Supporting Information

Ultra-Stable Water-in-Oil High Internal Phase Emulsions Featuring Interfacial and Biphasic Network Stabilization

Michelle C. Lee¹, Chen Tan¹, Raheleh Ravanfar¹, Alireza Abbaspourrad^{1*}

¹Department of Food Science, Cornell University, Stocking Hall, Ithaca, NY 14853,

USA

*Corresponding Author: alireza@cornell.edu; +1 607 255-2923

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Figure S1. Stability of HIPEs made from (A) only 1 wt% GMO, (B) only 3 wt% beeswax, and (C) 1 wt% GMO and 3 wt% beeswax in the oil phase stored at 25°C for one day.

Figure S2. Particle size distribution of O-HIPEs made with 1 wt% GMO and 3 wt% beeswax in the external phase and internal volume fractions of 0.70, 0.75, 0.80, and 0.85.

Figure S3. Stability of O-HIPEs made with 1 wt% GMO and 3 wt% beeswax in the external phase at internal volume fractions of 0.80 and 0.85. The samples were stored for 2 days at 25 °C.

Figure S4. Interfacial tension of water or 1 wt% carrageenans (internal phase) dispensed in different external phases. The interfacial tension measurements were conducted in a temperature-controlled chamber at 50 °C. All measurements are expressed in the unit of mN/m.

Figure S5. Interfacial tension of water (internal phase) dispensed in oil external phases composed of 1 wt% GMO and different beeswax concentration (0.5-10 wt%). The interfacial tension measurements were conducted in a temperature-controlled chamber at 50 °C. All measurements are expressed in the unit of mN/m.

Figure S6. (A)Visual images and (B) rheological measurement of bulk oleogel made with beeswax at different weight concentrations.

Figure S7. Particle size distribution of O-HIPEs made with 1 wt% GMO and beeswax concentrations of 0.5, 1, 3, 5, and 10 wt% in the external phase.

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Figure S9. Gel-in-gel HIPEs featuring different *i*-carrageenan concentrations. (A) Images and (B) rheological measurements of gel-in-gel HIPEs prepared with different *i*-carrageenan concentrations in the internal phase. (C) The optical microscopy showing microstructure of gel-in-gel HIPEs with different *i*-carrageenan concentrations.

Figure S10. Particle size distribution of gel-in-gel HIPEs made with various types of carrageenans (κ , ι , λ) in the internal phase at 1 wt% and 1 wt% GMO and 3 wt% beeswax in the external phase.

Figure S11. Images of the O-HIPE and gel-in-gel HIPEs made from κ -carrageenan (κ -car), 1-carrageenan (1-car), and λ -carrageenan (λ -car) stored over time under vacuumed

conditions.

Figure S12. Images of the O-HIPE and gel-in-gel HIPEs made from κ -carrageenan (κ -car), 1-carrageenan (1-car), and λ -carrageenan (λ -car) stored over time under 37 °C.

Figure S13. (A) Demonstration of the stability of an anthocyanin-incorporated ι carrageenan gel-in-gel HIPE at different pH and 25 °C. (B) Demonstration of β carotene and anthocyanin co-encapsulated in the gel-in-gel HIPE after extraction using n-Hexanes.

Figure S14. O-HIPEs featuring different GMO concentrations in the external phase. (A) Images and (B) rheological measurements of O-HIPEs prepared with different GMO concentrations. (C) The optical microscopy showing microstructure of O-HIPEs with different GMO concentrations.

Figure S15. Particle size distribution of O-HIPEs made with 3 wt% beeswax and different GMO concentrations (0.5- 3%) in the external phase at internal volume fractions of 0.75.

Figure S16. Gel-in-gel (ι-carrageenan) HIPEs featuring different GMO concentrations. (A) Images and (B) rheological measurements of gel-in-gel (ι-carrageenan) HIPEs prepared with different GMO concentrations. (C) The optical microscopy showing microstructure of gel-in-gel (ι-carrageenan) HIPEs with different GMO concentrations. **Figure S17.** Three phase contact angle of water on 3% beeswax oleogel film immersed

in either canola oil or canola oil- GMO mixture.

Table S1. Particle size of HIPEs stabilized by solely 1 wt% GMO, solely 3 wt% beeswax, and a combination of 1 wt% GMO + 3 wt% beeswax made at internal volume fractions of 0.75.

Table S2. Particle size of O-HIPEs made from different internal phase volume ratios.

Table S3. Particle size of O-HIPEs made from different beeswax concentrations.

Table S4. Particle size of gel-in-gel HIPEs made from different carrageenans in the internal phase.

Table S5. EDS elemental analysis of carbon, oxygen, and sulfur from the cryo-SEM.

Figures

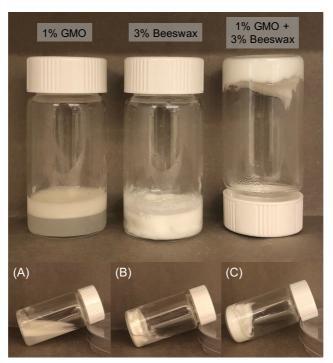


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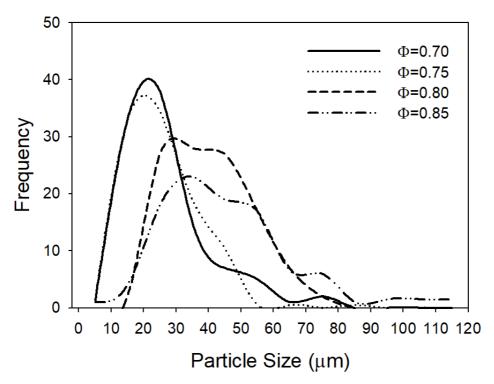


Figure S2. Particle size distribution of O-HIPEs made with 1 wt% GMO and 3 wt% beeswax in the external phase and internal volume fractions of 0.70, 0.75, 0.80, and 0.85.

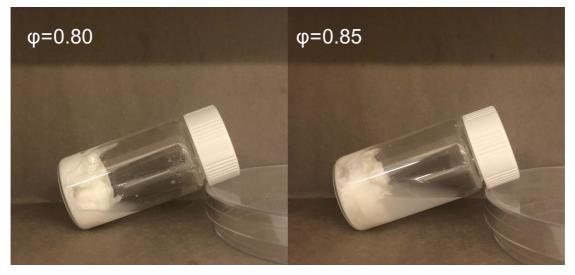


Figure S3. Stability of O-HIPEs made with 1 wt% GMO and 3 wt% beeswax in the external phase at internal volume fractions of 0.80 and 0.85. The samples were stored for 2 days at 25 °C.

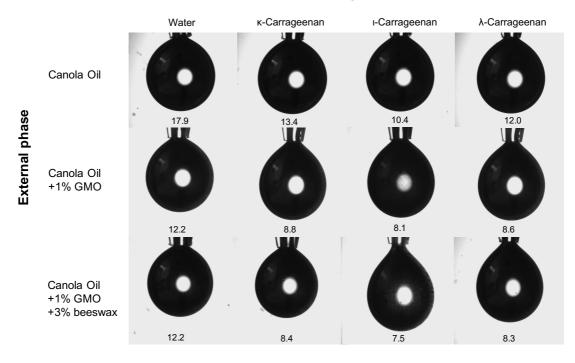


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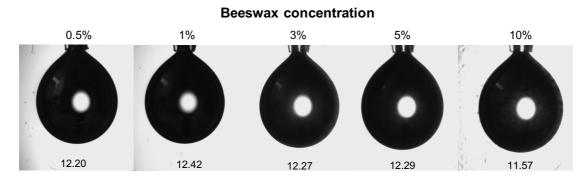


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Internal phase

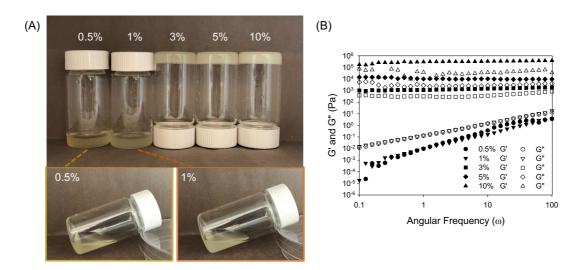


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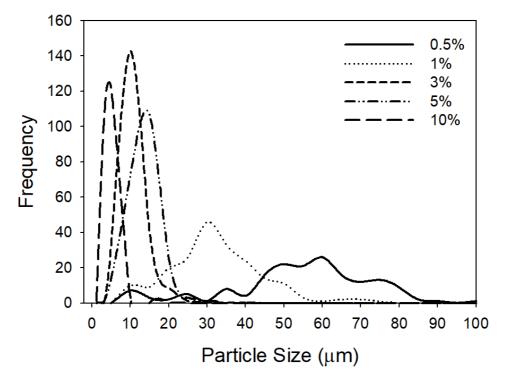


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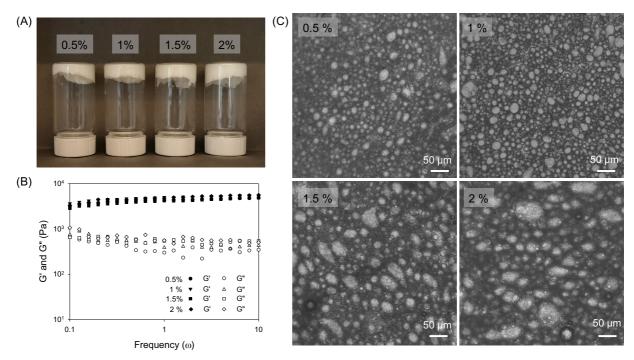


Figure S9. Gel-in-gel HIPEs featuring different 1-carrageenan concentrations. (A) Images and (B) rheological measurements of gel-in-gel HIPEs prepared with different 1-carrageenan concentrations in the internal phase. (C) The optical microscopy showing microstructure of gel-in-gel HIPEs with different 1-carrageenan concentrations.

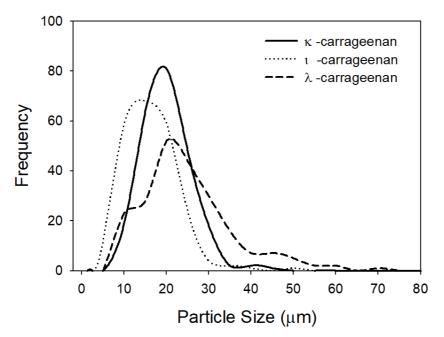


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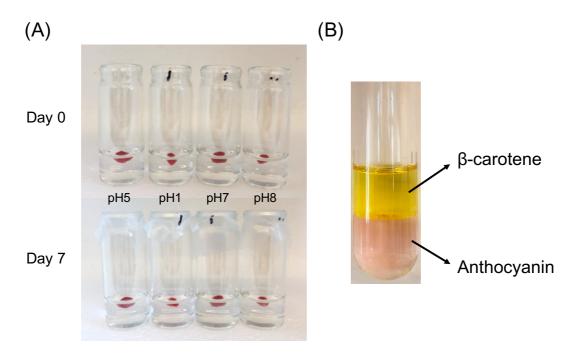


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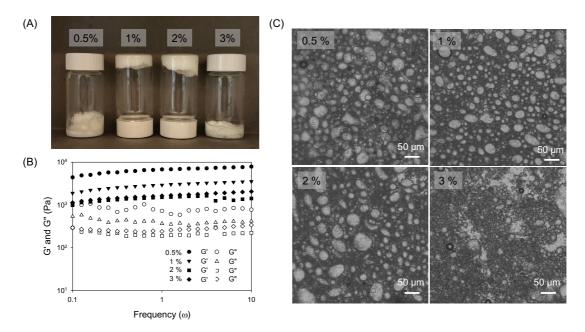


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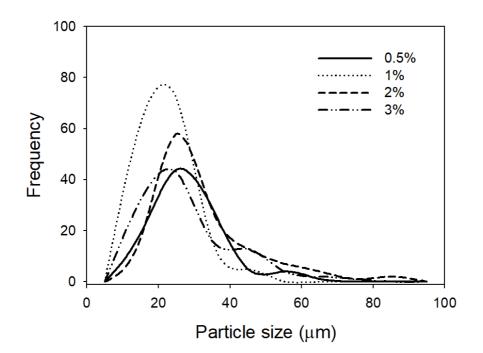


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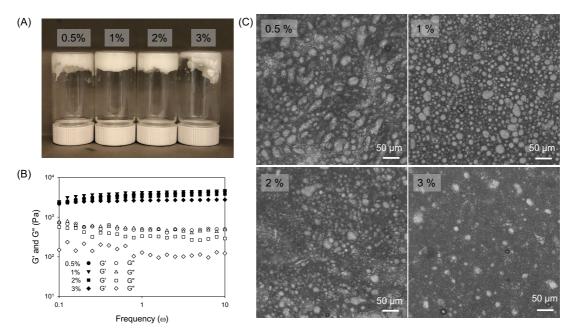


Figure S16. Gel-in-gel (*i*-carrageenan) HIPEs featuring different GMO concentrations. (A) Images and (B) rheological measurements of gel-in-gel (*i*-carrageenan) HIPEs prepared with different GMO concentrations. (C) The optical microscopy showing microstructure of gel-in-gel (*i*-carrageenan) HIPEs with different GMO concentrations.

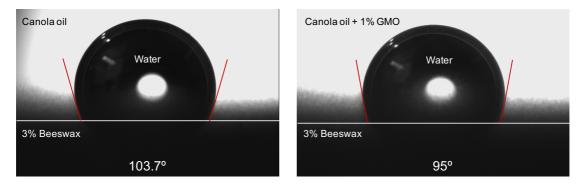


Figure S17. Three phase contact angle of water on 3% beeswax oleogel film immersed in either canola oil or canola oil- GMO mixture.

Tables

Table S1. Particle size of HIPEs stabilized by solely 1 wt% GMO, solely 3 wt% beeswax, and a combination of 1 wt% GMO + 3 wt% beeswax made at internal volume fractions of 0.75.

HIPE type	Emulsion type	Particle Size (µm)		
GMO	O/W	N/A		
Beeswax	W/O	$28.86 \pm 11.33^{\mathrm{a}}$		
GMO + Beeswax	W/O	$17.59\pm6.18^{\mathrm{a}}$		

Table S2. Particle size of O-HIPEs made from different internal phase volume ratios.

	Internal phase volume ratio (Φ)			
	0.70	0.75	0.80	0.85
Particle Size (µm)	27.16 ± 13.41^{a}	$25.78\pm11.22^{\mathrm{a}}$	$40.42\pm14.77^{\text{b}}$	$45.77\pm21.01^{\text{b}}$

Table S3. Particle size of O-HIPEs made from different beeswax concentrations.

	Beeswax concentration (wt%)				
	0.5	1	3	5	10
Particle Size (µm)	$105.25\pm38.34^{\mathrm{a}}$	59.74 ± 23.68^{b}	$16.50 \pm 6.42^{\circ}$	21.90 ± 7.72^{d}	5.75 ± 1.62^{e}

Table S4. Particle size of gel-in-gel HIPEs made from different carrageenans in the internal

	Internal Phase (Carrageenan)			
	К-	1-	λ-	
Particle Size (µm)	$34.90 \pm 11.40^{\mathrm{a}}$	$27.18 \pm 12.95^{\text{b}}$	$45.61 \pm 25.72^{\circ}$	

Element	Internal Phase (Atomic %)			External Phase (Atomic %)				
	Water	к-car	1-car	λ-car	Water	κ-car	1-car	λ-car
С	2.77	2.27	5.62	80.37	89.19	79.99	83.28	89.32
0	97.23	97.30	94.21	18.12	10.81	19.70	15.94	10.68
S	-	-	-	0.58	-	-	-	-

Table S5. EDS elemental analysis of carbon, oxygen, and sulfur from the cryo-SEM.