

Development of aqueous protein/polysaccharide mixture-based inks for 3D printing towards food applications

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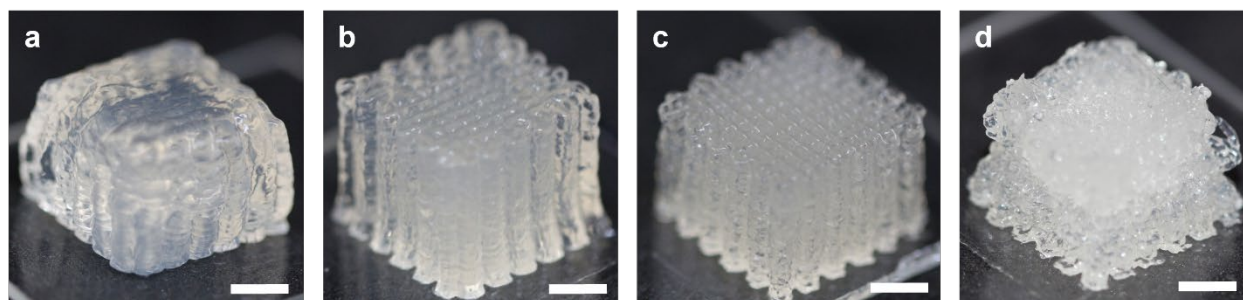


Figure S1. Comparison of the shape retention of as-printed 40-layers scaffolds using a 200- μm nozzle using various gel formulations right after printing at room temperature: with (a) 10 wt% pure XG ink under 224 kPa, (b) GB/XG ink with 12.5mM Ca^{2+} under 504 kPa, (c) GB/XG ink with 37.5mM Ca^{2+} under 588 kPa and (d) GB/XG ink with 62.5mM Ca^{2+} under 784 kPa. Scale bars are 5 mm.

Table S1 Viscosity at $\dot{\gamma} = 1 \text{ s}^{-1}$ for different food inks at room temperature (22 °C)

Food ink type	Viscosity (Pa.s)
Pure 10 wt% XG gel	158 ± 30
GB/XG gel with no salt	231 ± 27
GB/XG gel with 12.5 mM Ca^{2+}	313 ± 24
GB/XG gel with 37.5 mM Ca^{2+}	460 ± 67
GB/XG gel with 62.5 mM Ca^{2+}	720 ± 91
GB/XG gel with 75 mM Ca^{2+}	700 ± 94

Table S2. G' and $\tan \delta$ at $\omega = 1 \text{ rad/s}$ for different food inks

Food ink	G' (Pa)	$\tan \delta$
Pure 10 wt% XG gel, RT	452 ± 26	0.15
GB/XG gel with no salt, RT	692 ± 32	0.14
GB/XG gel with 12.5 mM Ca^{2+} , RT	980 ± 51	0.13
GB/XG gel with 37.5 mM Ca^{2+} , RT	1720 ± 68	0.11
GB/XG gel with no salt, 4 °C	1390 ± 46	0.12
GB/XG gel with 12.5 mM Ca^{2+} , 4 °C	2040 ± 97	0.12
GB/XG gel with 37.5 mM Ca^{2+} , 4 °C	2839 ± 73	0.08

Another option is:

Food ink	G' (Pa)		$\tan \delta$	
	RT	4 °C	RT	4 °C
Pure 10 wt% XG gel	452 ± 26	-	0.15	-
GB/XG gel with no salt	692 ± 32	1390 ± 46	0.14	0.12
GB/XG gel with 12.5 mM Ca^{2+}	980 ± 51	2040 ± 97	0.13	0.12
GB/XG gel with 37.5 mM Ca^{2+}	1720 ± 68	2839 ± 73	0.11	0.08

Frequency dependence of G' . The angular frequency dependence of G' was presented by the power-law equation:

$$G' = K\omega^a$$

with the linear trend of G' on log-log scale as the function of angular frequency, where a is the slope and K is the y -intercept of the trendline.

Table S3 Power-law parameter a comparing food inks at different temperature (RT and 4 °C) within angular frequency dependence

Food ink type	Power-law parameter (a)	
	RT	4 °C
GB/XG gel with no salt	0.095±0.006	0.084±0.003
GB/XG gel with 12.5 mM Ca^{2+}	0.084±0.005	0.081±0.006
GB/XG gel with 37.5 mM Ca^{2+}	0.083±0.003	0.050±0.002

Apparent yield stress determination. The process apparent yield stress was determined while extruding the ink by the 3D printer. The results were calculated based on the equation (Bruneaux, Therriault, & Heuzey, 2008):

$$\tau_{app} = \frac{P_{flow} - P_a}{2(\frac{L}{R})}$$

where τ_{app} is the apparent yield stress, P_{flow} is the minimum pressure required for flow, P_a is the atmospheric pressure, L and R are, respectively, the length inner radius of the nozzle.

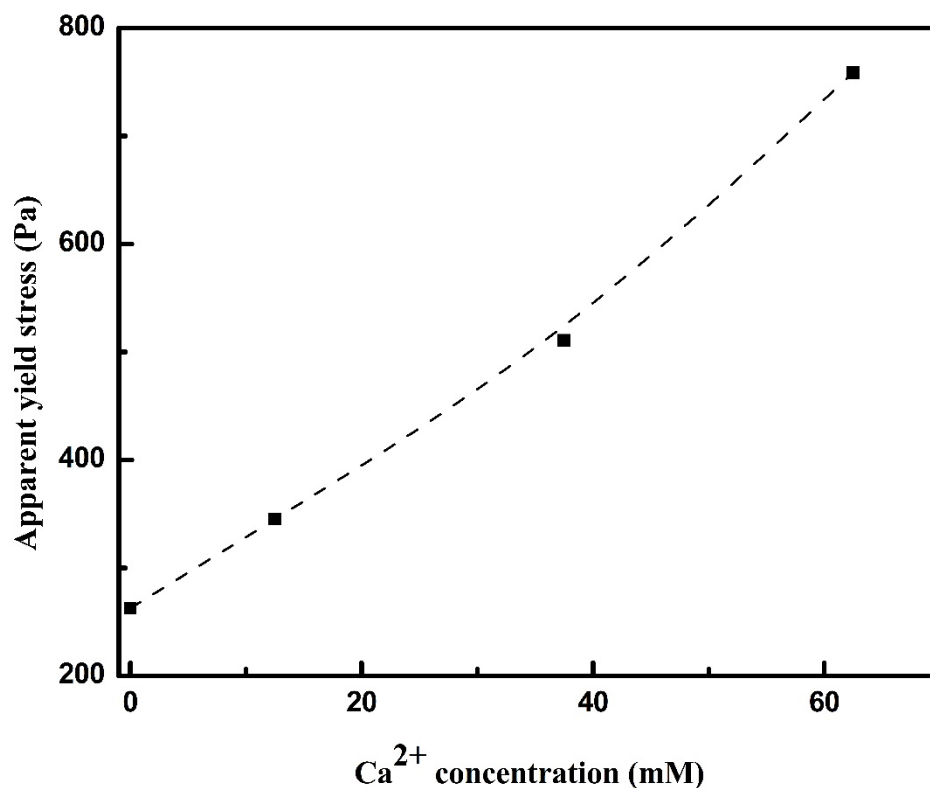


Figure S2. Apparent yield stress as a function of Ca²⁺ concentration.

Table S4. Textural properties of the 3D printed scaffolds made of different gel formulations *.

Effect	Food ink	Hardness (g)	Adhesiveness (g.s)	Cohesiveness	Gumminess (g)
Shape effect	25% IP GB/XG	8.3 (0.5) ^d	43.9 (2.2) ^d	0.64 (0.01) ^d	5.4 (0.3) ^d
	40% IP GB/XG	25.6 (1.1) ^c	84.0 (1.5) ^c	0.71 (0.01) ^c	18.2 (0.9) ^c
	64% IP GB/XG	36.2 (1.2) ^b	158.6 (4.0) ^b	0.75 (0.01) ^b	27.2 (0.8) ^b
	96% IP GB/XG	74.0 (2.1) ^a	183.2 (3.4) ^a	0.82 (0.02) ^a	60.7 (2.8) ^a
Ca ²⁺ addition effect	64% IP no salt	36.2 (1.2) ^c	158.6 (4.0) ^d	0.75 (0.01) ^c	27.2 (0.8) ^d
	64% IP 12.5 mM Ca ²⁺	39.9 (1.6) ^c	172.8 (4.0) ^c	0.78 (0.02) ^b	31.1 (1.6) ^c
	64% IP 37.5 mM Ca ²⁺	45.3 (2.4) ^b	198.3 (5.7) ^b	0.81 (0.01) ^a	36.7 (1.4) ^b
	64% IP 62.5 mM Ca ²⁺	57.3 (3.5) ^a	221.3 (8.3) ^a	0.82 (0.01) ^a	47.0 (3.8) ^a
Temperature effect	RT 64% IP GB/XG	36.2 (1.2) ^b	158.6 (4.0) ^a	0.75 (0.01) ^b	27.2 (0.8) ^b
	4 °C 64% IP GB/XG	95.1 (6.5) ^a	64.0 (2.7) ^b	0.84 (0.02) ^a	79.9 (7.3) ^a

*: Values in parenthesis are standard deviation $n = 3$. Mean values with the same lowercase superscript letter are not significantly different ($p > 0.05$) between the row.

IP = infill percentage

References

Bruneaux, J., Therriault, D., & Heuzey, M.-C. (2008). Micro-extrusion of organic inks for direct-write assembly. *Journal of Micromechanics and Microengineering*, 18(11).