

## Supplementary Material

# Emulgels Containing Propolis and Curcumin: The Effect of Type of Vegetable Oil, Poly(Acrylic Acid) and Bioactive Agent on Physicochemical Stability, Mechanical and Rheological Properties

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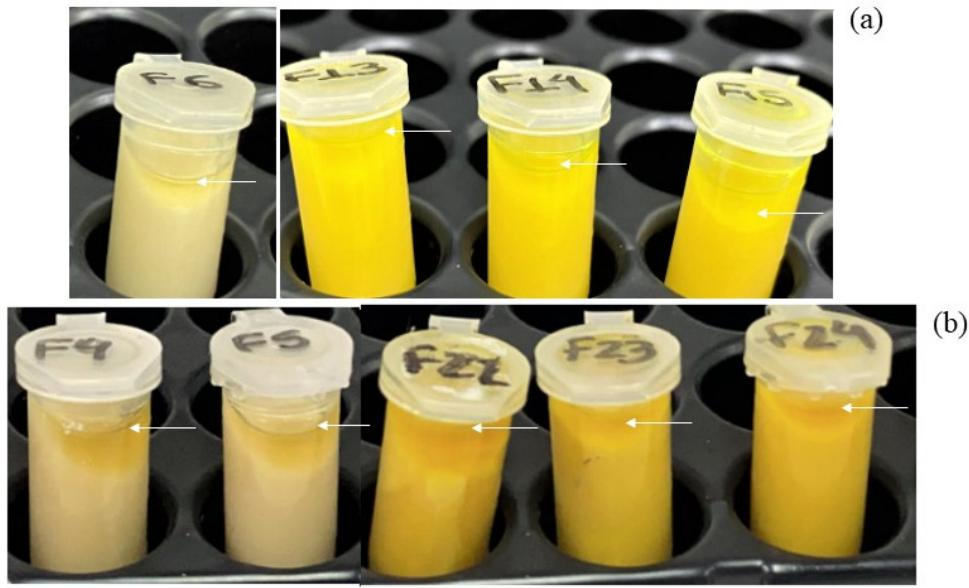
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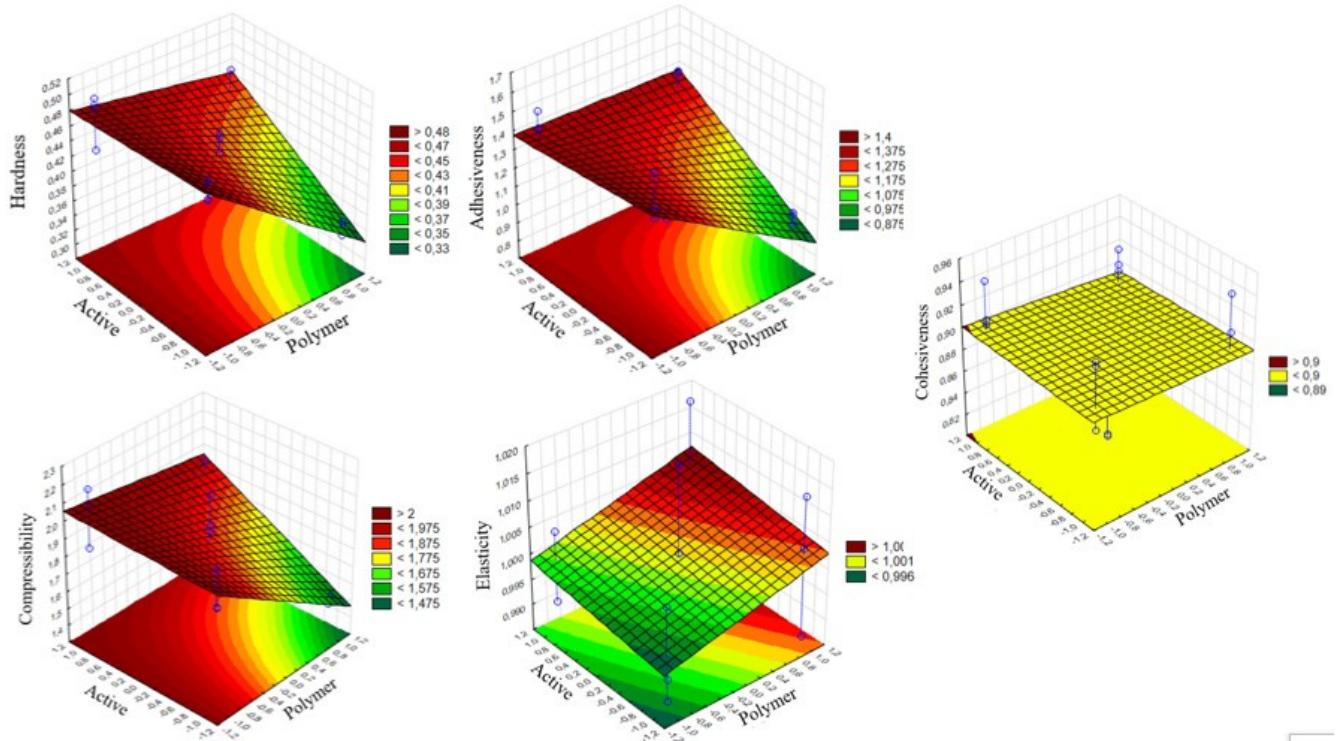
**Table S1.** Mean globule size and polydispersity index (PI) of dispersed phase of the emulgels containing PF or AN oils just after the preparation (T<sub>0</sub>) and after 12 days (T<sub>12</sub>) of the ice-thawing cycle.

Formulations	Mean size ( $\mu\text{m}$ )		PI	
	T <sub>0</sub>	T <sub>12</sub>	T <sub>0</sub>	T <sub>12</sub>
F1	32.13 ± 13.29	29.75 ± 11.89	0.6	0.6
F2	25.03 ± 13.01	27.63 ± 10.03	0.5	0.5
F3	27.11 ± 8.36	33.37 ± 12.66	0.4	0.5
F7	34.04 ± 11.25	37.67 ± 19.05	0.5	0.6
F8	27.28 ± 9.02	28.86 ± 9.77	0.5	0.5
F9	26.94 ± 10.71	25.13 ± 6.74	0.6	0.4
F10	43.49 ± 16.83	57.45 ± 40.99	0.6	1.2
F11	37.16 ± 16.10	46.92 ± 38.24	0.7	0.9
F12	36.45 ± 13.73	46.84 ± 48.19	0.6	1.0
F16	39.02 ± 15.75	54.34 ± 31.86	0.7	0.9
F17	38.20 ± 16.08	52.46 ± 36.71	0.7	0.4
F18	40.83 ± 16.80	47.78 ± 29.01	0.7	0.8
F19	39.84 ± 14.51	27.91 ± 9.99	0.6	0.4
F20	30.93 ± 10.73	32.40 ± 12.29	0.5	0.5
F21	32.35 ± 11.18	24.62 ± 8.40	0.5	0.4
F25	34.24 ± 10.65	34.99 ± 12.30	0.5	0.5
F26	28.39 ± 8.00	31.10 ± 8.95	0.4	0.4
F27	29.90 ± 8.91	30.68 ± 9.81	0.4	0.5

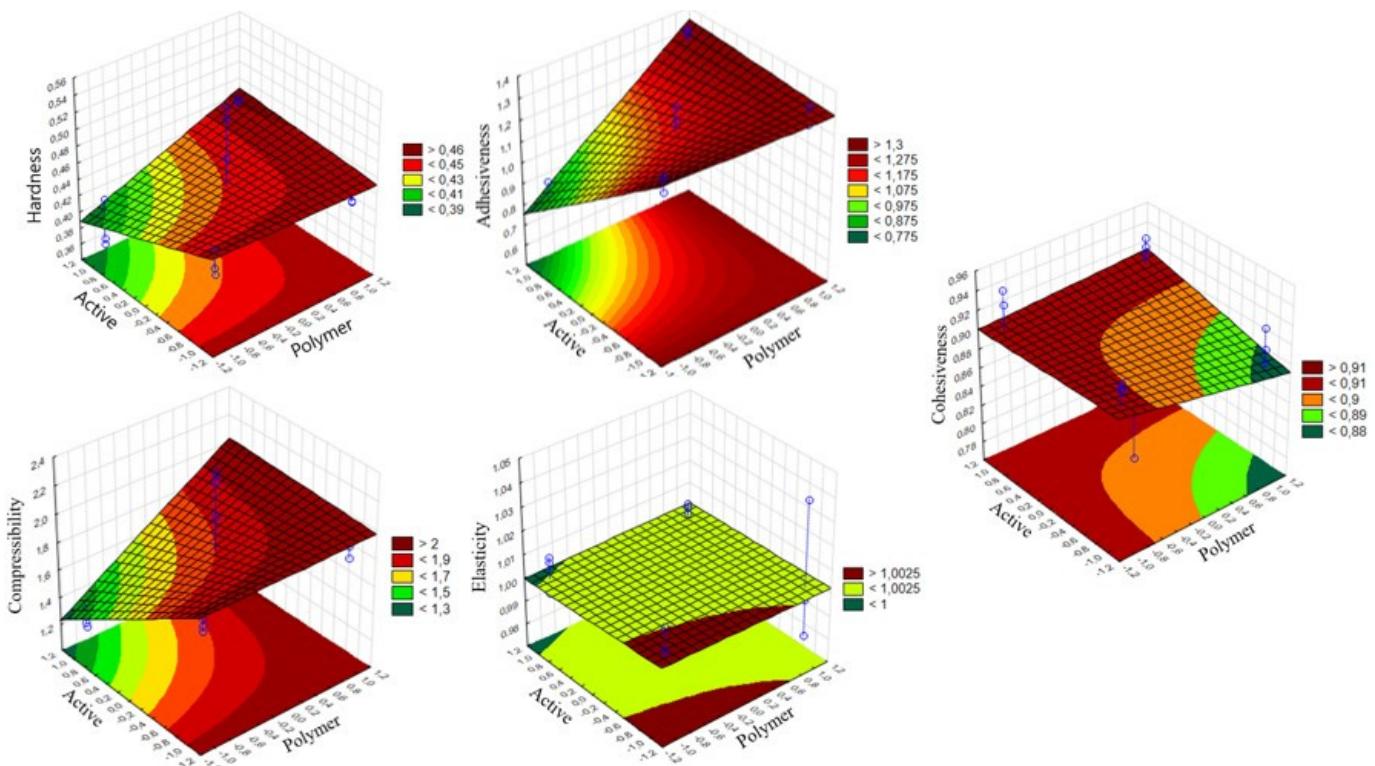
\*± Standard deviation.



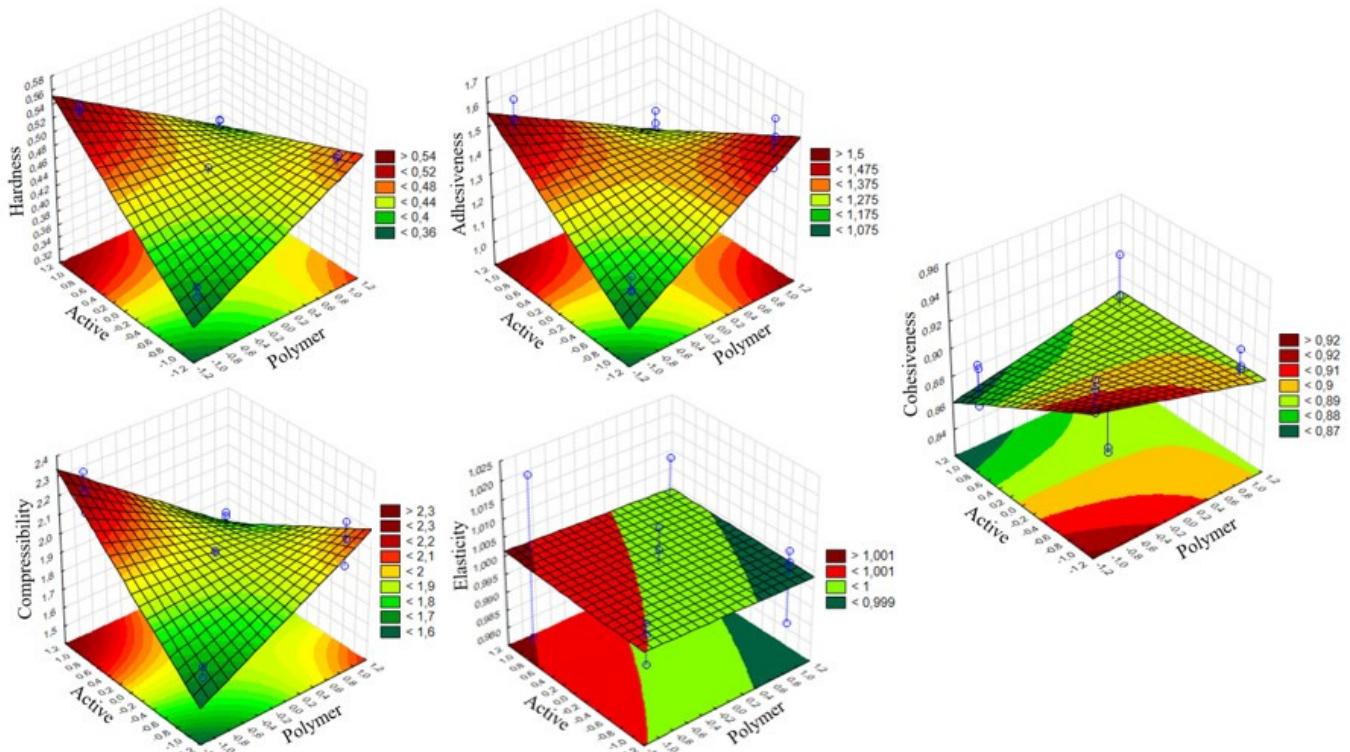
**Figure S1.** Phase separation of the systems during the physicochemical stability analysis at time zero (a): F6, F13, F14, F15; and after 12 days (b): F4, F5, F22, F23, and F24.



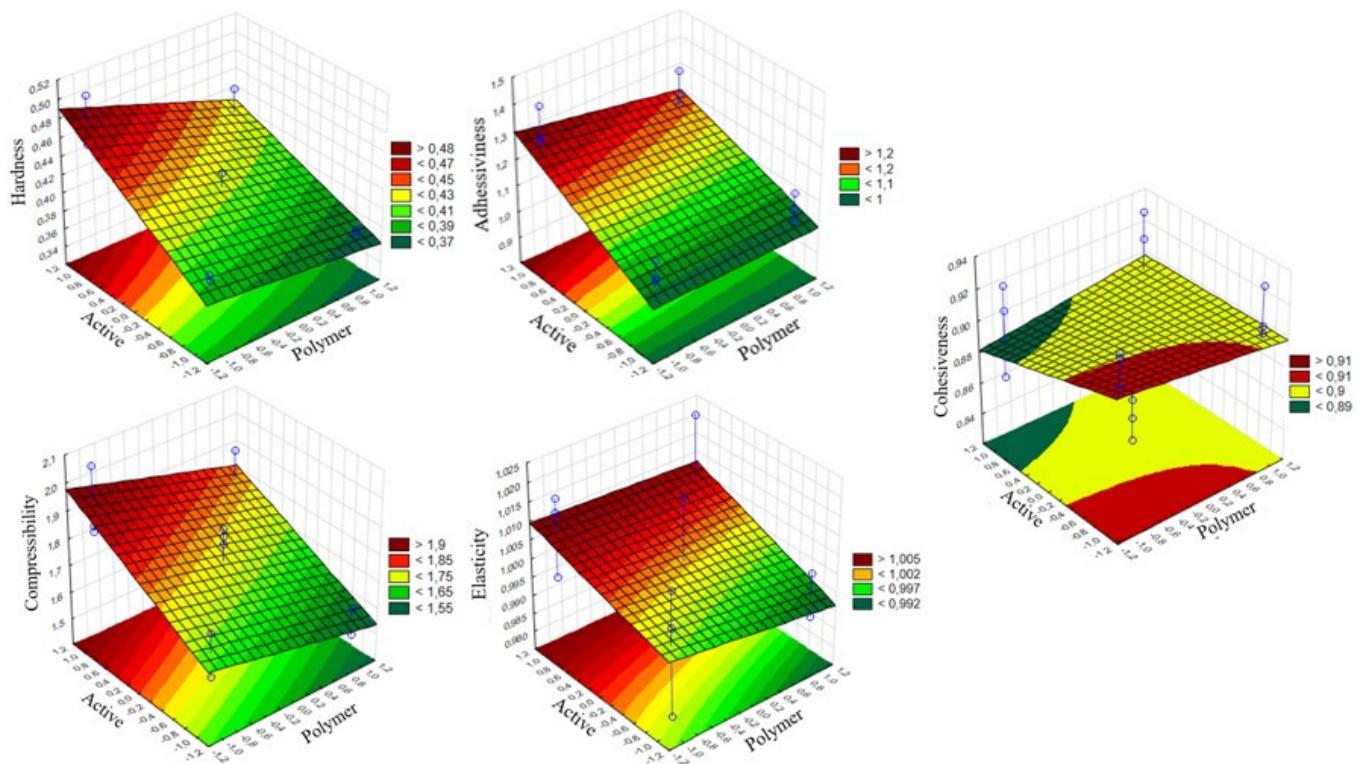
**Figure S2.** Response surface plots of mechanical properties (hardness, compressibility, adhesiveness, elasticity and cohesiveness) of PF emulgels influenced by different polymers (C934P, C974P, PC), different type of bioactive (PE, CUR, PE+CUR) at 25 °C. The color scale is indicated in each figure and shows the isoparametric values.



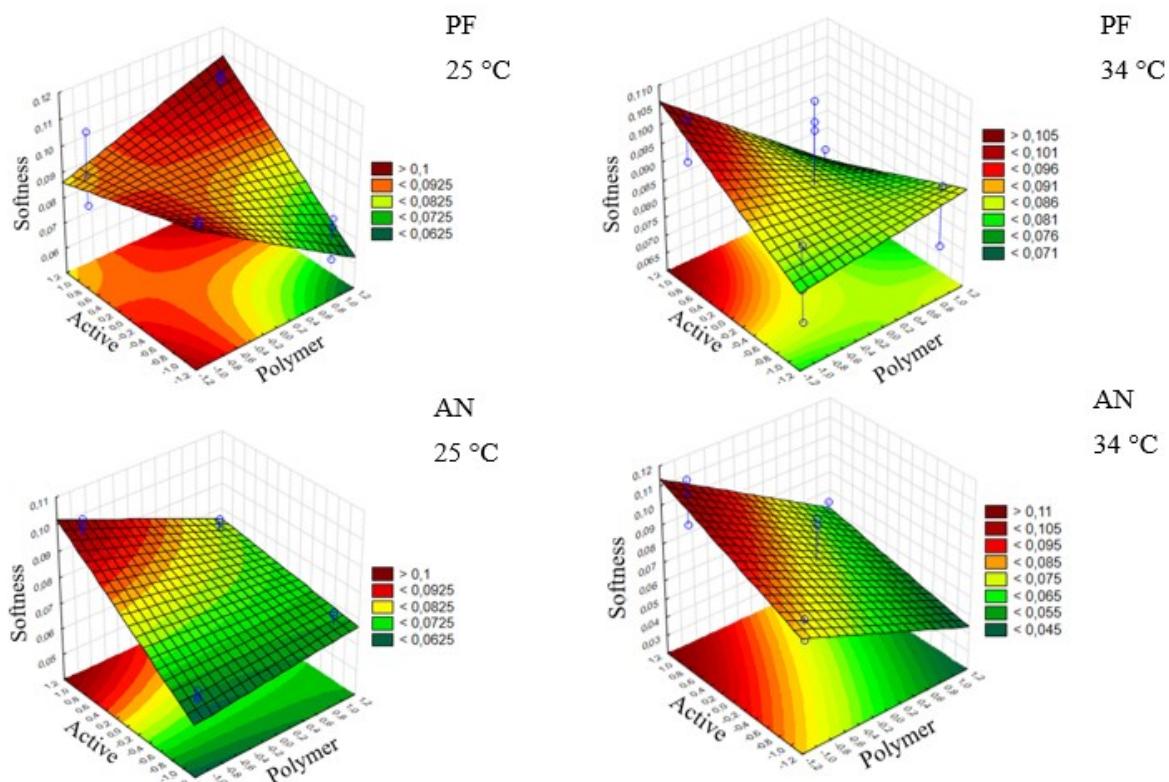
**Figure S3.** Response surface plots of mechanical properties (hardness, compressibility, adhesiveness, elasticity and cohesiveness) of PF emulgels influenced by different polymers (C934P, C974P, PC), different type of bioactive (PE, CUR, PE+CUR) at 34 °C. The color scale is indicated in each figure and shows the isoparametric values.



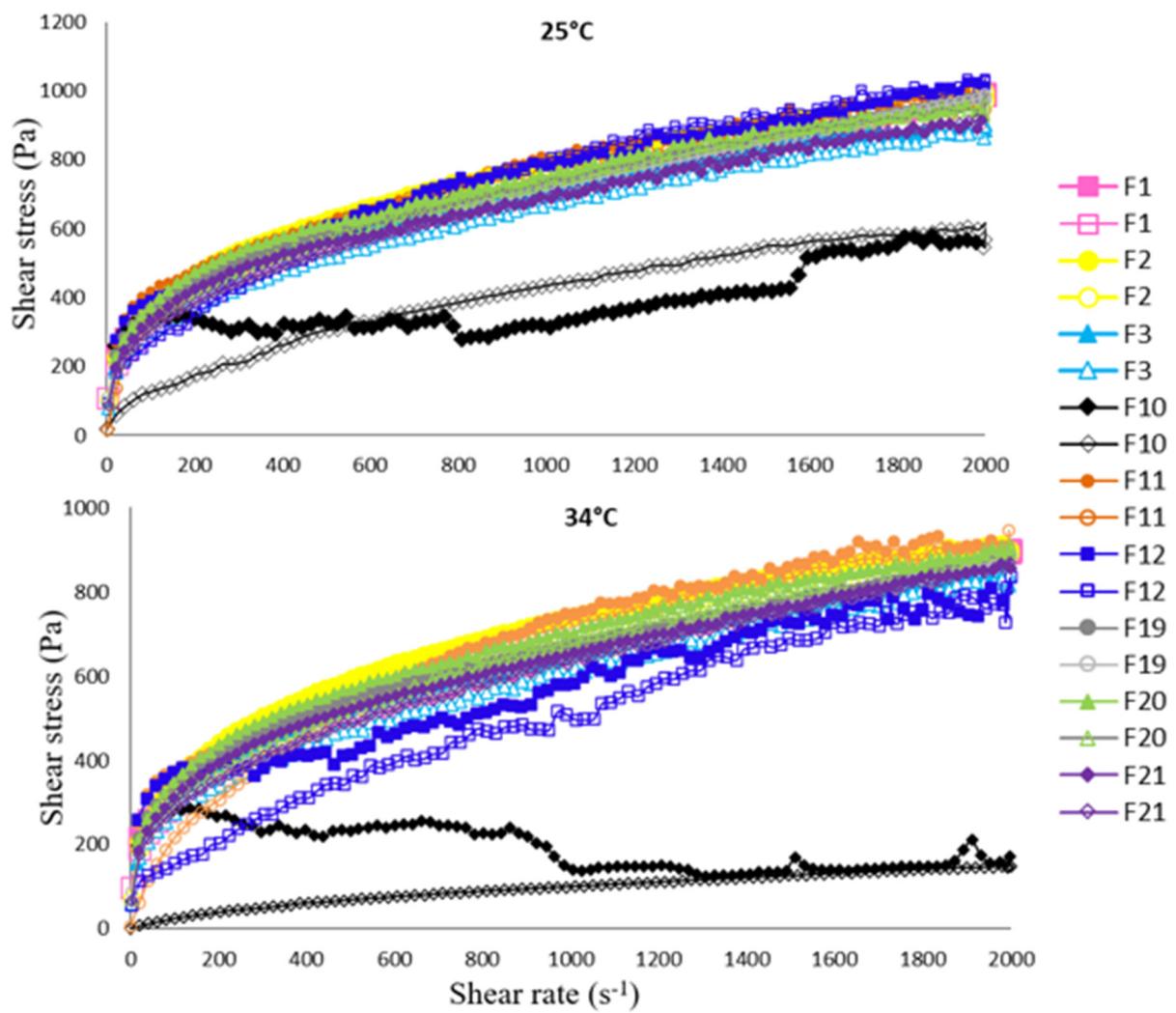
**Figure S4.** Response surface plots of mechanical properties (hardness, compressibility, adhesiveness, elasticity and cohesiveness) of AN emulgels influenced by different polymers (C934P, C974P, PC), different type of bioactive (PE, CUR, PE+CUR) at 25 °C. The color scale is indicated in each figure and shows the isoparametric values.



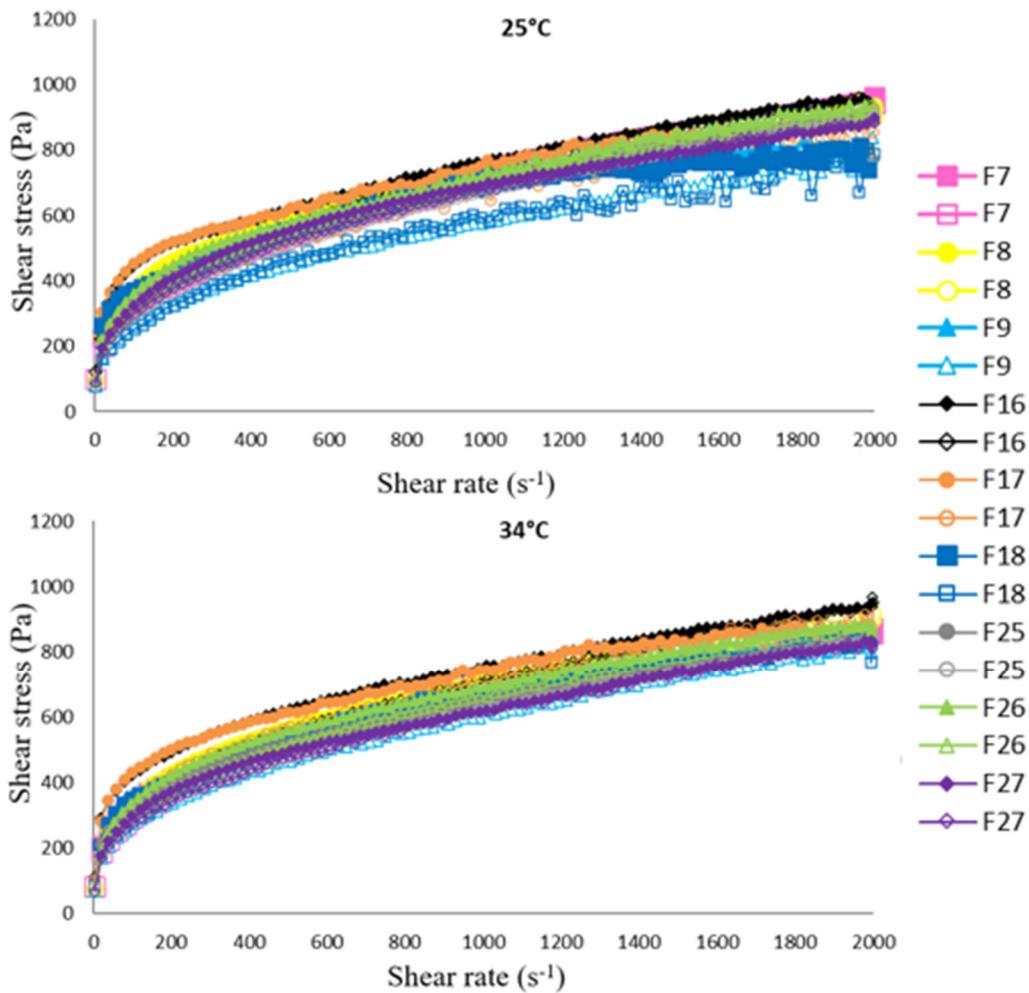
**Figure S5.** Response surface plots of mechanical properties (hardness, compressibility, adhesiveness, elasticity and cohesiveness) of AN emulgels influenced by different polymers (C934P, C974P, PC), different type of bioactive (PE, CUR, PE+CUR) at 34 °C. The color scale is indicated in each figure and shows the isoparametric values.



**Figure S6.** Response surface plots of softness as a function of different polymers (C934P, C974P, PC), different type of bioactive (PE, CUR, PE+CUR) and vegetable oils (PF or AN) for emulgels systems, at 25 and 34 °C. The color scale is indicated in each figure and shows the isoparametric values.



**Figure S7.** Flow curves of the formulations containing PF at different temperatures. Closed symbol represents the upcurve and open symbol represents the downcurve. Standard deviations have been omitted for clarity; however, in all cases, the relative standard deviation of replicate analysis was less than 10%.



**Figure S8.** Flow curves of the formulations containing AN at different temperatures. Closed symbol represents the upcurve and open symbol represents the downcurve. Standard deviations have been omitted for clarity; however, in all cases, the relative standard deviation of replicate analysis was less than 10%.

**Table S2.** Relative standard deviation (%) of emulgels systems F1, F2 and F3, containing PF and PE: elastic modulus ( $G'$ ) and viscous modulus ( $G''$ ) at 25 and 34 °C.

Frequency (Hz)	Relative standard deviation (%)											
	F1				F2				F3			
	25 °C		34 °C		25 °C		34 °C		25 °C		34 °C	
0.10	6.84	6.93	6.10	5.57	8.72	9.37	4.32	7.44	1.44	6.94	4.79	3.82
0.15	5.73	7.55	1.93	3.02	5.00	3.74	1.19	5.67	2.12	2.33	1.88	9.37
0.22	5.14	6.43	1.58	2.79	4.09	7.12	1.66	9.14	2.91	4.47	3.89	2.93
0.32	1.73	8.24	1.92	7.75	3.88	9.07	1.93	4.05	3.45	6.02	3.73	8.39
0.46	2.39	9.97	2.07	1.07	4.16	8.20	2.24	1.57	3.74	8.31	0.49	7.15
0.68	2.13	7.87	2.04	7.02	4.86	6.87	2.33	3.03	3.87	8.44	3.28	4.11
1.00	1.70	5.71	2.13	5.86	5.03	8.84	2.41	1.72	3.78	7.93	0.60	8.90
1.47	1.91	8.39	2.19	4.71	4.93	3.99	2.31	2.55	3.79	7.37	7.74	3.57
2.15	1.24	3.26	2.19	4.42	5.69	3.95	2.20	1.10	3.70	6.74	9.74	8.71
3.16	2.05	4.05	2.07	2.20	4.89	3.03	2.16	0.17	3.58	5.77	8.64	8.40
4.64	1.37	4.27	1.99	2.35	4.90	4.84	2.17	1.29	3.46	4.59	6.16	9.97
6.81	0.56	3.80	1.86	2.03	4.46	1.10	1.93	2.18	3.14	4.15	5.37	3.12
10.00	1.53	5.02	1.90	0.86	4.50	6.24	1.44	1.59	2.59	2.55	8.58	5.35

**Table S3.** Relative standard deviation (%) of emulgels systems F7, F8 and F9, containing AN and PE: elastic modulus ( $G'$ ) and viscous modulus ( $G''$ ) at 25 and 34 °C.

Frequency (Hz)	Relative standard deviation (%)											
	F7				F8				F9			
	25 °C		34 °C		25 °C		34 °C		25 °C		34 °C	
Frequency (Hz)	$G'$	$G''$	$G'$	$G''$	$G'$	$G''$	$G'$	$G''$	$G'$	$G''$	$G'$	$G''$
0.10	0.75	7.45	4.01	1.94	6.12	5.79	3.36	5.23	8.63	5.46	7.84	3.78
0.15	1.38	1.05	6.75	5.25	1.71	3.62	4.33	3.97	6.39	6.53	7.71	3.50
0.22	1.86	8.86	9.82	4.40	2.48	3.28	3.87	1.97	8.38	7.52	7.82	7.09
0.32	2.20	3.15	6.28	8.75	6.18	6.94	3.79	2.54	2.34	9.76	7.88	5.42
0.46	2.31	5.33	6.02	1.82	7.95	5.44	3.89	0.80	3.42	4.42	7.96	4.45
0.68	2.49	9.54	2.08	6.09	5.68	4.84	3.87	6.54	4.31	4.15	8.00	0.72
1.00	2.63	8.88	4.37	8.01	4.37	9.91	3.76	3.01	3.77	8.81	7.82	0.17
1.47	2.70	7.83	6.71	8.03	4.98	9.84	3.73	9.81	5.01	0.95	7.70	1.01
2.15	2.79	7.17	6.71	6.59	4.00	8.19	3.89	8.60	2.15	9.57	7.58	0.87
3.16	2.79	6.34	6.67	8.32	6.59	2.46	3.87	8.76	5.10	3.84	7.44	2.36
4.64	2.74	4.89	6.29	7.04	3.36	6.00	3.90	6.51	5.26	3.21	7.17	2.86
6.81	2.63	4.61	5.92	7.31	4.97	3.38	3.88	8.01	3.89	1.18	6.86	3.06
10.00	2.17	3.33	4.94	2.43	1.51	8.67	3.36	2.40	0.84	8.86	6.27	6.23

**Table S4.** Relative standard deviation (%) of emulgels systems F10, F11 and F12, containing PF and CUR: elastic modulus ( $G'$ ) and viscous modulus ( $G''$ ) at 25 and 34 °C.

Frequency (Hz)	Relative standard deviation (%)											
	F10				F11				F12			
	25 °C		34 °C		25 °C		34 °C		25 °C		34 °C	
Frequency (Hz)	$G'$	$G''$	$G'$	$G''$	$G'$	$G''$	$G'$	$G''$	$G'$	$G''$	$G'$	$G''$
0.10	0.85	1.17	8.40	5.99	1.35	2.75	6.04	6.30	1.68	5.75	8.46	2.43
0.15	1.89	9.18	5.75	4.97	1.39	2.70	0.09	6.33	0.48	0.33	9.73	8.22
0.22	4.91	5.70	7.98	7.37	1.36	2.67	4.84	5.67	1.11	1.89	9.23	8.42
0.32	9.48	5.54	4.99	4.95	1.36	2.48	0.00	6.07	0.93	3.98	9.41	6.01
0.46	8.72	5.56	3.08	3.34	1.38	2.44	0.72	5.23	0.16	9.79	9.55	7.90
0.68	8.00	4.94	1.74	4.76	1.47	2.42	1.21	0.99	0.74	9.03	9.59	6.56
1.00	7.40	4.20	0.65	5.68	1.45	2.08	1.48	9.27	0.77	3.55	9.59	6.16
1.47	6.79	8.98	1.94	2.64	0.45	2.30	1.52	9.24	0.15	4.77	9.64	7.87
2.15	6.29	7.95	4.30	4.29	1.46	2.23	2.17	1.05	1.23	4.81	9.78	8.62
3.16	5.97	7.39	5.42	2.99	1.50	2.40	2.12	9.68	0.66	9.14	9.34	6.25
4.64	5.34	5.80	6.00	8.91	1.47	2.44	1.74	2.53	0.22	9.20	9.26	7.13
6.81	5.18	7.55	7.51	6.10	1.36	3.71	1.79	0.43	0.14	1.48	9.86	7.80
10.00	4.87	1.47	7.47	2.62	1.25	2.09	1.94	7.64	8.29	9.26	8.39	9.94

**Table S5.** Relative standard deviation (%) of emulgels systems F16, F17 and F18, containing AN and CUR: elastic modulus ( $G'$ ) and viscous modulus ( $G''$ ) at 25 and 34 °C.

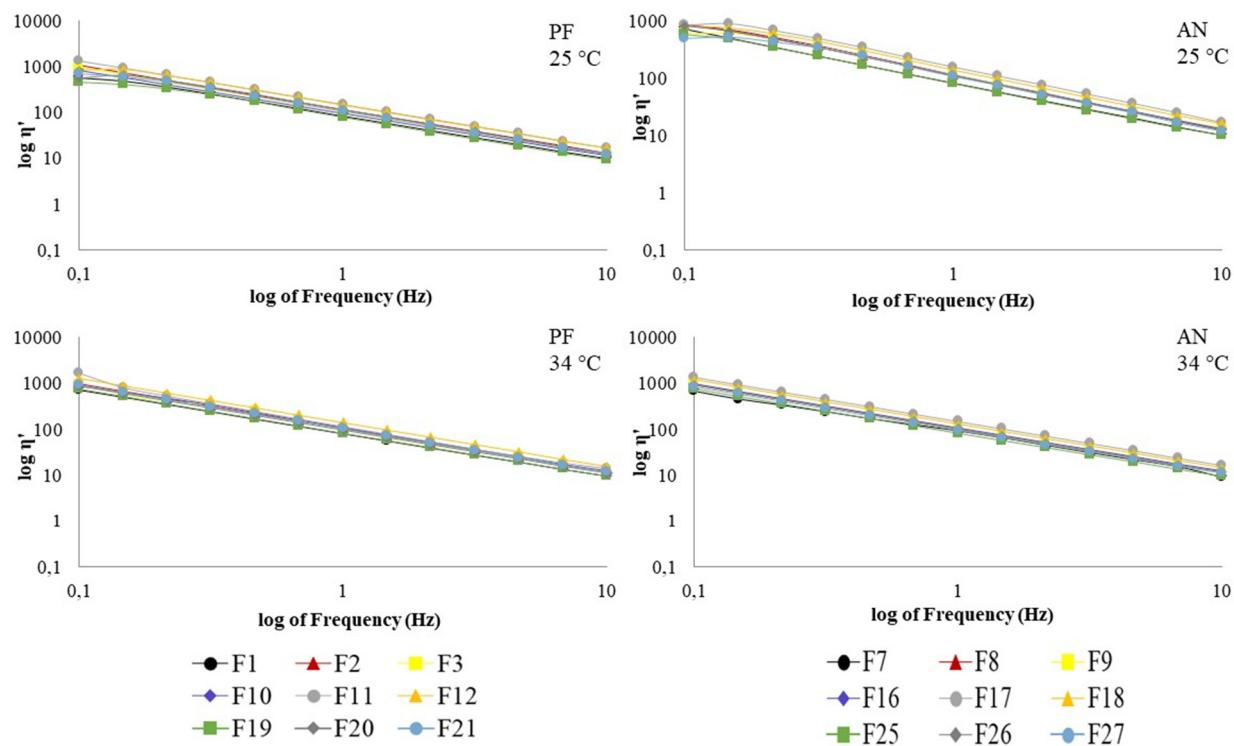
Frequency (Hz)	Relative standard deviation (%)											
	F16				F17				F18			
	25 °C		34 °C		25 °C		34 °C		25 °C		34 °C	
Frequency (Hz)	$G'$	$G''$	$G'$	$G''$	$G'$	$G''$	$G'$	$G''$	$G'$	$G''$	$G'$	$G''$
0.10	8.62	9.71	4.49	9.88	2.42	8.82	5.22	9.78	8.23	8.78	2.83	7.22
0.15	6.11	8.76	2.36	9.92	9.43	7.14	4.71	9.96	9.82	8.70	3.21	4.31
0.22	5.55	9.34	2.41	6.58	5.98	9.02	4.65	9.00	8.71	8.30	3.23	5.66
0.32	2.51	8.63	2.85	3.96	2.39	9.59	4.74	7.44	0.36	8.22	3.17	4.21
0.46	2.29	3.82	2.64	2.57	2.39	6.94	4.84	7.04	6.10	9.21	3.06	2.89
0.68	3.99	4.86	2.58	2.75	2.99	9.16	4.90	6.45	4.82	8.98	2.98	4.30
1.00	4.85	1.55	2.63	2.34	2.39	4.46	4.81	5.98	0.26	4.05	2.95	3.99
1.47	2.94	2.53	2.61	1.74	3.87	1.66	4.84	5.06	2.23	7.73	2.90	3.58
2.15	1.49	0.98	2.57	3.43	2.78	8.17	4.89	6.26	2.05	9.04	2.80	3.52
3.16	3.10	3.18	2.95	2.76	3.71	8.07	4.80	5.43	1.15	9.64	2.91	2.29
4.64	2.99	5.52	2.64	3.05	3.99	4.03	4.80	4.51	1.70	6.59	2.74	2.56
6.81	3.39	5.19	2.68	1.70	3.71	4.37	4.62	6.07	1.57	9.38	2.69	2.85
10.00	1.57	4.44	2.49	1.53	4.49	9.57	4.14	8.00	4.48	9.02	2.37	6.29

**Table S6.** Relative standard deviation (%) of emulgels systems F19, F20 and F21, containing PF and PE + CUR: elastic modulus ( $G'$ ) and viscous modulus ( $G''$ ) at 25 and 34 °C.

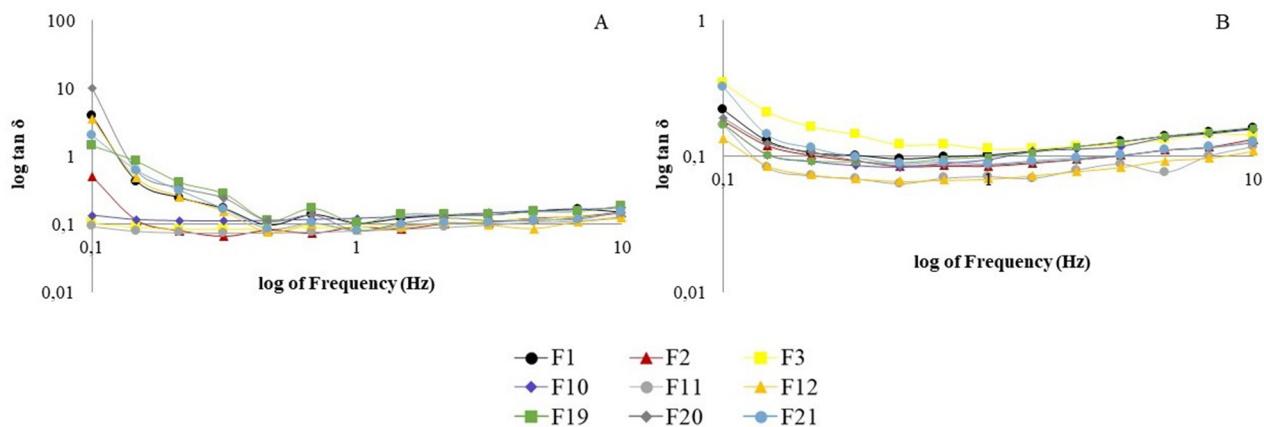
Frequency (Hz)	Relative standard deviation (%)											
	F19				F20				F21			
	25 °C		34 °C		25 °C		34 °C		25 °C		34 °C	
Frequency (Hz)	$G'$	$G''$	$G'$	$G''$	$G'$	$G''$	$G'$	$G''$	$G'$	$G''$	$G'$	$G''$
0.10	1.02	0.01	7.13	9.93	9.66	9.34	2.10	9.36	9.16	9.48	1.90	9.79
0.15	7.53	0.02	3.86	3.91	4.39	5.38	3.51	9.13	9.63	9.12	2.12	9.51
0.22	9.64	0.03	3.55	0.67	7.87	7.59	3.83	6.34	2.53	9.50	3.46	8.93
0.32	8.46	0.05	3.34	2.09	7.10	9.67	3.72	6.81	3.29	9.86	4.18	8.12
0.46	9.55	0.01	3.33	2.73	9.80	5.32	3.59	6.86	5.41	9.88	4.02	8.55
0.68	9.65	0.05	3.26	1.85	6.31	9.45	3.73	6.31	5.52	9.31	3.87	8.18
1.00	9.26	0.01	3.23	2.02	5.99	9.63	3.61	4.46	4.31	8.70	3.41	9.58
1.47	7.17	0.03	3.23	1.74	4.14	9.88	3.59	2.51	4.07	8.46	3.23	7.79
2.15	5.82	0.04	3.22	1.98	4.15	5.65	3.61	9.63	3.81	8.42	2.86	5.28
3.16	5.38	0.01	3.15	2.18	5.29	4.89	3.53	8.28	2.89	6.91	2.51	4.30
4.64	5.82	0.02	3.08	1.14	4.89	8.84	3.50	5.52	4.19	9.94	2.38	2.08
6.81	3.40	0.06	2.98	1.56	3.09	2.61	3.49	4.02	5.18	9.82	2.08	0.56
10.00	4.68	0.19	2.82	3.71	5.30	3.93	3.35	5.49	2.76	9.00	1.98	2.48

**Table S7.** Relative standard deviation (%) of emulgels systems F25, F26 and F27, containing PF and PE + CUR: elastic modulus ( $G'$ ) and viscous modulus ( $G''$ ) at 25 and 34 °C.

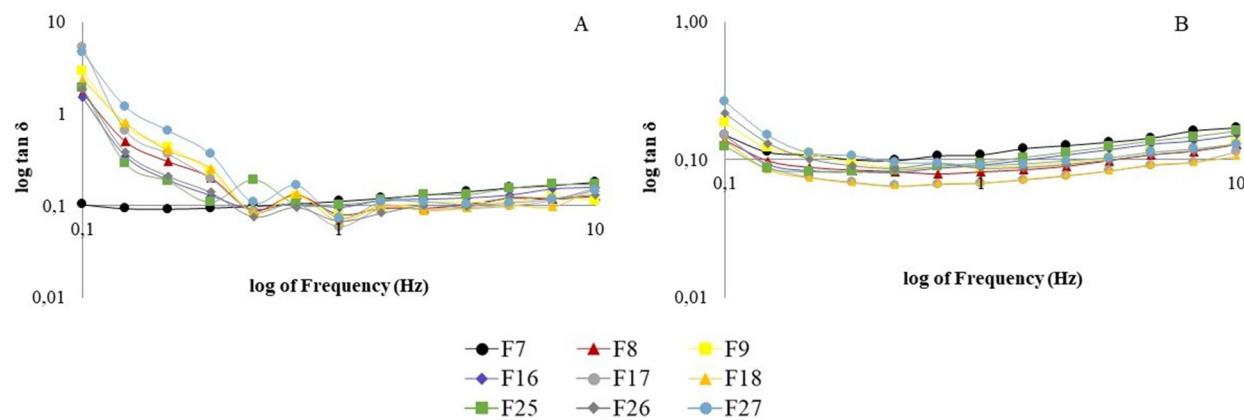
Frequency (Hz)	Relative standard deviation (%)											
	F25				F26				F27			
	25 °C		34 °C		25 °C		34 °C		25 °C		34 °C	
Frequency (Hz)	$G'$	$G''$	$G'$	$G''$	$G'$	$G''$	$G'$	$G''$	$G'$	$G''$	$G'$	$G''$
0.10	8.18	5.62	5.05	9.64	9.44	6.06	7.17	8.68	9.46	9.83	5.16	3.43
0.15	9.39	9.31	5.10	8.39	9.97	7.64	7.96	8.80	7.69	9.73	5.30	8.89
0.22	4.89	9.75	5.34	6.44	4.04	9.11	6.44	9.63	8.50	4.77	5.37	3.35
0.32	0.67	8.57	5.52	5.52	1.84	9.00	5.42	6.22	4.68	5.19	4.26	6.11
0.46	3.97	5.24	5.74	4.14	1.37	8.13	4.84	1.43	6.62	2.46	4.89	7.12
0.68	0.13	6.78	5.78	5.15	2.33	8.37	4.11	6.46	3.03	4.89	4.43	8.40
1.00	1.31	3.31	5.76	4.86	2.35	8.41	3.89	6.28	1.98	9.91	4.13	7.54
1.47	1.14	1.59	5.77	5.37	1.89	7.64	3.49	0.87	3.57	9.55	3.93	6.23
2.15	2.39	8.59	5.72	5.35	2.76	6.06	3.38	1.76	2.24	8.74	3.67	5.46
3.16	1.34	9.47	5.61	5.90	1.64	4.37	3.36	2.45	2.60	3.19	3.43	4.15
4.64	1.55	7.75	5.42	4.97	2.29	4.90	3.15	2.59	9.58	8.03	3.19	4.06
6.81	1.52	4.30	4.90	5.67	2.08	9.29	2.96	3.06	2.83	7.17	2.95	5.19
10.00	1.50	3.64	4.37	5.61	2.80	9.68	2.57	4.68	2.51	4.76	2.37	3.80



**Figure S9.** Viscosity  $\eta$  as a function of frequency of formulations without PF and AN oil at temperatures 25 and 34 °C. Each point is the mean of at least three replicates. Standard deviations have been omitted for clarity; however, in all cases, the relative standard deviation of replicate analysis was less than 10%.



**Figure S10.**  $\tan \delta$  as a function of frequency of formulations containing PF at temperatures of 25 (A) and 34 °C (B). Each point is the mean of at least three replicates. Standard deviations have been omitted for clarity; however, in all cases, the relative standard deviation of replicate analysis was less than 10%.



**Figure S11.**  $\tan \delta$  as a function of frequency of formulations containing AN at temperatures of 25 (A) and 34 °C (B). Each point is the mean of at least three replicates. Standard deviations have been omitted for clarity; however, in all cases, the relative standard deviation of replicate analysis was less than 10%.