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## Data Article

# Dataset on the small- and large deformation mechanical properties of emulsion-filled gelatin hydrogels as a model particle-filled composite food gel



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## ABSTRACT

In this article we present data related to the original research articles 'Effect of matrix architecture on the elastic behavior of an emulsion-filled polymer gel' (Gravelle et al., 2021) and 'The influence of network architecture on the large deformation and fracture behavior of emulsion-filled gelatin gels' (Gravelle and Marangoni, 2021). The small deformation elastic (Young's) modulus and large deformation fracture behavior of emulsion-filled composite gelatin gels are reported as a function of filler volume fraction ( $\phi_f = 0 - 0.32$ ). Homogeneous and heterogeneous network architectures were achieved by varying electrostatic interactions between matrix and filler. The effect of emulsion droplet physical state (solid fat or liquid oil) and gelator concentration (2, 4, 6, or 8% gelatin) were also evaluated. The reported elastic modulus, and fracture properties were obtained from large deformation uniaxial compression tests. Power law scaling behavior was identified for the elastic modulus as a function of both  $\phi_{\rm f}$  and gelator concentration, which are also reported. This data is relevant to the evaluation of network properties on the applicability of small deformation particle reinforcement theories and models describing the fracture mechanics of filled composites such as fat-filled food systems.

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## **Specifications Table**

Subject	Agricultural and Biological Sciences
Specific subject area	Food Science, Materials Science
Type of data	Tables
How data was acquired	Large deformation analysis acquired via uniaxial compression (TA.XT2, Stable Micro Systems, Texture Technologies Corp., Scarsdale, NY, USA)
Data format	Analyzed
Parameters for data collection	Gelatin gels: protein content $c_{gel}$ of 2, 4, 6, or 8 wt%; pH 4.0 or pH 6.0; emulsion volume fraction $\phi_f$ of 0, 0.054, 0.108, 0.161, 0.214, 0.266, and 0.318 Compression rate: 1.0 mm/s: final compression strain of 85%
	Data analyzed using the Exponent software (Stable Micro Systems, Texture Technologies Corp., Scarsdale, NY, USA)
Description of data collection	Cylindrical pucks (height: 10 mm; diameter: 22 mm) compressed via uniaxial compression to 85% strain to generate force/deformation profiles.
	Small deformation: Determination of elastic modulus of fat-filled composite gels $(E_c)$ from initial linear region ( $\leq$ 10% strain).
	Large deformation: Determination of true fracture strain ( $\varepsilon^*$ ) and true fracture stress ( $\sigma^*$ ) at fracture point.
Data source location	University of Guelph, Guelph, ON, Canada
Data accessibility	Open access via Mendeley Data: Gravelle, Andrew; Marangoni, Alejandro (2021), "Emulsion-filled gelatin hydrogels: Mechanical behavior under small and large
	deformations", Mendeley Data, V1, doi:10.17632/mzky273yy9.1
Related research article	A.J. Gravelle and A.G. Marangoni, Effect of matrix architecture on the elastic
	behavior of an emulsion-filled polymer gel, Food Hydrocolloids, 119 (2021)
	106,875. https://doi.org/10.1016/j.foodhyd.2021.106875
	A.J. Gravelle and A.G. Marangoni, The influence of network architecture on the
	large deformation and fracture behavior of emulsion-filled gelatin gels, Food
	Structure, 29 (2021) 100,193. https://doi.org/10.1016/j.foostr.2021.100193

## Value of the Data

- Emulsion-filled gelatin gels serve as a model composite food system for a variety of fat-filled food systems.
- This dataset will benefit researchers developing or validating new particle reinforcement models, or proposed modifications to existing theories.
- Adjusting parameters such as network structure, filler physical state, and gelator concentration (i.e., relative modulus of filler and matrix) provides insight into the contribution of fillers on the small deformation mechanical/rheological properties of composite food gels.
- Systematic variation in several parameters allows exploration of the properties which contribute to the observed mechanical behavior of particle-filled soft materials, which will be relevant to modifying or developing new particle reinforcement models.
- Evaluation of fracture behavior also provides insight into the applicability of established models describing fracture stress and fracture strain.
- Both small- and large deformation behavior of emulsion-filled hydrogels could provide insight into the initial sensory properties of fat-filled foods.

## 1. Data Description

This dataset corresponds to the small- and large deformation mechanical properties of thermally-induced gelatin containing whey protein isolate-stabilized emulsion droplets as filler particles. The tabulated data is distinguished by gelation conditions and filler physical state. Emulsion-filled gels were prepared using either B-Type gelatin at pH 6 (Gel- $6_B$ ), A-Type gelatin at pH 6 (Gel- $6_A$ ; elastic modulus only), or A-Type gelatin at pH 4 (Gel- $4_A$ ). Gels prepared at pH 6 produced a homogeneous filler distribution, whereas those prepared at pH 4 had a highly heterogeneous network architecture [1,2]. All composite gels were prepared using 2, 4, 6, and 8 wt% gelatin. For each gelatin type, emulsion droplets were prepared with either solid fat or liquid oil as the lipid phase (denoted as fat and oil, respectively).

*Note:* All data tables are also provided in an Excel worksheet as supporting information (Supplementary Data).

## 1.1. Small deformation elastic behavior

Data presented in this section corresponds to the elastic modulus of composite gels ( $E_c$ ) consisting of thermally-induced gelatin containing whey protein isolate-stabilized emulsion droplets as filler particles. The elastic constant was determined from the initial linear region of the force/deformation curves (strain  $\leq$ 10%) and  $E_c$  was determined using the sample geometry. Values presented in Tables 1–6 contain tabulated values of  $E_c$  as a function of filler volume fraction ( $\phi_f$ ) for composites prepared with Gel-6<sub>B</sub>, Gel-6<sub>A</sub>, and Gel-4<sub>A</sub>, each containing either solid fat or liquid oil emulsion droplets, as indicated in the Table captions.

Table 7 presents the scaling factor  $\gamma$  **obtained by fitting the data from each column** of Tables 1 and 2 (Gel-6<sub>B</sub>) and Tables 5 and 6 (Gel-4<sub>A</sub>) to the power law scaling relation  $E_c \sim (1 - \phi_f)^{-\gamma}$ .

Table 8 presents the scaling factor  $\delta$  **obtained by fitting the data from each row** of Tables 1 and 2 (Gel-6<sub>B</sub>) and Tables 5 and 6 (Gel-4<sub>A</sub>) to the power law scaling relation  $E_c \sim c_{gel}^{-\gamma}$  (where  $c_{gel}$  denotes gelator concentration).

Tal	bl	e	1

Elastic modulus of gelatin gels prepared with B-type gelatin at pH 6 (Gel-6<sub>B</sub>) and solid fat as the lipid phase with varying fraction filler ( $\phi_f$ ) and gelatin concentration. All values reported in kPa.

		Gelatin concentration			
$arphi_{ m f}$	2%	4%	6%	8%	
0	$3.20\pm0.24$	$10.57\pm0.64$	$24.16\pm0.37$	40.85 ± 1.81	
0.054	$3.77 \pm 0.19$	$11.47 \pm 0.04$	$27.41 \pm 1.33$	$48.44 \pm 2.82$	
0.108	$3.99\pm0.48$	$13.95 \pm 0.41$	$32.92 \pm 0.72$	$52.12 \pm 3.01$	
0.161	$4.42\pm0.08$	$15.89 \pm 0.62$	$36.68 \pm 1.20$	$59.84 \pm 4.82$	
0.214	$4.88\pm0.18$	$16.21 \pm 2.05$	$43.48 \pm 4.90$	$67.84~\pm~5.84$	
0.266	$5.45\pm1.07$	$22.44 \pm 0.75$	$50.12 \pm 1.52$	$78.48 \pm 5.05$	
0.318	$6.99\pm0.72$	$29.45\pm1.11$	$56.79\pm1.39$	$100.25\pm3.22$	

#### Table 2

Elastic modulus of gelatin gels prepared with B-type gelatin at pH 6 (Gel-6<sub>B</sub>) and liquid oil as the lipid phase with varying fraction filler ( $\phi_f$ ) and gelatin concentration. All values reported in kPa.

	Gelatin concentration			
$arphi_{ m f}$	2%	4%	6%	8%
0	$3.60\pm0.13$	$14.56 \pm 1.36$	$26.78\pm6.59$	47.29 ± 11.63
0.054	$4.32 \pm 0.21$	$16.70 \pm 1.13$	$29.95 \pm 6.10$	$48.58 \pm 6.62$
0.108	$4.49\pm0.45$	$17.17 \pm 0.26$	$32.41 \pm 6.45$	$53.57 \pm 7.77$
0.161	$4.88\pm0.52$	$19.01 \pm 1.06$	$34.35 \pm 6.20$	$59.05 \pm 9.62$
0.214	$4.96\pm0.82$	$21.18 \pm 1.30$	$36.95 \pm 6.06$	$62.21 \pm 6.76$
0.266	$5.56\pm0.79$	$23.35 \pm 2.02$	$41.99 \pm 8.42$	$68.04 \pm 11.12$
0.318	$5.47\pm0.35$	$24.14~\pm~5.71$	$43.93\pm9.99$	$71.94\pm10.37$

Elastic modulus of gelatin gels prepared with A-type gelatin at pH 6 (Gel- $6_A$ ) and solid fat as the lipid phase with varying fraction filler ( $\phi_f$ ) and gelatin concentration. All values reported in kPa.

	Gelatin concentration			
$arphi_{ m f}$	2%	4%	6%	8%
0	$3.60\pm0.20$	$10.78\pm0.28$	22.87 ± 1.12	$40.39 \pm 0.53$
0.054	$4.02\pm0.18$	$11.77 \pm 0.21$	$24.08\pm0.94$	$43.57\pm0.80$
0.108	$4.67 \pm 0.11$	$12.99 \pm 0.04$	$28.58 \pm 1.16$	$50.60\pm0.38$
0.161	$5.37 \pm 0.12$	$14.64 \pm 0.14$	$33.03 \pm 1.49$	$54.86 \pm 4.45$
0.214	$5.77 \pm 0.26$	$16.27 \pm 0.03$	37.25 ± 1.79	$64.71 \pm 5.02$
0.266	$6.69 \pm 0.14$	$18.89 \pm 0.31$	$43.29 \pm 1.35$	76.34 ± 1.71
0.318	$7.61\pm0.45$	$22.48\pm1.00$	$53.26\pm2.10$	$91.12~\pm~1.90$

## Table 4

Elastic modulus of gelatin gels prepared with A-type gelatin at pH 6 (Gel-6<sub>A</sub>) and liquid oil as the lipid phase with varying fraction filler ( $\phi_f$ ) and gelatin concentration. All values reported in kPa.

	Gelatin concentration				
$arphi_{ m f}$	2%	4%	6%	8%	
0	$4.02\pm0.08$	$10.80\pm0.05$	$22.90 \pm 0.39$	41.54 ± 1.10	
0.054	$4.40\pm0.27$	$11.89 \pm 0.48$	$25.13 \pm 0.80$	$42.35 \pm 0.20$	
0.108	$4.70 \pm 0.29$	$12.65 \pm 0.64$	$27.83 \pm 0.57$	$45.36 \pm 0.16$	
0.161	$5.13 \pm 0.11$	$14.38 \pm 0.82$	$29.59 \pm 0.62$	$53.53 \pm 0.28$	
0.214	$5.40\pm0.04$	$15.42 \pm 0.79$	$33.82 \pm 0.37$	$55.95 \pm 0.25$	
0.266	$5.77 \pm 0.17$	$17.46 \pm 0.27$	$38.52\pm0.50$	$62.18 \pm 0.42$	
0.318	$6.05\pm0.15$	$18.81\pm0.48$	$40.18~\pm~1.07$	$67.35\pm0.67$	

## Table 5

Elastic modulus of gelatin gels prepared with A-type gelatin at pH 4 (Gel-4<sub>A</sub>) and solid fat as the lipid phase with varying fraction filler ( $\phi_f$ ) and gelatin concentration. All values reported in kPa.

	Gelatin concentration				
$arphi_{ m f}$	2%	4%	6%	8%	
0	$3.19\pm0.14$	$11.02\pm0.78$	$23.71 \pm 2.75$	$41.72 \pm 1.48$	
0.054	$4.71 \pm 0.11$	$12.97 \pm 0.64$	$31.44 \pm 2.42$	$49.75 \pm 1.51$	
0.108	$6.29\pm0.87$	$16.96 \pm 0.68$	$38.87 \pm 4.71$	$64.20 \pm 4.68$	
0.161	$7.49 \pm 0.74$	$21.34 \pm 1.50$	$47.23 \pm 4.20$	$73.81 \pm 4.18$	
0.214	$9.43\pm0.08$	$29.35 \pm 1.34$	$58.26 \pm 6.93$	$96.34 \pm 6.19$	
0.266	$12.85 \pm 1.60$	$34.92 \pm 1.92$	$72.39 \pm 9.51$	$112.81 \pm 4.85$	
0.318	$18.45\pm3.63$	$48.88~\pm~5.13$	$98.34 \pm 9.48$	$144.06\pm12.42$	

## Table 6

Elastic modulus of gelatin gels prepared with A-type gelatin at pH 4 (Gel-4<sub>A</sub>) and liquid oil as the lipid phase with varying fraction filler ( $\phi_f$ ) and gelatin concentration. All values reported in kPa.

	Gelatin concentration				
$arphi_{ m f}$	2%	4%	6%	8%	
0	3.28 ± 0.31	$13.19\pm0.77$	$34.16 \pm 9.53$	$50.19\pm4.33$	
0.054	$4.49\pm0.46$	$14.35 \pm 0.04$	$37.65 \pm 9.01$	$56.59 \pm 3.26$	
0.108	$5.81 \pm 0.07$	$17.07 \pm 0.93$	$45.46 \pm 11.57$	$58.31 \pm 2.93$	
0.161	$6.63 \pm 0.56$	$21.74 \pm 3.21$	$51.06 \pm 14.69$	$69.17~\pm~9.04$	
0.214	$8.28 \pm 0.21$	$28.07\pm2.66$	$56.20 \pm 13.37$	$75.89 \pm 9.97$	
0.266	$10.89 \pm 0.13$	$33.32 \pm 3.41$	$64.70 \pm 14.53$	83.92 ± 13.10	
0.318	$14.03\pm0.37$	$36.80\pm5.06$	$72.27 \pm 12.22$	$86.64 \pm 8.82$	

Scaling factors ( $\gamma$ ) corresponding to power law scaling relation  $E_r \sim (1 - \phi_f)^{-\gamma}$  for varying gelator concentration ( $c_{gel}$ ), gelatin type (B-Type, pH 6; Gel-6<sub>B</sub> or A-Type, pH 4; Gel-4<sub>A</sub>) and lipid physical state (solid fat or liquid oil). Scaling factor values are unitless.

	Gel-6 <sub>B</sub>		Gel-4 <sub>A</sub>	
c <sub>gel</sub> (wt%)	Fat	Oil	Fat	Oil
2	$1.94\pm0.28$	$1.12\pm0.29$	$4.31\pm0.16$	$3.56\pm0.13$
4	$2.82\pm0.22$	$1.30 \pm 0.29$	$3.92\pm0.07$	$3.19\pm0.26$
6	$2.19\pm0.14$	$1.26 \pm 0.47$	$3.54\pm0.10$	$1.91\pm0.55$
8	$2.30\pm0.17$	$1.15\pm0.36$	$3.15\pm0.06$	$1.45\pm0.28$

### Table 8

Scaling factors ( $\delta$ ) corresponding to power law scaling relation  $E_c \sim c_{gel}^{\delta}$  for varying gelator filler volume fraction ( $\phi_f$ ), gelatin type (B-Type, pH 6; Gel-6<sub>B</sub> or A-Type, pH 4; Gel-4<sub>A</sub>) and lipid physical state (solid fat or liquid oil). Scaling factor values are unitless.

	Gel-6 <sub>B</sub>		Gel	-4 <sub>A</sub>
$\phi_{ m f}$	Fat	Oil	Fat	Oil
0	$1.89\pm0.09$	$1.80\pm0.47$	$1.92\pm0.14$	$1.93\pm0.38$
0.054	$2.01 \pm 0.12$	$1.62 \pm 0.28$	$1.79 \pm 0.12$	$1.78 \pm 0.29$
0.108	$1.80\pm0.12$	$1.70\pm0.29$	$1.82~\pm~0.18$	$1.65\pm0.37$
0.161	$1.84\pm0.15$	$1.73 \pm 0.31$	$1.70~\pm~0.14$	$1.54\pm0.36$
0.214	$1.87\pm0.22$	$1.67\pm0.23$	$1.72 \pm 0.15$	$1.42\pm0.29$
0.266	$1.76 \pm 0.12$	$1.63 \pm 0.31$	$1.63 \pm 0.15$	$1.32 \pm 0.29$
0.318	$1.85\pm0.08$	$1.67\pm0.33$	$1.49\pm0.16$	$1.13\pm0.24$

#### Table 9

True fracture strain ( $\varepsilon^*$ ; unitless) of gelatin gels prepared with B-type gelatin at pH 6 (Gel-6<sub>B</sub>) and solid fat as the lipid phase with varying fraction filler ( $\phi_f$ ) and gelatin concentration. Values are unitless.

	Gelatin concentration			
$arphi_{ m f}$	2%	4%	6%	8%
0	$1.155\pm0.025$	$1.503\pm0.063$	$1.386 \pm 0.044$	$1.609\pm0.042$
0.054	$1.263 \pm 0.009$	$1.542 \pm 0.013$	$1.534 \pm 0.008$	$1.502\pm0.085$
0.108	$1.230 \pm 0.002$	$1.587 \pm 0.082$	$1.439 \pm 0.060$	$1.459 \pm 0.008$
0.161	$1.355 \pm 0.063$	$1.598 \pm 0.152$	$1.464 \pm 0.036$	$1.470\pm0.076$
0.214	$1.540 \pm 0.032$	$1.724 \pm 0.119$	$1.444 \pm 0.070$	$1.575 \pm 0.062$
0.266	$1.849 \pm 0.004$	$1.747 \pm 0.094$	$1.579 \pm 0.003$	$1.607\pm0.002$
0.318	$1.847\pm0.050$	$1.753 \pm 0.115$	$1.617\pm0.081$	$1.803\pm0.075$

## 1.2. Large deformation (fracture mechanics)

Data presented in this section corresponds to the true fracture strain ( $\varepsilon^*$ ) and true fracture stress ( $\sigma^*$ ) for thermally-induced composite gelatin containing whey protein isolate-stabilized emulsion droplets as filler particles. The fracture properties were determined from the fracture point of the force/deformation curves, and converted to true strain and true stress values using the sample geometry. Values presented in Tables 9–12 and 13–16, respectively, present tabulated values of  $\varepsilon^*$  and  $\sigma^*$  as a function of filler volume fraction ( $\phi_f$ ) for composites prepared with Gel-6<sub>B</sub> and Gel-4<sub>A</sub>, each containing either solid fat or liquid oil emulsion droplets, as indicated in the Table captions.

True fracture strain ( $\varepsilon^*$ ) of gelatin gels prepared with B-type gelatin at pH 6 (Gel-6<sub>B</sub>) and liquid oil as the lipid phase with varying fraction filler ( $\phi_f$ ) and gelatin concentration. Values are unitless.

	Gelatin concentration			
$arphi_{ m f}$	2%	4%	6%	8%
0	$1.118\pm0.003$	$1.433 \pm 0.125$	$1.633\pm0.132$	$1.676\pm0.067$
0.054	$1.151 \pm 0.031$	$1.333 \pm 0.082$	$1.400 \pm 0.025$	$1.456 \pm 0.030$
0.108	$1.134 \pm 0.024$	$1.288\pm0.087$	$1.310 \pm 0.009$	$1.320\pm0.031$
0.161	$1.172 \pm 0.049$	$1.332\pm0.062$	$1.262 \pm 0.038$	$1.224 \pm 0.029$
0.214	$1.171 \pm 0.010$	$1.323 \pm 0.004$	$1.232 \pm 0.016$	$1.188\pm0.061$
0.266	$1.171\pm0.002$	$1.168 \pm 0.006$	$1.204 \pm 0.039$	$1.263\pm0.025$
0.318	$1.274\pm0.088$	$1.304\pm0.005$	$1.308\pm0.012$	$1.362\pm0.040$

#### Table 11

True fracture strain ( $\varepsilon^*$ ) of gelatin gels prepared with A-type gelatin at pH 4 (Gel-4<sub>A</sub>) and solid fat as the lipid phase with varying fraction filler ( $\phi_f$ ) and gelatin concentration. Values are unitless.

	Gelatin concentration			
$arphi_{ m f}$	2%	4%	6%	8%
0	$1.173 \pm 0.033$	$1.335\pm0.026$	$1.477 \pm 0.030$	$1.585\pm0.048$
0.054	$1.030 \pm 0.034$	$1.300 \pm 0.045$	$1.419 \pm 0.078$	$1.504\pm0.047$
0.108	$1.020\pm0.024$	$1.120 \pm 0.032$	$1.281 \pm 0.013$	$1.288\pm0.030$
0.161	$0.976 \pm 0.023$	$1.063 \pm 0.031$	$1.161 \pm 0.023$	$1.163 \pm 0.013$
0.214	$0.929\pm0.009$	$0.959\pm0.009$	$1.005 \pm 0.002$	$1.092\pm0.003$
0.266	$0.844 \pm 0.003$	$0.877 \pm 0.004$	$0.952 \pm 0.019$	$1.027 \pm 0.013$
0.318	$0.764\pm0.017$	$0.793\pm0.030$	$0.852\pm0.029$	$0.929\pm0.009$

## Table 12

True fracture strain ( $\varepsilon^*$ ) of gelatin gels prepared with A-type gelatin at pH 4 (Gel-4<sub>A</sub>) and liquid oil as the lipid phase with varying fraction filler ( $\phi_f$ ) and gelatin concentration. Values are unitless.

	Gelatin concentration			
$arphi_{ m f}$	2%	4%	6%	8%
0	$1.184 \pm 0.102$	$1.377 \pm 0.066$	$1.498\pm0.079$	$1.664 \pm 0.101$
0.054	$1.068 \pm 0.033$	$1.424 \pm 0.003$	$1.276 \pm 0.055$	$1.510\pm0.026$
0.108	$1.105 \pm 0.095$	$1.312 \pm 0.154$	$1.264 \pm 0.070$	$1.399\pm0.017$
0.161	$1.060 \pm 0.087$	$1.231 \pm 0.162$	$1.172 \pm 0.055$	$1.268\pm0.018$
0.214	$1.016 \pm 0.102$	$1.121 \pm 0.146$	$1.170 \pm 0.010$	$1.241\pm0.022$
0.266	$0.847 \pm 0.019$	$1.129 \pm 0.131$	$1.114 \pm 0.018$	$1.112 \pm 0.006$
0.318	$0.831\pm0.053$	$1.095\pm0.148$	$1.063\pm0.060$	$1.102\pm0.025$

## Table 13

True fracture stress ( $\sigma^*$ ) of gelatin gels prepared with B-type gelatin at pH 6 (Gel-6<sub>B</sub>) and solid fat as the lipid phase with varying fraction filler ( $\phi_f$ ) and gelatin concentration. Values are reported in kPa.

$arphi_{ m f}$	Gelatin concentration			
	2%	4%	6%	8%
0	$9.84\pm0.24$	$21.42\pm0.13$	32.66 ± 1.37	$51.86\pm0.42$
0.054	$10.14 \pm 0.43$	$20.45 \pm 2.32$	$31.71 \pm 0.07$	$52.04 \pm 2.99$
0.108	$8.72\pm0.78$	$20.74 \pm 0.56$	$34.04 \pm 0.47$	$48.57\pm0.67$
0.161	$8.39 \pm 0.51$	$19.37 \pm 0.14$	$30.49\pm0.89$	$47.66 \pm 0.19$
0.214	$8.57 \pm 0.05$	$18.54 \pm 0.99$	$30.45 \pm 0.18$	$43.52 \pm 0.56$
0.266	9.40 ± 0.17	$19.86 \pm 0.80$	$29.09 \pm 3.87$	$45.14 \pm 1.94$
0.318	$9.81\pm1.14$	$20.52 \pm 1.17$	$31.62~\pm~1.45$	$53.61  \pm  3.48$

True fracture stress ( $\sigma^*$ ) of gelatin gels prepared with B-type gelatin at pH 6 (Gel-6<sub>B</sub>) and liquid oil as the lipid phase with varying fraction filler ( $\phi_f$ ) and gelatin concentration. Values are reported in kPa.

	Gelatin concentration			
$arphi_{ m f}$	2%	4%	6%	8%
0	9.13 ± 1.22	$29.95 \pm 0.43$	$39.88 \pm 4.24$	$59.23 \pm 8.73$
0.054	$9.78\pm2.37$	$25.01 \pm 0.06$	$36.75 \pm 5.79$	$52.01 \pm 3.48$
0.108	$9.76 \pm 0.34$	$21.91 \pm 2.64$	$33.45 \pm 3.44$	$45.16 \pm 1.16$
0.161	8.13 ± 1.12	$23.06\pm0.35$	$29.99\pm2.90$	$43.72 \pm 1.28$
0.214	$7.24 \pm 1.41$	$21.65 \pm 2.44$	$27.87 \pm 1.56$	$42.88 \pm 1.80$
0.266	$7.04 \pm 1.29$	$19.49 \pm 3.34$	$27.56 \pm 4.03$	$39.95 \pm 2.65$
0.318	$6.18\pm1.36$	$16.87\pm4.40$	$26.65\pm5.33$	$34.57\pm2.70$

## Table 15

True fracture stress ( $\sigma^*$ ) of gelatin gels prepared with A-type gelatin at pH 4 (Gel-4<sub>A</sub>) and solid fat as the lipid phase with varying fraction filler ( $\phi_f$ ) and gelatin concentration. Values are reported in kPa.

	Gelatin concentration			
$arphi_{ m f}$	2%	4%	6%	8%
0	$8.82\pm0.82$	$21.24 \pm 1.41$	$34.52\pm2.66$	$62.70 \pm 3.15$
0.054	$10.54 \pm 0.48$	$21.81 \pm 0.41$	$38.66 \pm 3.50$	$62.41 \pm 5.96$
0.108	$9.75 \pm 0.68$	$22.57 \pm 0.40$	$40.72 \pm 4.15$	$63.19\pm6.86$
0.161	$11.17 \pm 0.11$	$24.87\pm0.84$	$41.97 \pm 1.99$	$62.20 \pm 3.54$
0.214	$10.95 \pm 0.40$	$29.16 \pm 1.96$	$46.31 \pm 3.72$	$68.33 \pm 1.53$
0.266	$12.20 \pm 0.35$	$30.44 \pm 0.91$	$47.95 \pm 5.03$	$67.57 \pm 0.96$
0.318	$14.65\pm0.94$	$33.72\pm2.27$	$53.66 \pm 2.97$	$68.01\pm3.18$

## Table 16

True fracture stress ( $\sigma^*$ ) of gelatin gels prepared with A-type gelatin at pH 4 (Gel-4<sub>A</sub>) and liquid oil as the lipid phase with varying fraction filler ( $\phi_f$ ) and gelatin concentration. Values are reported in kPa.

	Gelatin concentration				
$arphi_{ m f}$	2%	4%	6%	8%	
0	$7.08~\pm~1.45$	$21.26\pm3.13$	$51.23\pm8.52$	$59.56 \pm 2.91$	
0.054	$7.87\pm0.99$	$21.91 \pm 4.71$	$41.54 \pm 7.06$	$67.22~\pm~3.16$	
0.108	7.83 ± 1.24	$21.69 \pm 0.06$	$48.55 \pm 11.94$	$53.56 \pm 0.48$	
0.161	$7.98~\pm~1.42$	$23.69 \pm 2.64$	$46.64 \pm 13.42$	$60.00\pm6.94$	
0.214	$8.96 \pm 0.71$	$25.54 \pm 1.72$	$47.55 \pm 7.69$	$56.45 \pm 1.76$	
0.266	$9.43\pm0.46$	$26.48 \pm 0.67$	$51.01 \pm 8.39$	$51.89\pm0.49$	
0.318	$11.55\pm1.25$	$25.34\pm0.41$	$47.07\pm5.50$	51.59 ± 3.79	

## 2. Experimental Design, Materials and Methods

## 2.1. Experimental design

Emulsion-filled gelatin gels were prepared at either pH 6 using either B-Type gelatin (Gel-6<sub>B</sub>) or A-Type gelatin (Gel-6<sub>A</sub>; only E<sub>c</sub> reported) or pH 4 using A-Type gelatin (Gel-4<sub>A</sub>). Variation in pH was used to induce either a homogeneous filler distribution or heterogeneous network architecture [1,2]. The emulsion droplets were added at fixed intervals of 0, 5, 10, 15, 20, 25, and 30 wt% emulsion in the final formulation, corresponding to a filler volume fraction  $\phi_f = 0$ , 0.054, 0.108, 0.161, 0.214, 0.266, and 0.318, respectively. For each gelatin type and pH, composites were prepared with emulsions containing either solid fat or liquid oil as the lipid phase. All formulations were repeated at gelatin concentrations of 2, 4, 6, and 8 wt% gelatin in the gel phase.

## 2.2. Materials

A-Type (porcine skin) and B-Type (bovine bone) gelatin were obtained from Nitta Gelatin NA Inc. (Toronto, ON, Canada). Both gelatin types had a bloom strength of 250. Whey protein isolate (WPI; BiPro<sup>TM</sup>, 91% protein) was purchased through Davisco Foods International (Le Sur, MN, USA). High oleic canola oil was sourced from a local retailer, and fully hydrogenated soybean oil flakes were purchased from Stratas Foods LLC. (Memphis, TN, USA). All solutions were prepared with potable deionized water. NaOH and HCl used for pH adjustment were obtained from Fisher Scientific (Ottawa, ON, Canada).

## 2.3. Methods

## 2.3.1. Emulsion preparation

All stock WPI-stabilized emulsions (40 wt% lipid) were prepared using a pilot-scale high pressure homogenizer unit (Model M110EH; Microfluidics, Westwood, MA, USA). A 1 wt% WPI solution was prepared by mixing for 2 h under gentle mixing (initial hydration) and subsequently heating to ~45 °C for 1 hour under mixing (full protein dissolution). The lipid phase used was liquid high oleic canola oil ("Oil") or a pre-mixed 60/40 blend of hydrogenated soy flakes with high oleic canola oil ("Fat"). The latter produced a solid fat at room temperature, but reduced the melting point to ~60 °C. The appropriate lipid phase was heated to 70 °C and combined with the 1 wt% WPI solution at a water/lipid ratio of 60:40. Crude emulsions were formed by mixing for 2 min at ~12,500 rpm with a IKA T18 Basic high shear mixer outfitted with a S18N-19 G dispersing probe (IKA Works Inc., Wilmington, NC, USA). Crude emulsion was then processed in the high pressure homogenizer (4 passes at 350 bar). The homogenizer unit was maintained above 70 °C to ensure the lipid phase remained liquid during processing. The emulsions were then cooled to room temperature before further use.

## 2.3.2. Preparation of emulsion-filled gelatin gels

Stock gelatin solutions were prepared using 3x the target matrix concentration. Powdered gelatin was hydrated in deionized water for  $\sim$ 30 min under gentle mixing, and subsequently heated to 70 °C in an incubator until use (minimum 30 min). pH of the stock gelatin was adjusted with dilute NaOH or HCl prior to emulsion addition. Stock emulsion was diluted with deionized water to achieve the target formulation, and pre-heated to  $\sim$ 70 °C in an incubator. Addition of emulsion did not notably alter pH of stock gelatin.

All samples were prepared in 35 g batches. Gelatin was added to diluted emulsion, mixed using a benchtop stir plate, and divided into three 10 ml disposable polystyrene beakers coated with a thin layer of mineral oil. Beakers were then placed in an ice bath to immediately induce gelation and avoid filler aggregation [3]. After 30 min, samples were refrigerated at 8 °C overnight, prior to further analysis.

## 2.3.3. Large deformation mechanical analysis

Mechanical analysis was carried out using a uniaxial compression test on a texture analyzer (Model TA.XT2, Stable Micro Systems, Texture Technologies Corp., Scarsdale, NY, USA) outfitted with a 30 kg load cell and a 200 mm diameter cylindrical aluminum probe. Samples were extracted from the disposable beakers (diameter 22 mm) and cut into 10 mm tall cylindrical pucks with a dye (fabricated in-house). A thin layer of mineral oil was applied to the upper and lower plates to reduce interfacial friction. Each gel was compressed for a single cycle to a strain of 85% at cross-head speed of 1.0 mm/s. All tests were performed immediately after removing each sample from the refrigerator. Data was collected and analyzed using the Exponent software (Stable Micro Systems).

The apparent elastic constant was taken as the slope of the linear region at the onset of compression of the force-deformation profile (see Fig. 1); i.e., strain  $\leq 10\%$  [3], and the elastic (Young's) modulus was determined based on the sample geometry (all values reported in kPa).



**Fig. 1.** Representative force/deformation profile (Gel-6<sub>B</sub>, 6% gelatin,  $\phi_f = 0.20$ ) denoting analysis parameters. Fracture point denoted by arrow. Inset highlights initial linear region, with dashed lines indicating region of slope determination ( $\leq$ 10% strain).

True fracture strain was taken as  $\varepsilon^* = ln(h^*/h)$ , where *h* and *h*<sup>\*</sup> denote the initial sample height and the height at fracture, respectively (values are unitless). True fracture stress is given by  $\sigma^* = F^*/A^*$ , where  $F^*$  and  $A^*$  are the force and cross-sectional area at the fracture point (all values reported in kPa).

## **Declaration of Competing Interest**

All authors declare they have no known competing financial interests or personal relationships which have, or could be perceived as having influenced the work reported in this article.

## **CRediT Author Statement**

**Andrew J. Gravelle:** Conceptualization, Methodology, Validation, Formal analysis, Investigation, Writing – original draft, Writing – review & editing, Visualization, Funding acquisition; **Alejandro G. Marangoni:** Conceptualization, Resources, Writing – review & editing, Supervision, Funding acquisition.

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## Supplementary Materials

Supplementary material associated with this article can be found in the online version at doi:10.1016/j.dib.2021.107410.

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