

Supporting Information

**A tough, injectable, naturally-derived, and cost-effective composite hydrogel for tissue sealing**

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**Table S1.** Formulation of hybrid hydrogels.

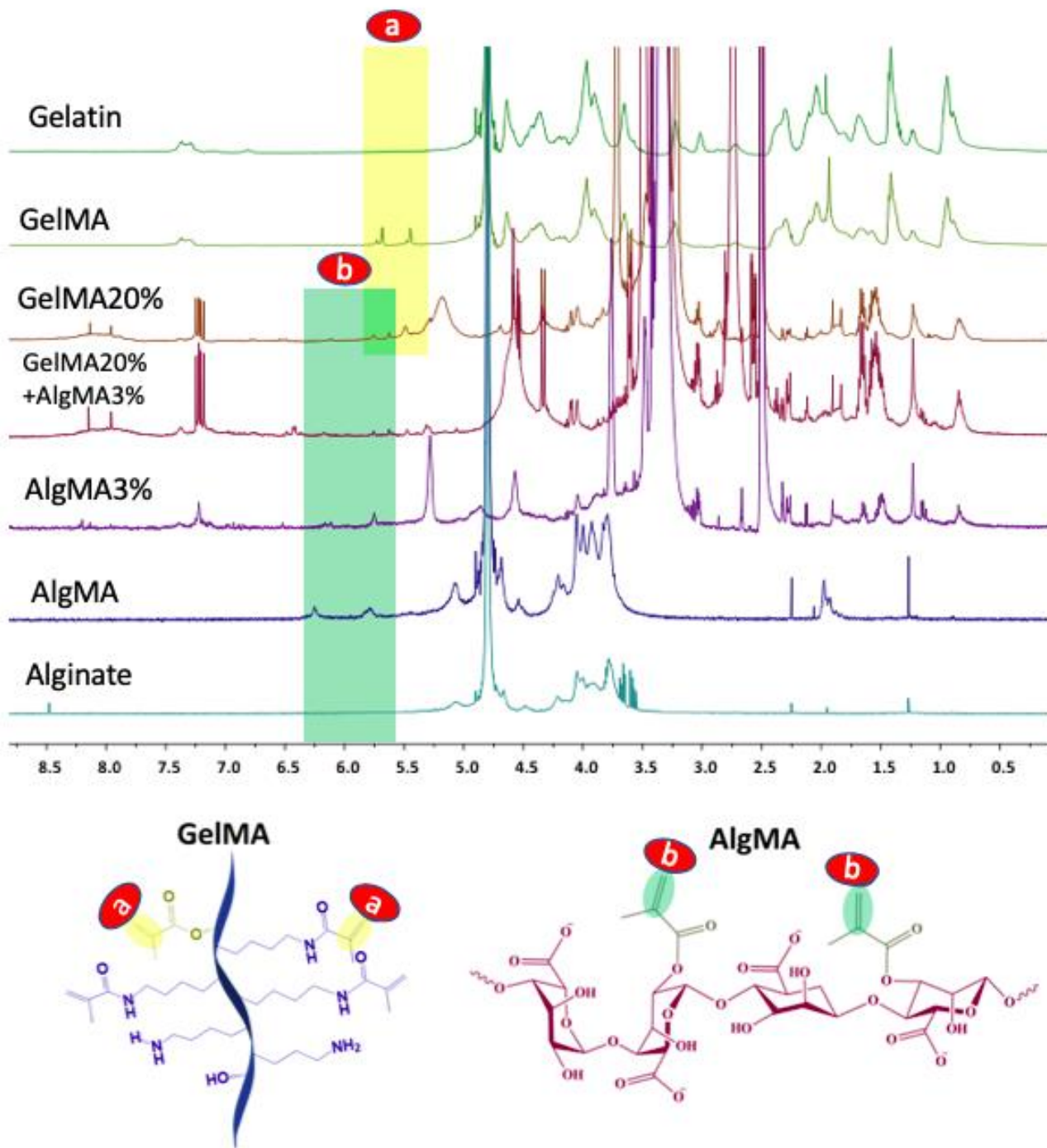
$$\text{VC\% (w/v)} = 0.05 \times [\text{GelMA+AlgMA}]\% \text{ (w/v)}$$

$$\text{TEA\% (w/v)} = 0.075 \times [\text{GelMA+AlgMA}]\% \text{ (w/v)}$$

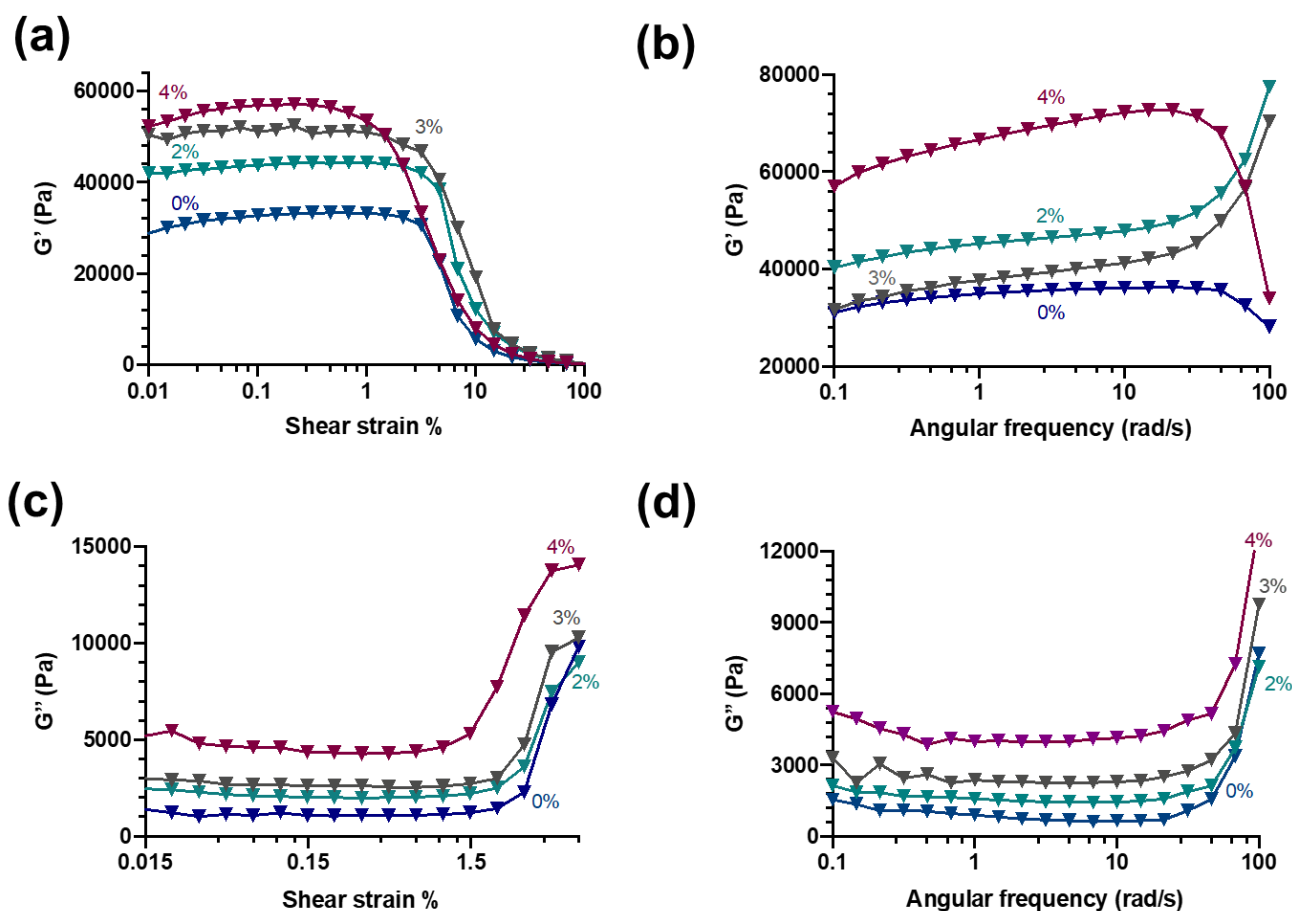
$$\text{Eosin Y\% (w/v)} = 0.0003 \times [\text{GelMA+AlgMA}]\% \text{ (w/v)}$$

$$\text{CaCl}_2\% \text{ (w/v)} = 0.13 \times \text{AlgMA}\% \text{ (w/v)}$$

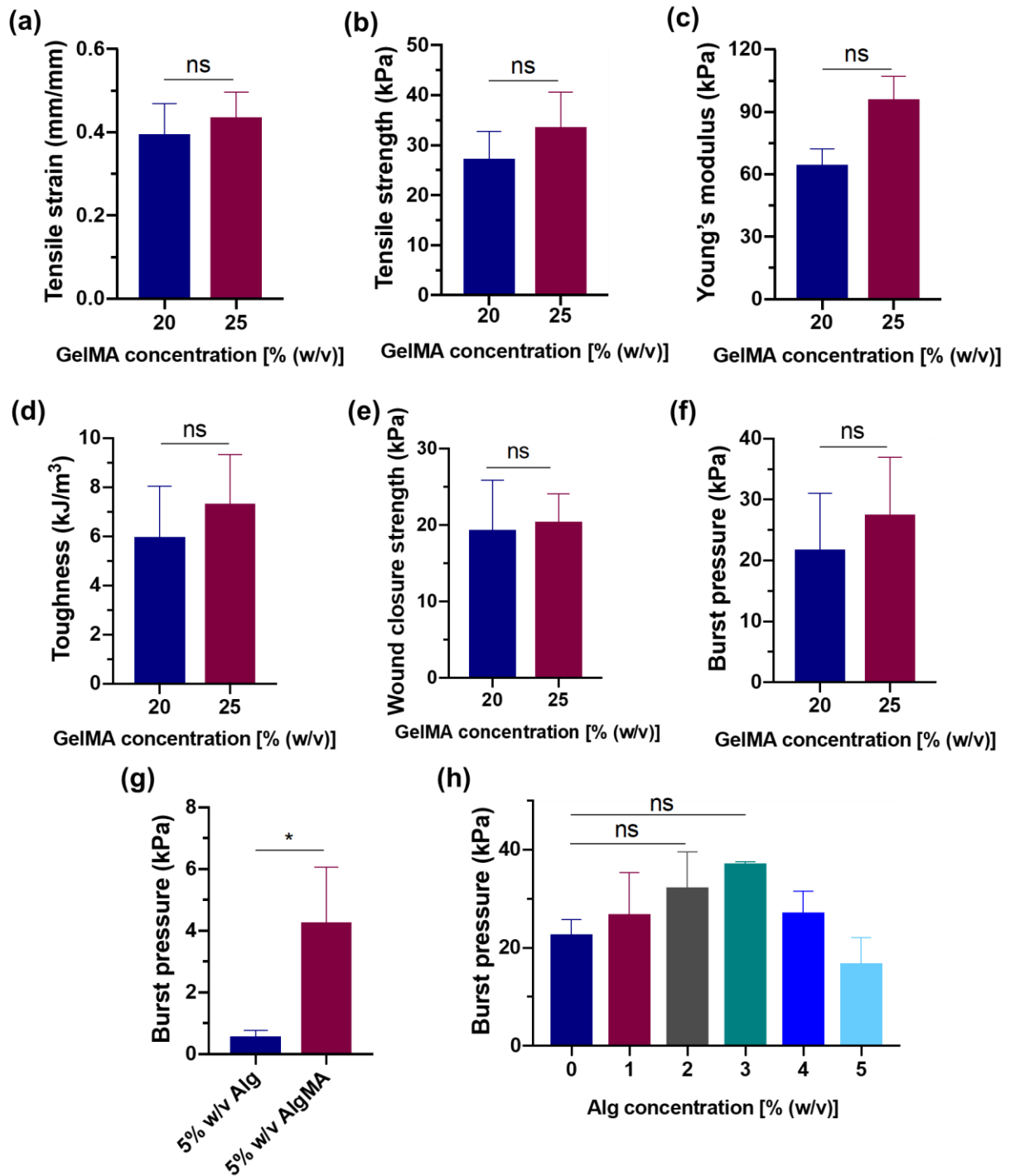
<b>Sample</b>	<b>AlgMA</b>	<b>GelMA</b>	<b>VC</b>	<b>TEA</b>	<b>Eosin Y</b>	<b>DPBS</b>	<b>CaCl<sub>2</sub></b>
	<b>% (w/v)</b>	<b>% (w/v)</b>	<b>% (w/v)</b>	<b>% (w/v)</b>	<b>% (w/v)</b>	<b>(<math>\mu</math>l)</b>	<b>% (w/v)</b>
<b>1</b>	0%	20%	1	1.5	0.006	600	0
<b>2</b>	1%	20%	1.05	1.575	0.0063	580	0.13
<b>3</b>	2%	20%	1.1	1.65	0.0066	560	0.27
<b>4</b>	3%	20%	1.15	1.725	0.0069	540	0.4
<b>5</b>	4%	20%	1.2	1.8	0.0072	520	0.53
<b>6</b>	5%	20%	1.25	1.875	0.0075	500	0.67
<b>7</b>	20%	0%	1	1.5	0.006	600	2.667
<b>8</b>	0%	25%	1.25	1.25	0.0075	500	0



**Figure S1.** <sup>1</sup>H NMR spectra of pre-gel solutions and crosslinked hydrogels. Biopolymers include gelatin, GelMA, crosslinked GelMA (20%), alginate, AlgMA, crosslinked AlgMA (3%), and crosslinked GelMA (20%)-AlgMA (3%) hybrid hydrogels.



**Figure S2. Viscoelastic properties of GelMA-AlgMA hybrid hydrogels.** (a) Storage modulus and (b) loss modulus versus oscillatory shear strain (at angular frequency  $\sim 1 \text{ rad s}^{-1}$ ) and angular frequency (at oscillatory shear strain  $\sim 0.1\%$ ) for the hybrid hydrogels composed of GelMA (20% w/v) and varying concentrations of AlgMA.



**Figure S3. Mechanical and adhesive properties of various biopolymeric systems.** (a) Tensile strain at break (b) tensile strength at break, (c) Young's modulus, (d) toughness, (e) wound closure strength, and (f) burst pressure of GelMA 20% compared to GelMA 25% showing no significant difference. (g)

Burst pressure of alginate (5%) compared to crosslinked AlgMA (5%). **(h)** Burst pressure of hybrid hydrogels made up of GelMA (25%) and varying concentrations of alginate. Data are reported as the mean values of at least 5 experiments  $\pm$  their standard deviation. The statistical analysis was done according to the methods explained in “Statistical analysis” section. Asterisks show the results that are statically significant with p-values less than 0.05 (\*).