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## Effect of ethanol on the stability of sodium caseinate stabilised emulsions

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## Effect of ethanol on the stability of sodium caseinate stabilised emulsions

Stephan W. J. Erxleben, Eddie Pelan and Bettina Wolf

**Supplementary material** 

Table S1: Source data for solubility (Figure 1), identical lowercase letters indicate no significant differences between samples for the same heating history

non	heated	heated	
Ethanol concentration (wt. %)	Solubility (%)	Ethanol concentration (wt. %)	Solubility (%)
0	89.22 ± 0.64 <sup>a</sup>	0	91.37 ± 1.33 <sup>g</sup>
5	92.06 ± 0.28 <sup>b</sup>	5	91.17 ± 0.37 <sup>g</sup>
10	83.30 ± 1.65°	10	94.08 ± 0.36 <sup>h</sup>
15	82.47 ± 0.34°	15	91.88 ± 0.25 <sup>g</sup>
20	75.47 ± 2.20 <sup>d</sup>	20	93.08 ± 0.52 <sup>g</sup>
25	60.99 ± 2.67 <sup>e</sup>	25	91.55 ± 0.72 <sup>g</sup>
30	57.07 ± 2.91 <sup>e</sup>	30	91.35 ± 0.49 <sup>g</sup>
35	54.36 ± 1.54 <sup>e</sup>	35	86.21 ± 1.03 <sup>i</sup>
40	53.78 ± 6.31 <sup>e</sup>	40	78.27 ± 0.88 <sup>j</sup>
45	48.05 ± 4.55 <sup>e</sup>	45	66.13 ± 1.58 <sup>k</sup>
50	27.89 ± 2.78 <sup>f</sup>	50	52.89 ± 0.33 <sup>m</sup>

Table S2: Source data for soluble fraction (Figure 2), identical lowercase letters indicate no significant differences between samples for the same heating history. Identical uppercase letters indicate no significant differences between samples for the same ethanol concentration

non heated		heat	ed
Ethanol concentration (wt. %)	Soluble fraction (%)	Ethanol concentration (wt. %)	Soluble fraction (%)
0	89.22 ± 0.64 <sup>a</sup>	0	91.37 ± 1.33 <sup>f</sup>
5	94.86 ± 0.35 <sup>b, A</sup>	5	95.28 ± 0.80 <sup>g, A</sup>
10	91.05 ± 1.88 <sup>a, c</sup>	10	96.06 ± 2.21 <sup>g</sup>
15	91.98 ± 1.66°	15	95.79 ± 0.14 <sup>g</sup>
20	93.98 ± 0.04 <sup>c</sup>	20	95.83 ± 0.54 <sup>g</sup>
25	90.42 ± 0.10 <sup>c</sup>	25	95.86 ± 0.66 <sup>g</sup>
30	91.88 ± 1.22 <sup>c</sup>	30	95.27 ± 0.10 <sup>g</sup>
35	92.01 ± 1.42°	35	95.25 ± 0.48 <sup>g</sup>
40	88.97 ± 0.22 <sup>a</sup>	40	93.09 ± 0.47 <sup>f</sup>
45	87.12 ± 0.43 <sup>d</sup>	45	90.89 ± 0.96 <sup>f</sup>
50	83.45 ± 0.11 <sup>e</sup>	50	88.02 ± 0.39 <sup>h</sup>

Table S3: Source data for aggregate size, part I – dispersion in ethanol-water mixtures (Figure 3), identical lowercase letters indicate no significant differences between samples for the same heating history and the same dispersion procedure. Identical uppercase letters indicate no significant differences between samples for the same ethanol concentration and the same dispersion procedure. Identical Greek letters indicate no significant changes between samples for the same heating history and the same ethanol concentration

non heated		heated	
Ethanol concentration (wt. %)	Z-average (nm)	Ethanol concentration (wt. %)	Z-average (nm)
0	434.6 ± 5.3 <sup>a, α</sup>	0	403.3 ± 6.2 h, η
5	330.4 ± 7.9 <sup>b, β</sup>	5	290.6 ± 2.9 i
10	281.0 ± 9.6 <sup>c, A, γ</sup>	10	264.9 ± 1.0 <sup>j, A, θ</sup>
15	245.1 ± 3.1 <sup>d</sup>	15	220.7 ± 4.7 <sup>k, ι</sup>
20	194.6 ± 2.0 <sup>e, Β, δ</sup>	20	191.1 ± 0.3 <sup>m, B</sup>
25	186.9 ± 1.7 <sup>f</sup>	25	180.1 ± 3.2 <sup>n</sup>
30	208.5 ± 4.2 <sup>g, ε</sup>	30	230.5 ± 3.2 <sup>k</sup>
35	208.7 ± 4.1 <sup>g, ζ</sup>	35	183.6 ± 1.8 <sup>n</sup>
40	267.8 ± 3.1 <sup>c</sup>	40	286.6 ± 3.3 <sup>i</sup>
45	285.3 ± 0.9°	45	280.5 ± 1.4 <sup>i</sup>
50	331.4 ± 7.6 <sup>b</sup>	50	520.8 ± 5.5 °

Table S4: Source data for aggregate size, part II — dispersion in water (Figure 3), identical lowercase letters indicate no significant differences between samples for the same heating history and the same dispersion procedure. Identical uppercase letters indicate no significant differences between samples for the same ethanol concentration and the same dispersion procedure. Identical Greek letters indicate no significant changes between samples for the same heating history and the same ethanol concentration

non hea	ted	heate	d
Ethanol concentration (wt. %)	Z-average (nm)	Ethanol concentration (wt. %)	Z-average (nm)
0	434.6 ± 5.3 a, α	0	403.3 ± 6.2 <sup>f, η</sup>
5	331.6 ± 6.4 b, β	5	315.2 ± 1.6 <sup>g</sup>
10	289.9 ± 1.5 <sup>c, γ</sup>	10	272.3 ± 5.1 h, θ
15	225.2 ± 1.0 <sup>d, A</sup>	15	231.4 ± 5.2 <sup>I, k, A, ι</sup>
20	198.0 ± 1.3 e, β, δ	20	201.2 ± 3.5 <sup>j, B</sup>
25	193.6 ± 1.1 <sup>f, C</sup>	25	203.4 ± 6.0 <sup>j, C</sup>
30	215.4 ± 1.0 <sup>e, ε</sup>	30	221.7 ± 2.4 <sup>i</sup>
35	204.5 ± 1.3 <sup>e, ζ</sup>	35	209.2 ± 1.5 <sup>I, j</sup>
40	207.5 ± 4.4 <sup>e</sup>	40	228.5 ± 5.4 <sup>i</sup>
45	215.6 ± 2.6 <sup>e</sup>	45	241.8 ± 3.8 <sup>k</sup>
50	203.4 ± 3.5 <sup>e</sup>	50	247.3 ± 1.9 <sup>k</sup>

Table S5: Source data for aggregate zeta potential, part I — dispersion in ethanol-water mixtures (Figure 4), identical lowercase letters indicate no significant differences between samples for the same heating history and the same dispersion procedure. Identical uppercase letters indicate no significant differences between samples for the same ethanol concentration and the same dispersion procedure. Identical Greek letters indicate no significant changes between samples for the same heating history and the same ethanol concentration

non heated		heate	ed
Ethanol concentration (wt. %)	ζ-potential (mV)	Ethanol concentration (wt. %)	ζ-potential (mV)
0	-21.9 ± 0.5 a, A, α	0	-22.0 ± 0.5 <sup>e, A, ε</sup>
5	-25.4 ± 1.0 a, B	5	-27.2 ± 0.9 e, f, B
10	-32.1 ± 3.7 b, d, C	10	-30.6 ± 1.8 <sup>f, C</sup>
15	-34.0 ± 0.2 <sup>b</sup>	15	-45.4 ± 1.0 <sup>g</sup>
20	-46.3 ± 1.9 °	20	-55.7 ± 1.8 <sup>h</sup>
25	-40.7 ± 0.5 <sup>d, D</sup>	25	-42.4 ± 2.4 <sup>g, D</sup>
30	-35.6 ± 3.9 b, d, E, β	30	-41.3 ± 0.8 <sup>g, E</sup>
35	-33.7 ± 2.0 b	35	-41.2 ± 1.5 <sup>g</sup>
40	-35.7 ± 3.1 <sup>b, d, F, γ</sup>	40	-39.3 ± 2.5 <sup>g, F</sup>
45	-32.2 ± 1.8 <sup>b, δ</sup>	45	-24.8 ± 2.6 <sup>e</sup>
50	-24.3 ± 2.3 <sup>a, G</sup>	50	-26.1 ± 1.6 <sup>e, G</sup>

Table S6: Source data for aggregate zeta potential, part II – dispersion in water (Figure 4), identical lowercase letters indicate no significant differences between samples for the same heating history and the same dispersion procedure. Identical uppercase letters indicate no significant differences between samples for the same ethanol concentration and the same dispersion procedure. Identical Greek letters indicate no significant changes between samples for the same heating history and the same ethanol concentration

non hea	ted	heate	d
Ethanol concentration (wt. %)	ζ-potential (mV)	Ethanol concentration (wt. %)	ζ-potential (mV)
0	-21.9 ± 0.5 a, A, α	0	-22.0 ± 0.5 <sup>f, A, ε</sup>
5	-57.6 ± 2.3 b	5	-49.5 ± 1.7 <sup>g</sup>
10	-66.7 ± 2.4 <sup>c, B</sup>	10	-69.4 ± 1.1 h, B
15	-67.9 ± 4.9 <sup>c</sup>	15	-82.8 ± 1.2 i
20	-67.1 ± 1.9 °	20	-80.9 ± 4.4 <sup>i</sup>
25	-63.2 ± 4.7 b, c, C	25	-68.9 ± 1.0 h, C
30	-40.3 ± 1.4 <sup>d, β</sup>	30	-54.4 ± 4.0 <sup>g, j</sup>
35	-42.1 ± 2.7 <sup>d</sup>	35	-65.9 ± 3.6 h, j
40	-37.3 ± 4.3 <sup>d, e, γ</sup>	40	-59.4 ± 3.2 a, j
45	-28.5 ± 2.6 <sup>e, δ</sup>	45	-47.9 ± 3.3 <sup>g</sup>
50	-31.6 ± 2.3 <sup>e</sup>	50	-45.4 ± 2.0 <sup>g</sup>

Table S7: Source data for steady state surface tension (Figure 5), identical lowercase letters indicate no significant differences between samples for the same initial protein concentration

	with sodium caseinate	without sodium caseinate
Ethanol	Surface tension (mN/m)	Surface tension (mN/m)
concentration (wt. %)	Surface tension (milym)	Surface tension (mity m)
0	41.46 ± 0.32 a	72.28 ± 0.03 <sup>e</sup>
15	31.10 ± 0.66 b	46.55 ± 0.05 <sup>f</sup>
25	27.84 ± 0.49 °	44.37 ± 0.05 <sup>g</sup>
40	25.45 ± 0.87 <sup>d</sup>	36.65 ± 0.03 h
50	25.17 ± 1.09 <sup>d</sup>	35.21 ± 0.05 <sup>i</sup>

Table S8: Surface tension data (steady state) for various sodium caseinate containing ethanol-water mixtures, acquired at 20 °C, identical lowercase letters indicate no significant differences between samples for the same ethanol concentration

	dispersion in ethanol-water mixture		dispersion	ı in water
Ethanol concentration (wt. %)	Surface tension (mN/m)		Surface tens	sion (mN/m)
	non heated	heated	non heated	heated
0	41.23 ± 0.23 <sup>a</sup>	41.68 ± 0.19 °	-	-
15	30.60 ± 0.53 b	31.68 ± 0.44 b	30.46 ± 0.63 b	31.66 ± 0.42 b
25	27.56 ± 0.28 °	27.83 ± 0.35 °	27.44 ± 0.42 °	28.54 ± 0.55 °
40	26.43 ± 0.31 <sup>d</sup>	25.53 ± 0.17 <sup>e</sup>	25.53 ± 0.43 <sup>e</sup>	24.30 ± 0.36 <sup>f</sup>
50	25.41 ± 0.21 <sup>g</sup>	25.96 ± 0.25 <sup>g</sup>	23.57 ± 0.23 <sup>h</sup>	25.74 ± 0.15 <sup>g</sup>

Table S9: Density difference data (oil phase density – aqueous phase density), acquired at 20 °C, identical lowercase letters indicate no significant differences between samples for the same protein concentration

	with sodium caseinate	without sodium caseinate
Ethanol	Density difference (g/cm³)	Density difference (g/cm³)
concentration (wt. %)	,,	, G
0	-0.090 ± 0.001 a	-0.082 ± 0.001 <sup>f</sup>
15	-0.066 ± 0.000 b	-0.059 ± 0.001 <sup>g</sup>
25	-0.053 ± 0.001 °	-0.044 ± 0.000 h
40	-0.026 ± 0.002 <sup>d</sup>	-0.014 ± 0.001 i
50	-0.003 ± 0.001 <sup>e</sup>	0.004 ± 0.001 <sup>j</sup>

Table S10: Source data for emulsion droplet size, ethanol addition pre-processing, part I (0 wt.% ethanol) (Figure 8), identical lowercase letters indicate no significant differences between samples for the same ethanol concentration

Time (d)	Z-average (nm)	PDI <sub>DLS</sub> (-)
0	471.1 ± 13.7 a, b	0.24 ± 0.02 <sup>e</sup>
2	480.5 ± 6.5 <sup>a</sup>	0.25 ± 0.03 <sup>e</sup>
4	505.7 ± 8.5 <sup>c</sup>	0.24 ± 0.02 <sup>e</sup>
9	515.1 ± 10.2 °	0.25 ± 0.05 <sup>e</sup>
11	492.3 ± 7.8 °	0.24 ± 0.04 <sup>e</sup>
18	462.6 ± 5.2 b	0.23 ± 0.03 <sup>e</sup>
26	450.2 ± 5.2 b	0.24 ± 0.02 <sup>e</sup>
31	427.6 ± 5.4 <sup>d</sup>	0.23 ± 0.02 <sup>e</sup>

Table S11: Source data for emulsion droplet size, ethanol addition pre-processing, part II (15 wt.% ethanol) (Figure 8), identical lowercase letters indicate no significant differences between samples for the same ethanol concentration

Time (d)	Z-average (nm)	PDI <sub>DLS</sub> (-)
0	336.8 ± 3.8 °	0.26 ± 0.01 d
2	342.7 ± 3.1 °	0.29 ± 0.01 <sup>e</sup>
4	364.2 ± 2.4 <sup>b</sup>	0.27 ± 0.01 <sup>e</sup>
9	361.5 ± 21.7 a, b	0.23 ± 0.02 <sup>f</sup>
11	365.3 ± 3.4 <sup>b</sup>	0.24 ± 0.04 <sup>e, f</sup>
18	340.4 ± 4.5 °	0.25 ± 0.04 <sup>e, f</sup>
26	408.9 ± 7.3 °	0.24 ± 0.03 <sup>e, f</sup>
31	415.2 ± 0.6 °	0.27 ± 0.05 <sup>e, f</sup>

Table S12: Source data for emulsion droplet size, ethanol addition pre-processing, part III (25 wt.% ethanol) (Figure 8), identical lowercase letters indicate no significant differences between samples for the same ethanol concentration

Time (d)	Z-average (nm)	PDI <sub>DLS</sub> (-)
0	250.6 ± 3.4 °	0.24 ± 0.02 °
2	253.3 ± 1.6 ª	0.25 ± 0.03 °
4	263.7 ± 4.0 b	0.26 ± 0.05 °
9	278.9 ± 12.9 a, b	0.22 ± 0.04 <sup>c</sup>
11	265.3 ± 1.3 b	0.24 ± 0.03 °
18	257.0 ± 1.8 ª	0.22 ± 0.02 °
26	289.7 ± 16.4 a, b	0.23 ± 0.02 °
31	267.1 ± 5.1 b	0.24 ± 0.02 °

Table S13: Source data for emulsion droplet size, ethanol addition pre-processing, part IV (40 wt.% ethanol) (Figure 8), identical lowercase letters indicate no significant differences between samples for the same ethanol concentration

Time (d)	D[4,3] (μm)	PDI <sub>SLS</sub> (-)
0	0.84 ± 0.04 a	0.59 ± 0.02 °
2	6.03 ± 0.13 b	0.42 ± 0.01 <sup>d</sup>
4	5.83 ± 0.03 °	0.42 ± 0.01 <sup>d</sup>
9	6.18 ± 0.12 b	0.41 ± 0.01 <sup>d</sup>
11	5.99 ± 0.02 b	0.42 ± 0.01 <sup>d</sup>
18	6.06 ± 0.08 b	0.41 ± 0.01 <sup>d</sup>
26	6.00 ± 0.07 b	0.41 ± 0.00 d
31	5.96 ± 0.04 b	0.42 ± 0.00 <sup>d</sup>

Table S14: Source data for emulsion droplet size, ethanol addition pre-processing, part V (50 wt.% ethanol) (Figure 8), identical lowercase letters indicate no significant differences between samples for the same ethanol concentration

Time (d)	D[4,3] (μm)	PDI <sub>SLS</sub> (-)
0	1.94 ± 0.07 °	1.71 ± 0.11 <sup>f</sup>
2	36.48 ± 0.01 b	0.17 ± 0.01 <sup>g</sup>
4	34.28 ± 0.31 °	0.20 ± 0.01 g
9	38.50 ± 0.43 <sup>d</sup>	0.20 ± 0.02 <sup>g</sup>
11	40.78 ± 0.60 e	0.17 ± 0.01 <sup>g</sup>
18	40.40 ± 0.31 e	0.17 ± 0.01 <sup>g</sup>
26	41.51 ± 0.59 <sup>e</sup>	0.17 ± 0.01 <sup>g</sup>
31	40.32 ± 1.26 <sup>e</sup>	0.18 ± 0.00 <sup>g</sup>

Table S15: Droplet size versus oil concentration immediately after homogenisation and after addition of water or ethanol (Table 1). The asterisk indicates the use of  $d_{4,3}$  which was acquired by static light scattering and the use of the corresponding  $PDI_{SLS}$  value

before solvent addition			after solvent addition				
			Water Et		Eth	anol	
Oil concentration	Droplet size	PDI (-)	Oil concentration	Droplet size	PDI (-)	Droplet size	PDI (-)
(wt.%)	(nm)	PDI (-)	(wt.%)	(nm)	PDI (-)	(nm)	PDI (-)
11.8	601 ± 19	0.24 ± 0.05	10	492 ± 14	0.20 ± 0.03	353 ± 3	0.18 ± 0.05
13.3	712 ± 33	0.24 ± 0.03	10	549 ± 13	0.20 ± 0.03	369 ± 4	0.18 ± 0.02
16.7	1468 ± 37	0.29 ± 0.02	10	476 ± 16	0.24 ± 0.05	652 ± 12	0.21 ± 0.02
20	3429 ± 53*	4.04 ± 0.01*	10	456 ± 10	0.19 ± 0.03	2766 ± 97*	1.09 ± 0.14*

Table S16: Source data for emulsion droplet size, ethanol addition post-processing, part I (0 wt.% ethanol) (Figure 9), identical lowercase letters indicate no significant differences between samples for the same ethanol concentration

Time (d)	Z-average (nm)	PDI <sub>DLS</sub> (-)
0	471.1 ± 13.7 a, b	0.24 ± 0.02 <sup>e</sup>
2	480.5 ± 6.5 °	0.25 ± 0.03 <sup>e</sup>
4	505.7 ± 8.5 °	0.24 ± 0.02 <sup>e</sup>
9	515.1 ± 10.2 °	0.25 ± 0.05 <sup>e</sup>
16	462.6 ± 2.3 b	0.23 ± 0.03 <sup>e</sup>
26	450.2 ± 5.2 b	0.24 ± 0.02 <sup>e</sup>
31	427.6 ± 5.4 <sup>d</sup>	0.23 ± 0.02 <sup>e</sup>

Table S17: Source data for emulsion droplet size, ethanol addition post-processing, part II (15 wt.% ethanol) (Figure 9), identical lowercase letters indicate no significant differences between samples for the same ethanol concentration

Time (d)	Z-average (nm)	PDI <sub>DLS</sub> (-)
0	352.5 ± 3.4 a, b	0.18 ± 0.05 a
2	370.9 ± 8.6 °	0.20 ± 0.03 <sup>a</sup>
4	368.2 ± 5.4 °	0.19 ± 0.03 a
9	370.5 ± 2.5 °	0.18 ± 0.05 a
16	338.3 ± 2.2 <sup>d</sup>	0.17 ± 0.03 <sup>a</sup>
26	372.6 ± 6.5 °	0.18 ± 0.04 <sup>a</sup>
31	365.8 ± 11.6 <sup>b, c</sup>	0.20 ± 0.02 a

Table S18: Source data for emulsion droplet size, ethanol addition post-processing, part III (25 wt.% ethanol) (Figure 9), identical lowercase letters indicate no significant differences between samples for the same ethanol concentration

Time (d)	Z-average (nm)	PDI <sub>DLS</sub> (-)
0	369.4 ± 3.7 °	0.18 ± 0.02 °
2	381.8 ± 2.1 b	0.17 ± 0.05 <sup>c</sup>
4	372.8 ± 5.5 a, b	0.19 ± 0.03 °
9	387.5 ± 4.6 <sup>b</sup>	0.19 ± 0.04 °
16	372.1 ± 9.8 a, b	0.19 ± 0.04 °
26	385.7 ± 1.6 b	0.22 ± 0.03 °
31	379.5 ± 3.9 a, b	0.23 ± 0.06 °

Table S19: Source data for emulsion droplet size, ethanol addition post-processing, part IV (40 wt.% ethanol) (Figure 9), identical lowercase letters indicate no significant differences between samples for the same ethanol concentration

Time (d)	Z-average (nm)	PDI <sub>DLS</sub> (-)	
0	651.8 ± 11.6 °	0.21 ± 0.02 <sup>f</sup>	
2	774.3 ± 17.0 b	0.23 ± 0.05 <sup>f</sup>	
4	827.7 ± 21.1 °	0.22 ± 0.05 <sup>f</sup>	
9	901.8 ± 15.9 <sup>d</sup>	0.26 ± 0.02 <sup>f</sup>	
16	1004.3 ± 20.6 <sup>e</sup>	0.20 ± 0.04 <sup>f</sup>	
26	1047.0 ± 75.4 <sup>d, e</sup>	0.26 ± 0.04 <sup>f</sup>	
31	1082.5 ± 91.2 <sup>d, e</sup>	0.24 ± 0.03 <sup>f</sup>	

Table S20: Source data for emulsion droplet size, ethanol addition post-processing, part V (50 wt.% ethanol) (Figure 9), identical lowercase letters indicate no significant differences between samples for the same ethanol concentration

Time (d)	D[4,3] (μm)	PDI <sub>SLS</sub> (-)
0	2.77 ± 0.10 a	1.09 ± 0.14 <sup>e</sup>
2	2.93 ± 0.00 b	0.44 ± 0.01 <sup>f</sup>
4	2.97 ± 0.01 <sup>a</sup>	0.43 ± 0.01 <sup>f</sup>
9	2.95 ± 0.01 a	0.46 ± 0.04 <sup>f</sup>
16	3.01 ± 0.01 °	0.45 ± 0.01 <sup>f</sup>
26	3.09 ± 0.00 <sup>d</sup>	0.31 ± 0.02 <sup>g</sup>
31	3.08 ± 0.02 <sup>d</sup>	0.31 ± 0.00 <sup>g</sup>

Table S21: Source data for emulsion zeta potential size, ethanol addition pre-processing, part I (Figure 10), identical lowercase letters indicate no significant differences between samples for the same ethanol concentration

0% Ethanol		15% Ethanol		25% Ethanol	
Time (d)	ζ-potential (mV)	Time (d)	ζ-potential (mV)	Time (d)	ζ-potential (mV)
0	-49.6 ± 2.0 ª	0	-60.7 ± 2.6 °	0	-61.4 ± 1.9 <sup>e</sup>
2	-56.0 ± 0.4 <sup>b</sup>	2	-72.6 ± 1.8 <sup>d</sup>	2	-64.5 ± 2.5 <sup>e</sup>
4	-49.0 ± 1.4 a	4	-66.5 ± 3.8 °	4	-57.1 ± 6.4 <sup>e</sup>
9	-50.3 ± 2.3 a	9	-65.6 ± 3.5 °	9	-59.2 ± 1.0 <sup>e</sup>
11	-54.5 ± 2.8 <sup>a</sup>	11	-74.5 ± 3.4 <sup>d</sup>	11	-69.8 ± 4.9 <sup>e</sup>
18	-51.9 ± 1.5 ª	18	-77.0 ± 3.2 <sup>d</sup>	18	-58.6 ± 0.8 <sup>e</sup>
26	-49.9 ± 1.6 ª	26	-63.9 ± 2.3 °	26	-65.3 ± 2.4 <sup>e</sup>
31	-51.9 ± 1.8 ª	31	-67.3 ± 2.1 °	31	-47.2 ± 4.4 <sup>f</sup>

Table S22: Source data for emulsion zeta potential size, ethanol addition pre-processing, part II (Figure 10), identical lowercase letters indicate no significant differences between samples for the same ethanol concentration

40% Ethanol		50% Ethanol	
Time (d)	ζ-potential (mV)	Time (d)	ζ-potential (mV)
0	-38.8 ± 0.9 °	0	-15.2 ± 0.9 <sup>d</sup>
2	-54.4 ± 1.7 <sup>b</sup>	2	-33.5 ± 0.9 <sup>e</sup>
4	-48.7 ± 1.2 <sup>a, c</sup>	4	-32.2 ± 1.3 <sup>e</sup>
9	-53.6 ± 3.4 <sup>b, c</sup>	9	-34.0 ± 1.1 <sup>e</sup>
11	-46.2 ± 3.1 <sup>a, c</sup>	11	-39.9 ± 2.4 °
18	-53.7 ± 1.6 <sup>b</sup>	18	-30.7 ± 2.9 °
26	-53.7 ± 0.1 <sup>b</sup>	26	-35.9 ± 2.5 °
31	-46.2 ± 2.0 °	31	-32.1 ± 3.6 °

Table S23: Source data for emulsion zeta potential size, ethanol addition post-processing, part I (Figure 11), identical lowercase letters indicate no significant differences between samples for the same ethanol concentration

0% Ethanol		15% Ethanol		25% Ethanol	
Time (d)	ζ-potential (mV)	Time (d)	ζ-potential (mV)	Time (d)	ζ-potential (mV)
0	-49.6 ± 2.0 ª	0	-65.6 ± 1.7 °	0	-58.4 ± 0.8 <sup>d</sup>
2	-56.0 ± 0.4 <sup>b</sup>	2	-74.0 ± 4.6 <sup>c</sup>	2	-55.9 ± 0.5 <sup>e</sup>
4	-49.0 ± 1.4 ª	4	-70.8 ± 3.4 °	4	-56.5 ± 3.2 <sup>d, e, f</sup>
9	-50.3 ± 2.3 a	9	-67.8 ± 0.9 °	9	-57.8 ± 5.6 <sup>d, e, f</sup>
16	-49.5 ± 1.2 ª	16	-63.4 ± 2.5 °	16	-61.2 ± 1.0 <sup>a, f</sup>
26	-49.9 ± 1.6 ª	26	-64.7 ± 3.4 °	26	-69.5 ± 4.1 a, f
31	-51.9 ± 1.8 ª	31	-68.7 ± 3.0 °	31	-56.2 ± 2.2 <sup>d, e, f</sup>

Table S24: Source data for emulsion zeta potential size, ethanol addition post-processing, part II (Figure 11), identical lowercase letters indicate no significant differences between samples for the same ethanol concentration

40% Ethanol		50% Ethanol	
Time (d)	ζ-potential (mV)	Time (d)	ζ-potential (mV)
0	-40.9 ± 2.2 °	0	-29.3 ± 3.2 °
2	-43.9 ± 1.1 °	2	-33.6 ± 4.3 <sup>c, d</sup>
4	-40.2 ± 0.6 a	4	-33.2 ± 6.1 <sup>c, d</sup>
9	-34.6 ± 0.2 <sup>b</sup>	9	-29.7 ± 2.4 °
16	-40.9 ± 1.5 ª	16	-30.4 ± 4.2 <sup>c, d</sup>
26	-35.5 ± 2.1 <sup>b</sup>	26	-38.0 ± 1.3 <sup>d</sup>
31	-42.0 ± 1.9 °	31	-28.2 ± 1.7 °

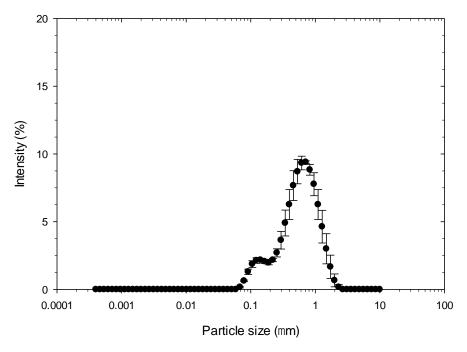


Figure S1: Intensity-based size distribution for sodium caseinate containing aqueous solution (heated to 60 °C, protein), data were acquired at 20 °C

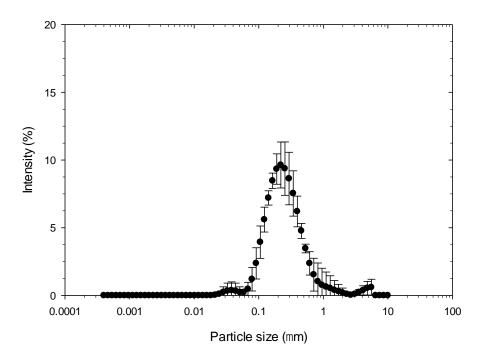


Figure S2: Intensity-based size distribution for sodium caseinate containing water-ethanol (25 wt.%) mixture (non heated, protein dispersion in water), data were acquired at 20 °C

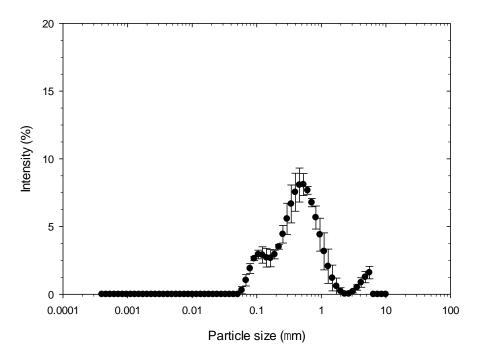


Figure S3: Intensity-based size distribution for sodium caseinate containing water-ethanol (50 wt.%) mixture (non heated, protein dispersion in water-ethanol mixture), data were acquired at 20  $^{\circ}$ C

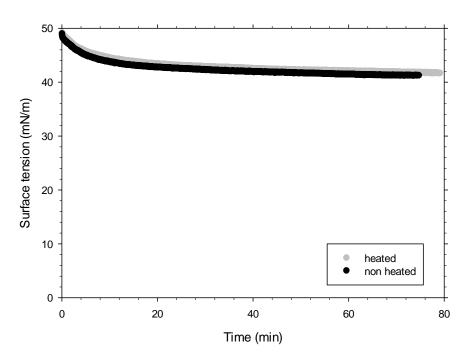


Figure S4: Dynamic surface tension of aqueous sodium caseinate systems containing 0% Ethanol, data were acquired at 20 °C

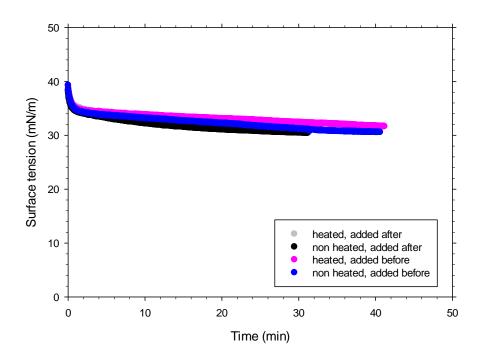


Figure S5: Dynamic surface tension of aqueous sodium caseinate systems containing 15% Ethanol, data were acquired at  $20\,^{\circ}\text{C}$ 

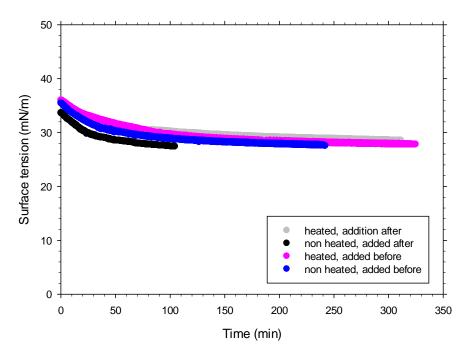


Figure S6: Dynamic surface tension of aqueous sodium caseinate systems containing 25% Ethanol, data were acquired at  $20\,^{\circ}\text{C}$ 

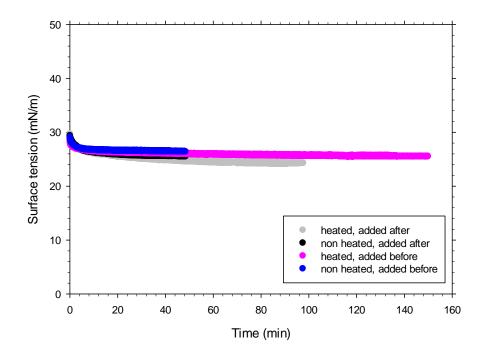


Figure S7: Dynamic surface tension of aqueous sodium caseinate systems containing 40% Ethanol, data were acquired at  $20\,^{\circ}\text{C}$ 

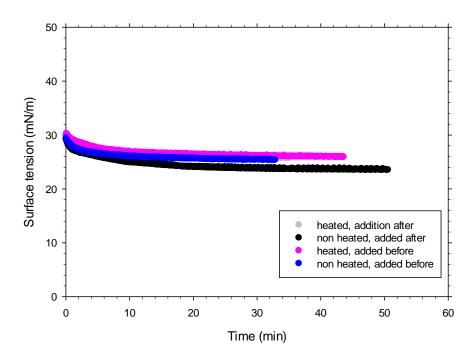


Figure S8: Dynamic surface tension of aqueous sodium caseinate systems containing 50% Ethanol, data were acquired at  $20\,^{\circ}\text{C}$ 

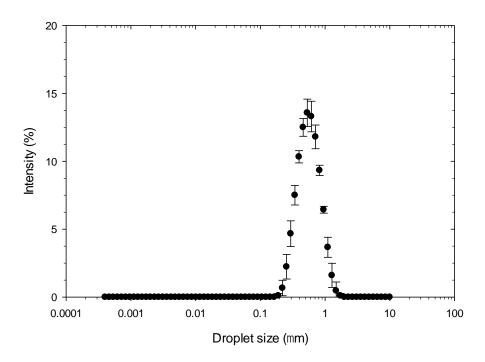


Figure S9: Intensity-based size distribution for oil-in-water emulsions (0 wt.% ethanol), Day 0, data were acquired at 20 °C

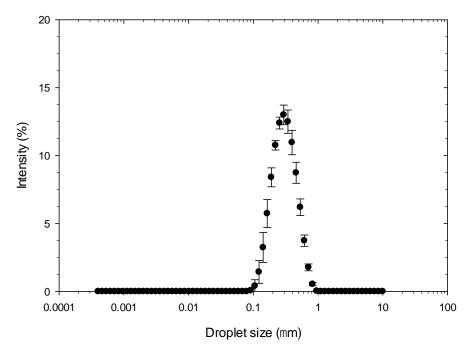


Figure S10: Intensity-based size distribution for oil-in-water emulsions (25 wt.% ethanol, ethanol addition pre-processing), Day 9, data were acquired at 20 °C

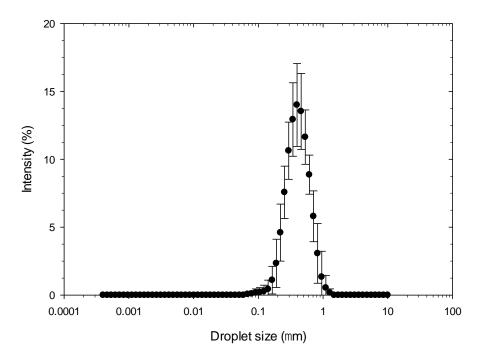


Figure S11: Intensity-based size distribution for oil-in-water emulsions (15 wt.% ethanol, ethanol addition post-processing), Day 31, data were acquired at 20  $^{\circ}$ C