deformed state.

Solving the equation of $\frac{dU_{cyt}}{d\lambda} = 0$ leads to the expression of pressure balance with respect to the deformation:

$$P = \frac{\mu}{2}\left(1 + \ln(\lambda^2) - \lambda^{-2}\right) + \frac{\gamma}{A}\lambda^{-1}. \tag{S3.4}$$

This is Equation 6 in the manuscript.

S4. Void inspection method

The void inspection method is developed to determine G_c of ultra-soft solids by calculating the elasto-adhesion length scale $l_{EA}=\frac{G_c}{E}$ [4]. The ultra-soft hydrogels have the same formulation as presented in this work. Rectangular prism voids are introduced by placing PTFE films (separators) with different thicknesses into the precursor solutions prior to fully crosslinking. Upon removal of the separator, the void occurs, and the surrounding solid undergoes deformation due to the interplay between surface and elastic energy. As the separator thickness increases, the voids in the hydrogels exhibit a configuration transition from a fully closed interface, a partly closed interface with opened edges, to a fully opened void. We measured the ratio of the closed width to the total width, w_c/w, and the separator thickness (t). The governing equation $l_{EA}=\frac{Kt^2f'\left(\frac{w_c}{w}\right)}{w}$ is utilized to determine l_{EA}. In the above equation, K is a geometric parameter and $f'\left(\frac{w_c}{w}\right)$ is an empirical polynomial function. The original result of void inspection method can be found in Table S2.

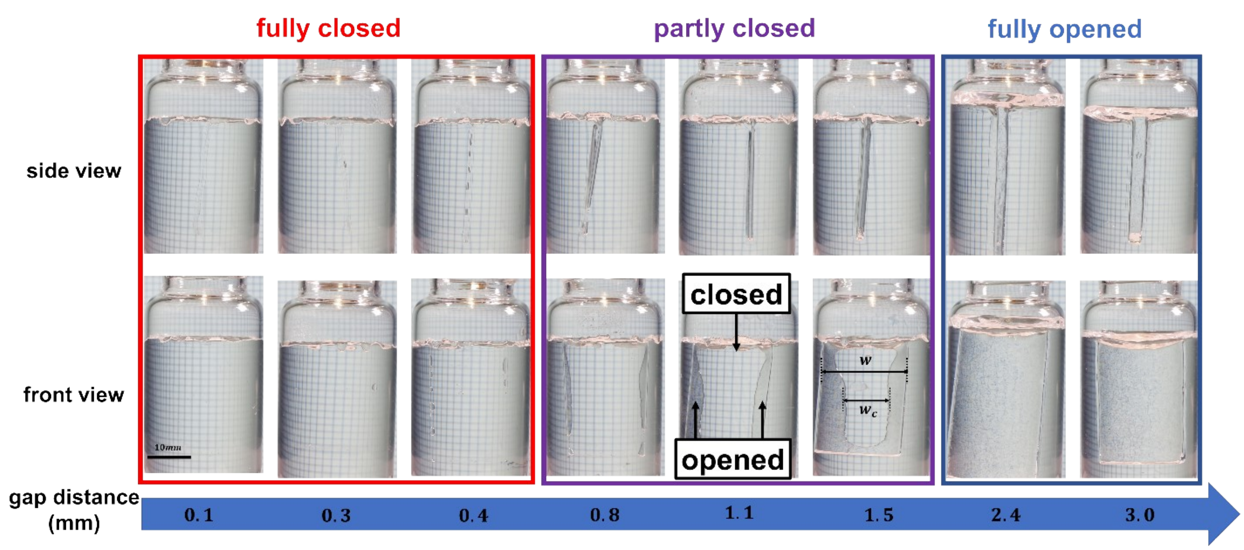

Figure S 1 The hydrogel interface configurations with varied separator thicknesses. For the partly closed interfaces, the ratio of closed width to the total width is of interest.

Table S 2 Determining G_c by void inspection method

$\frac{w_c}{w}$	$t\ (mm)$	$E\ (kPa)$	$l_{EA}\ (mm)$	$G_c = l_{EA} * E$ $(J/m2)$
8.20E-01	7.80E-01	2.45E+00	3.87E-01	9.48E-01
6.40E-01	1.08E+00	2.56E+00	4.31E-01	1.10E+00
3.70E-01	1.52E+00	2.14E+00	3.38E-01	7.23E-01
			Average	9.25E-01
			Standard deviation	1.91E-01

Table S 3 Test results and G_c fitting of Equation 10

$A\ (mm)$	$\mu\ (kPa)$	P_{c0}/μ	λ_c	$C = \frac{d\lambda}{dA}\ (mm^{-1})$	Y	$\frac{1}{\mu A}\ \frac{m}{(N)}$	P_{cNIC}/μ
0.865	0.783	0.663	1.408	0.0831	0.396	1.476	2.707
0.685		0.722	1.448	0.1367	0.492	1.864	2.761
0.570		0.928	1.671	0.2166	1.092	2.241	2.814
0.440		1.190	2.036	0.4026	2.630	2.903	2.906
0.290		1.262	2.074	0.9758	3.062	4.404	3.117

S5. A video of the interface NIC with a synchronized pressure profile.

The needle outer diameter is 0.58 mm. The thickness of the preset PTFE separator is 1.08 mm. The video is displayed at eight-time playback speed.

Bibliography

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