

## RESEARCH ARTICLE

Development of a 3D-printable matrix using cellulose microfibrils/ guar gum-based hydrogels and its post-printing antioxidant activity

# Supplementary File



Figure S1. Morphology of cellulose microfibrils produced at different blending times. (A) 0 min, (B) 80 min, and (C) 160 min.



**Figure S2**. Effect of different blending times on (A) sedimentation, (B) reduced viscosity, (C) water retention value and *Ct*CBD3 binding ability, and (D) FTIR spectra of cellulose microfibrils.



Figure S3. 3D-printed CMFs with different types of hydrocolloid such as (A) alginate, (B) starch, and (C-F) guar gum at different concentrations from 1% to 7% using (G) 2D design.

Designed shape			w/1% gg	w/3% gg	w/5% gg	w/7% gg
2D Square	Overall width (cm)	3.2	$3.19\pm0.00^{\rm a}$	$3.21\pm0.01^{\rm b}$	$3.21\pm0.01^{\rm b}$	N.E.
	Line width (cm)	0.6	$0.57\pm0.06^{a}$	$0.61\pm0.03^{\rm a}$	$0.58\pm0.01^{\rm a}$	N.E.
Designed shape			w/ 3% gg	w/ 4% gg	w/ 5% gg	
3D Cube	Width (cm)	1.6	$1.72\pm0.02^{\rm b}$	$1.48\pm0.01^{\rm a}$	$1.48\pm0.02^{\text{a}}$	
	Length (cm)	1.6	$1.65 \pm 0.01^{\mathrm{b}}$	$1.54\pm0.02^{a}$	$1.56 \pm 0.01^{a}$	
	Height (cm)	1.0	$0.42\pm0.02^{a}$	$0.96\pm0.01^{\rm b}$	$0.99\pm0.01^{\circ}$	
	Volume (cm <sup>3</sup> )	2.56	$1.17\pm0.00^{a}$	$2.16\pm0.03^{\rm b}$	$2.27 \pm 0.01^{\circ}$	
	Dimensional error (%)		$54.30\pm0.01^{\rm b}$	$15.63 \pm 0.01^{b}$	$11.33\pm0.01^{\text{a}}$	

### Table S1. Dimensions of a 2D square and 3D cube printed using 1% CMF with the addition of different concentrations of guar gum

Data are expressed as mean  $\pm$  standard deviation (n = 3).

<sup>a-c</sup> In the same row with different letters are significantly different at p < 0.05.

Abbreviations: gg, guar gum; N.E., not extruded; w/, 100 mL of 1% (w/v) CMFs.

	Side view		Top view	
Designed shape				
Infill density (%)	30	40	50	60
Printing speed (mm/s)	1	5	10	-
Nozzle diameter (mm)	0.4	0.8	1.0	1.5
Layer height (mm)	0.3	0.4	0.5	0.6

## Table S2. The appearance of the printed constructs based on printing parameters

	Storage time (h)			
Treatment time (min)	0	3	6	12
0				
Dimensional error (%)	$12.76\pm0.81^{a,A}$	$24.69 \pm 1.70^{\text{b},\text{B}}$	$30.58 \pm 4.10^{\rm b,C}$	$40.00 \pm 3.09^{a,D}$
30				
Dimensional error (%)	$11.88 \pm 1.53^{a,A}$	21.11 ± 1.63 <sup>a,B</sup>	$26.06 \pm 1.75^{a,C}$	30.66 ± 2.33 <sup>a,D</sup>

#### Table S3. Stability of the ink printed during storage

Data are expressed as mean  $\pm$  standard deviation (n = 3).

<sup>a-b</sup> In the same column with different letters are significantly different at p < 0.05.

<sup>A-D</sup> In the same row with different letters are significantly different at p < 0.05.

## Table S4. pH value of the respective substances and composite hydrogel or borax-treated hydrogel for CMFs, guar gum, and borax

Sample			pH value		
1% CMFs		$4.96 \pm 0.05$			
GG		$6.33 \pm 0.02$			
1 M Borax		9.46 ± 0.02			
Various concentration of GG	1%	3%	5%	7%	
CMFs/GG	$6.04\pm0.05^{\rm a}$	$6.31\pm0.02^{\rm b}$	$6.33\pm0.01^{\rm b}$	$6.37\pm0.02^{\rm b}$	
CMFs/GG/1 M Borax	$7.16 \pm 0.01^{a}$	$7.13 \pm 0.02^{a}$	$7.17 \pm 0.02^{a}$	$7.19\pm0.05^{a}$	

Data are expressed as the mean  $\pm$  standard deviation (n = 3).

 $^{\rm a-b}$  In the same row with different letters are significantly different at p < 0.05.

Abbreviations: CMFs, cellulose microfibrils; GG, guar gum.