

Supplemental Information:

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I. Chemicals and their Properties

Table S1. Ternary System Chemicals and Properties

Chemical	Abbv.	O:C	X:C	Solubility (in H ₂ O)	Molecular Weight	Density
Ammonium Sulfate	AS	0	0	74.4 g/100 g ^a	132.14 g/mol ^c	1.78 g/cm ³ ^d
2-methylglutaric acid	2-MGA	0.67	0.67	40.6 g/L ^b	146.14 g/mol ^b	1.33 g/cm ³ ^e
Proline	Pro	0.4	0.6	365 g/L ^b	115.13 g/mol ^c	1.40 g/cm ³ ^e
Valine	Val	0.4	0.6	88.5 g/L ^b	117.14 g/mol ^c	1.23 g/cm ³ ^d
Leucine	Leu	0.33	0.5	22.4 g/L ^b	131.18 g/mol ^c	1.29 g/cm ³ ^d

^aCRC Handbook

^bhmdb.ca

^cNIST WebBook

^d PubChem, National Institute of Health

^eEMD Millipore

II. O:C and X:C ratios of Ternary Mixtures

Proline System

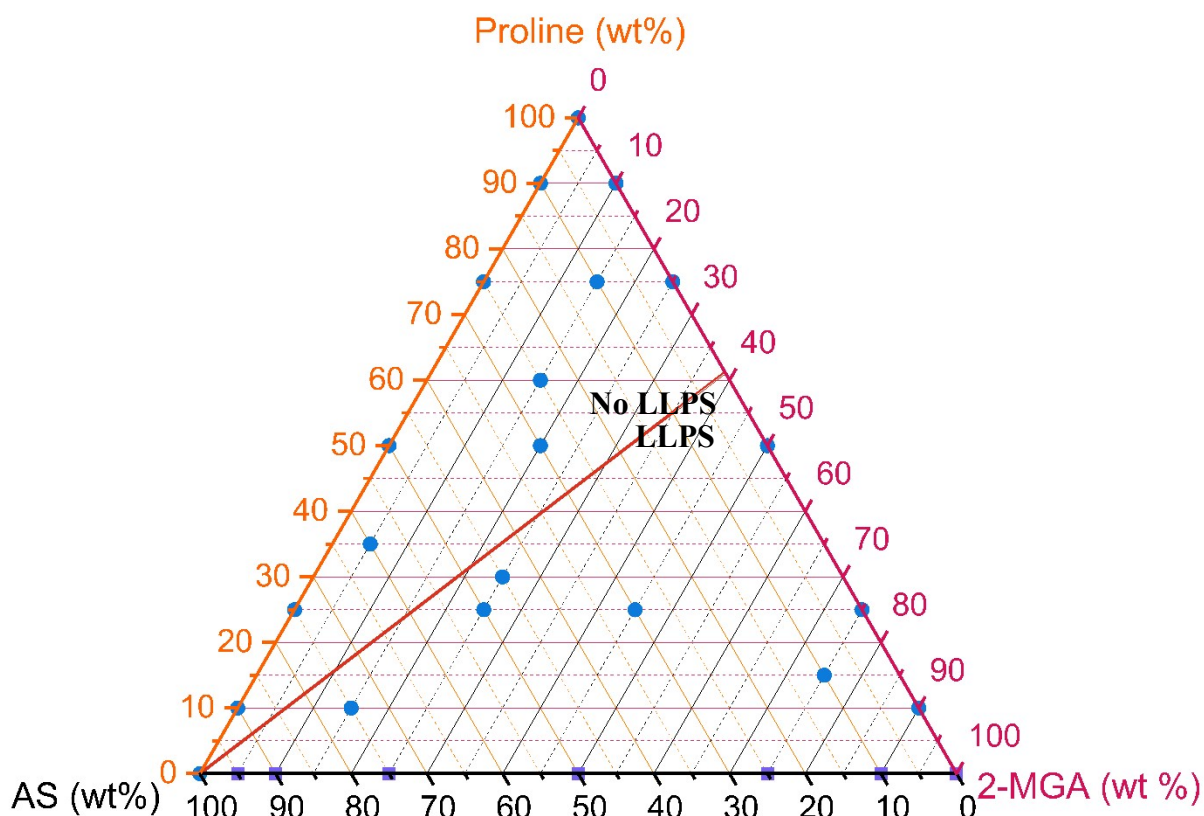
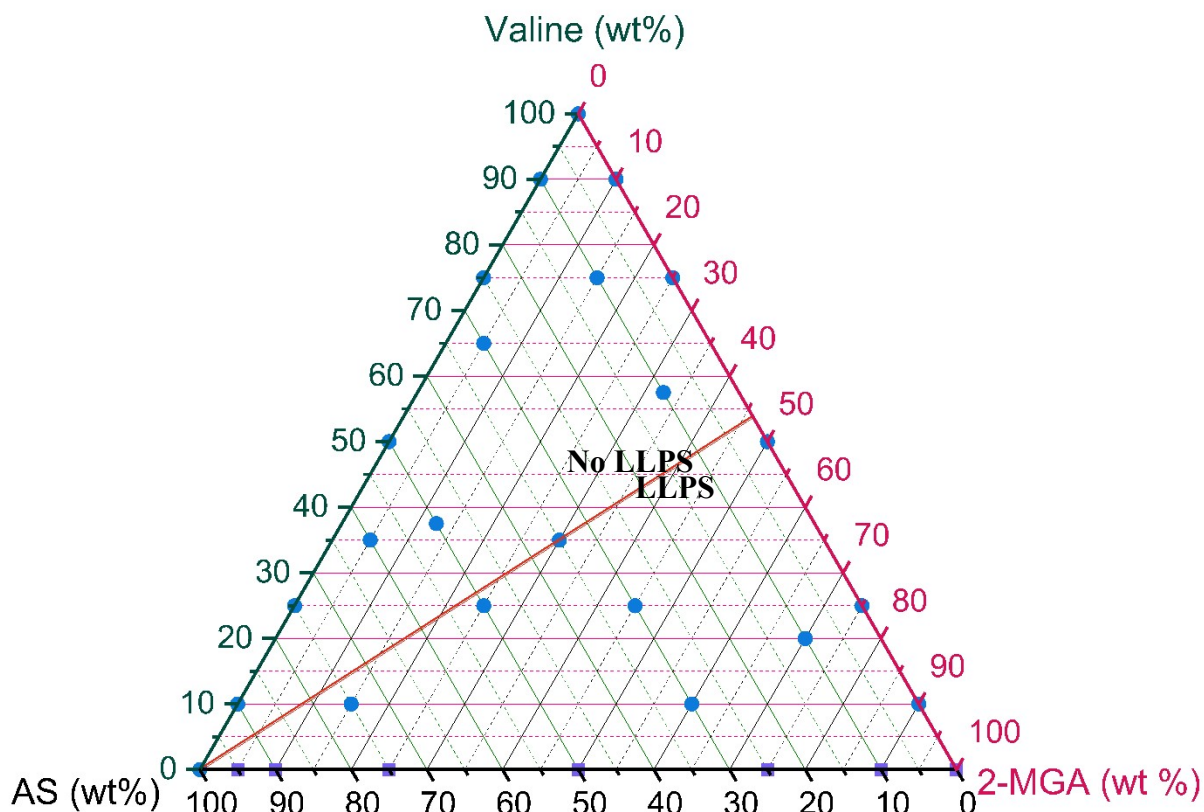


Figure S1. Weight percent ternary plots for Pro system. Each blue circle represents the solute chemical composition of each experiment. Purple squares are data points obtained from Ferdousi-Rokib et al (in prep). The red line represents the phase separation threshold, where the region below this line are expected to be phase separated while above the threshold are expected to be well mixed. The estimated threshold (red-line) is derived from the LLPS model in Malek et al.¹

Table S2. Experimental Mixture Composition and O:C/X:C ratios for Pro System.

Experiment	AS (wt%)	Proline (wt%)	2-MGA (wt%)	O:C	X:C
1	100	0	0	0.00	0.00
2	0	100	0	0.40	0.60
3 [#]	0	0	100	0.67	0.67
4	0	10	90	0.64	0.66
5	0	25	75	0.59	0.65
6	0	50	50	0.52	0.63
7	0	75	25	0.46	0.61
8	0	90	10	0.42	0.61
9	10	90	0	0.40	0.60
10	25	75	0	0.40	0.60
11	50	50	0	0.40	0.60
12	75	25	0	0.40	0.60
13	90	10	0	0.40	0.60
14 [#]	95	0	5	0.67	0.67
15 [#]	90	0	10	0.67	0.67
16 [#]	75	0	25	0.67	0.67
17 [#]	50	0	50	0.67	0.67
18 [#]	25	0	75	0.67	0.67
19 [#]	10	0	90	0.67	0.67
20	45	30	25	0.51	0.63
21	75	10	15	0.55	0.64
22	25	60	15	0.44	0.61
23	60	35	5	0.43	0.61
24	50	25	25	0.52	0.63
25	30	25	45	0.56	0.64
26	30	50	20	0.46	0.62
27	10	75	15	0.44	0.61
28	10	15	75	0.62	0.66

[#]Data points from Ferdousi-Rokib et al (in review)



Valine System

Figure S2. Weight percent ternary plots for Val system. Each blue circle represents the chemical composition of each experiment. Purple squares are data points obtained from Ferdousi-Rokib et al (in prep). The red line represents the estimated phase separation threshold, where the region below this line are expected to be phase separated while above the threshold are expected to be well mixed. The estimated threshold (red-line) is derived from the LLPS model in Malek et al.¹

Table S3. Experimental Mixture Composition and O:C/X:C ratios for Val System

Experiment	AS (wt%)	Valine (wt%)	2-MGA (wt%)	O:C	X:C
1	100	0	0	0.00	0.00
2	0	100	0	0.40	0.60
3 [#]	0	0	100	0.67	0.67
4	0	10	90	0.64	0.66
5	0	25	75	0.59	0.65
6	0	50	50	0.52	0.63
7	0	75	25	0.46	0.61
8	0	90	10	0.42	0.61
9	10	90	0	0.40	0.60
10	25	75	0	0.40	0.60
11	50	50	0	0.40	0.60
12	75	25	0	0.40	0.60
13	90	10	0	0.40	0.60
14 [#]	95	0	5	0.67	0.67
15 [#]	90	0	10	0.67	0.67
16 [#]	75	0	25	0.67	0.67
17 [#]	50	0	50	0.67	0.67
18 [#]	25	0	75	0.67	0.67
19 [#]	10	0	90	0.67	0.67
20	10	57.5	32.5	0.48	0.62
21	50	37.5	12.5	0.49	0.62
22	10	20	70	0.54	0.64
23	75	10	15	0.43	0.61
24	30	65	5	0.52	0.63
25	60	35	5	0.56	0.64
26	50	25	25	0.47	0.62
27	35	35	30	0.62	0.66
28	30	25	45	0.44	0.61
29	30	10	60	0.00	0.00
30	10	75	15	0.40	0.60

[#]Data points from Ferdousi-Rokib et al (in review)

Leucine System

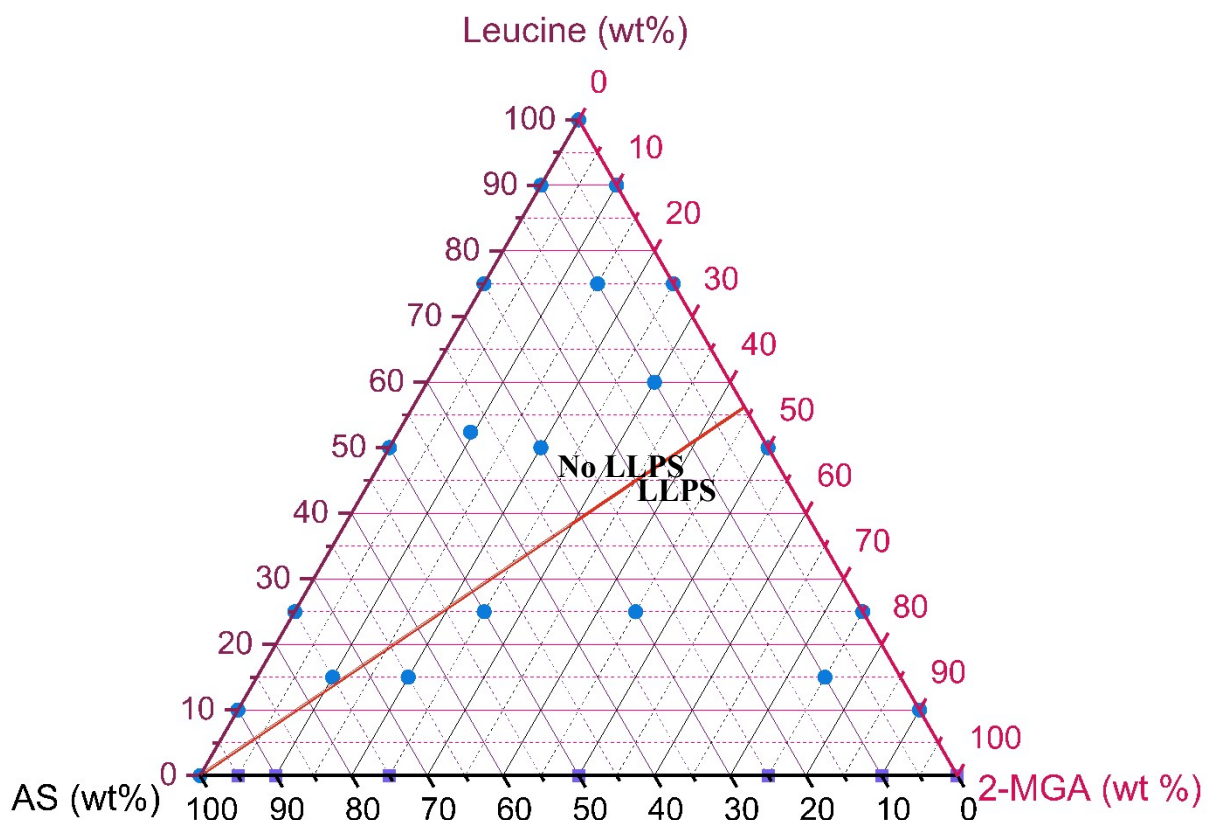


Figure S3. Weight percent ternary plots for Leu system. Each blue circle represents the chemical composition of each experiment. Purple squares are data points obtained from Ferdousi-Rokib et al (in prep). The red line represents the estimated phase separation threshold, where the region below this line are expected to be phase separated while above the threshold are expected to be well mixed. The estimated threshold (red-line) is derived from the LLPS model in Malek et al.¹

Table S4. Experimental Mixture Composition and O:C/X:C ratios for Leu System

Experiment	AS (wt%)	Leucine (wt%)	2-MGA (wt%)	O:C	X:C
1	100	0	0	0.00	0.00
2	0	100	0	0.33	0.60
3 [#]	0	0	100	0.67	0.67
4	0	10	90	0.63	0.66
5	0	25	75	0.57	0.65
6	0	50	50	0.48	0.63
7	0	75	25	0.40	0.61
8	0	90	10	0.36	0.61
9	10	90	0	0.33	0.60
10	25	75	0	0.33	0.60
11	50	50	0	0.33	0.60
12	75	25	0	0.33	0.60
13	90	10	0	0.33	0.60
14 [#]	95	0	5	0.67	0.67
15 [#]	90	0	10	0.67	0.67
16 [#]	75	0	25	0.67	0.67
17 [#]	50	0	50	0.67	0.67
18 [#]	25	0	75	0.67	0.67
19 [#]	10	0	90	0.67	0.67
20	10	60	30	0.43	0.62
21	75	15	10	0.45	0.62
22	65	15	20	0.50	0.64
23	40	55	10	0.37	0.61
24	50	25	25	0.48	0.63
25	30	25	45	0.53	0.64
26	30	50	20	0.41	0.62
27	10	15	75	0.60	0.66
28	10	75	15	0.38	0.61

[#]Data points from Ferdousi-Rokib et al (in review)

III. CCNC Experiments

Table S5. Weight of Chemical Compounds in 200mL Ultra purified Millipore Water for Pro System

Experiment	AS (mg)	Proline (mg)	2-MGA (mg)
1	20	0	0
2	0	20	0
3 [#]	0	0	20
4	0	2	18
5	0	5	15
6	0	10	10
7	0	15	5
8	0	18	2
9	2	18	0
10	5	15	0
11	10	10	0
12	15	5	0
13	18	2	0
14 [#]	19	0	1
15 [#]	18	0	2
16 [#]	15	0	5
17 [#]	10	0	10
18 [#]	5	0	15
19 [#]	2	0	18
20	9	6	5
21	15	2	3
22	5	12	3
23	12	7	1
24	10	5	5
25	6	5	9
26	6	10	4
27	2	15	3
28	2	3	15

[#]Data points from Ferdousi-Rokib et al (in review)

Table S6. Weight of Chemical Compounds in 200mL Ultra purified Millipore Water for Val System

Experiment	AS (mg)	Valine (mg)	2-MGA (mg)
1	20	0	0
2	0	20	0
3 [#]	0	0	20
4	0	2	18
5	0	5	15
6	0	10	10
7	0	15	5
8	0	18	2
9	2	18	0
10	5	15	0
11	10	10	0
12	15	5	0
13	18	2	0
14 [#]	19	0	1
15 [#]	18	0	2
16 [#]	15	0	5
17 [#]	10	0	10
18 [#]	5	0	15
19 [#]	2	0	18
20	2	12	6
21	15	3	2
22	13	3	4
23	8	11	2
24	10	5	5
25	6	5	9
26	6	10	4
27	2	3	15
28	2	15	3
29	20	0	0
30	0	20	0

[#]Data points from Ferdousi-Rokib et al (in review)

Table S7. Weight of Chemical Compounds in 200mL Ultra purified Millipore Water for Leu System

Experiment	AS (mg)	Leucine (mg)	2-MGA (mg)
1	20	0	0
2	0	20	0
3 [#]	0	0	20
4	0	2	18
5	0	5	15
6	0	10	10
7	0	15	5
8	0	18	2
9	2	18	0
10	5	15	0
11	10	10	0
12	15	5	0
13	18	2	0
14 [#]	19	0	1
15 [#]	18	0	2
16 [#]	15	0	5
17 [#]	10	0	10
18 [#]	5	0	15
19 [#]	2	0	18
20	2	12	6
21	15	3	2
22	13	3	4
23	8	11	2
24	10	5	5
25	6	5	9
26	6	10	4
27	2	3	15
28	2	15	3

[#]Data points from Ferdousi-Rokib et al (in review)

IV. CCNC Measurement Setup

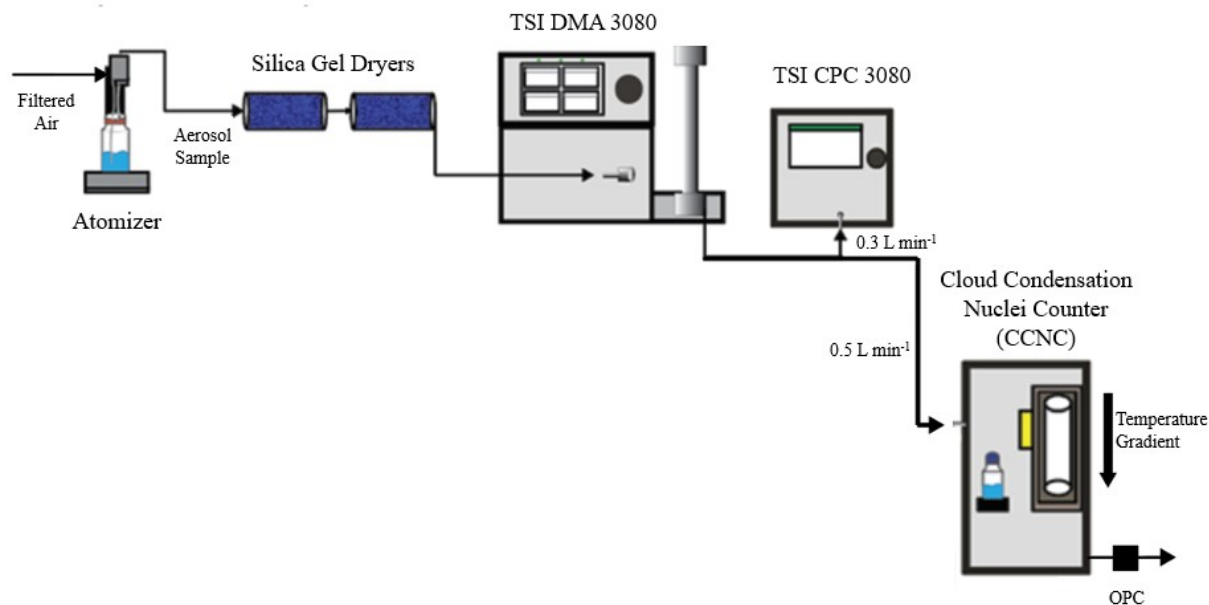


Figure S4. Experimental set up for Cloud Condensation Nuclei (CCN) experiments; dry, polydisperse aerosols were passed through the SMPS at a 1:10 aerosol to sheath flow rate; aerosols were flowed into the CPC and CCN at 0.3 L min^{-1} and 0.5 L min^{-1} , respectively.

V. Ammonium Sulfate CCN Calibration

Table S8. Ammonium Sulfate Calibration Data for CCNC

Calibrated Supersaturation (%)	Activation Diameter (nm)
0.42 ± 0.07	50.87 ± 4.47
0.61 ± 0.09	39.84 ± 3.17
0.78 ± 0.03	33.43 ± 0.86
0.99 ± 0.08	28.71 ± 1.45
1.21 ± 0.03	25.12 ± 0.42
1.57 ± 0.16	21.19 ± 1.35
1.72 ± 0.03	19.86 ± 0.21

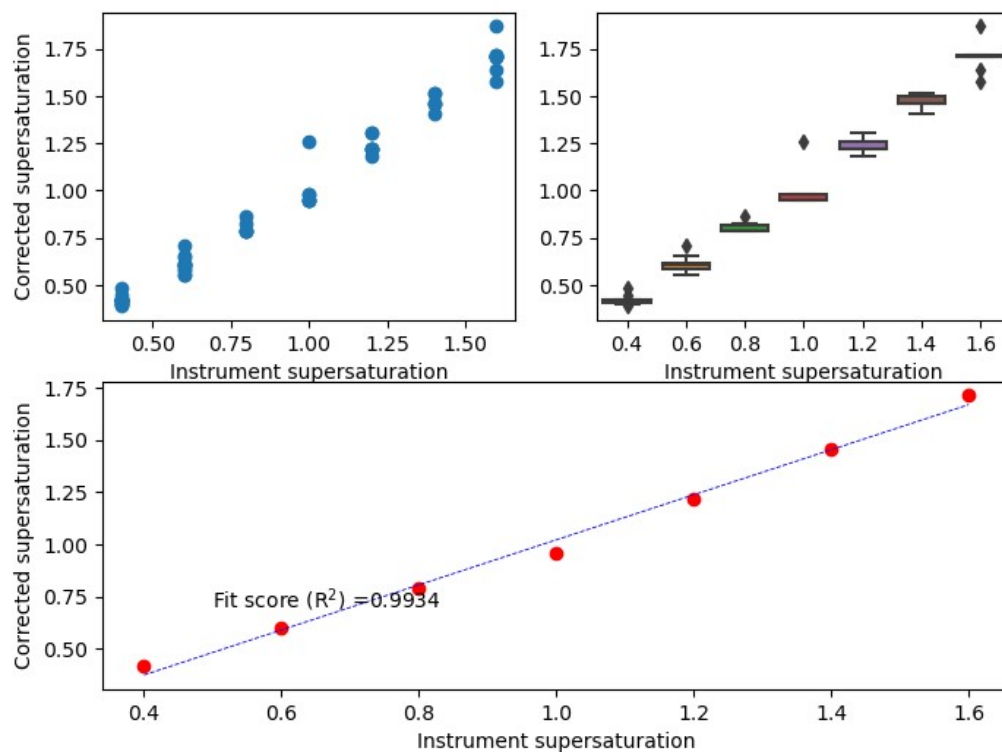


Figure S5. Ammonium sulfate (AS) CCNC instrument calibration results. Instrument supersaturation is set between 0.4 - 1.6% SS and corresponding corrected supersaturation is calculated using PyCAT analysis. Instrument supersaturation and corrected supersaturation are then plotted against each other to obtain R^2 of a linear fit; $R^2 > 0.98$ is considered a well calibrated CCNC within our study.

VI. X:C Parameterizations

Table S9. Commonly found amino acids with their X:C Ratio and Solubility

Compound	X:C Ratio	Solubility (v/v) ^{\$}
Glycine	1.50	0.155
Alanine	1.00	0.117
Glutamic acid	1.00	0.018
Valine	0.60	0.056
Isoleucine	0.50	0.018
Proline	0.60	0.261
dl-Leucine	0.50	0.014
Aspartic Acid	1.25	0.003
Tyrosine	0.44	0.0003
Arginine	1.00	0.487
Histidine	0.83	0.029
Glutamine	1.00	0.017
Serine	1.33	0.156
Phenylalanine	0.33	0.010
Tryptophan	0.36	0.011
Asparagine	1.25	0.019

^{\$}hmdb.ca

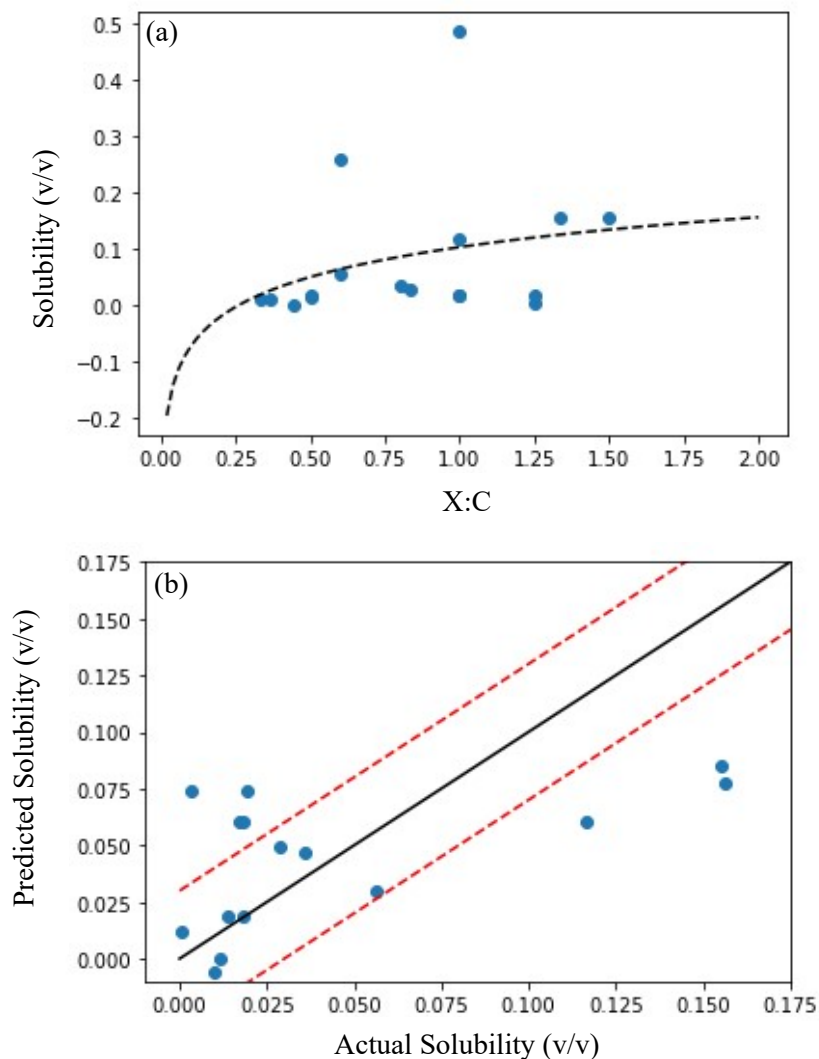
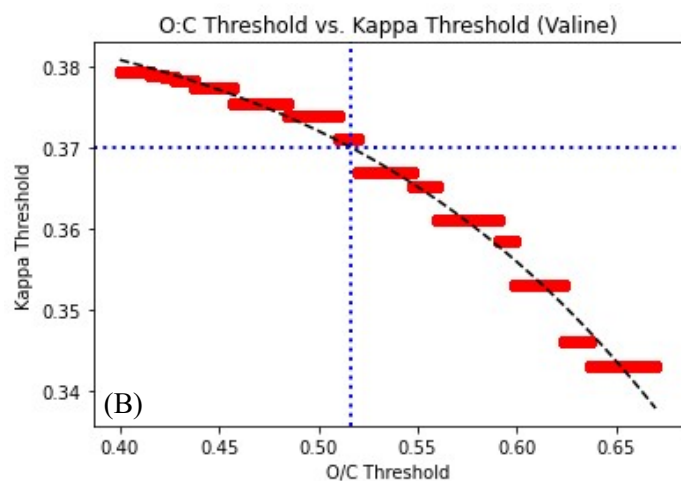
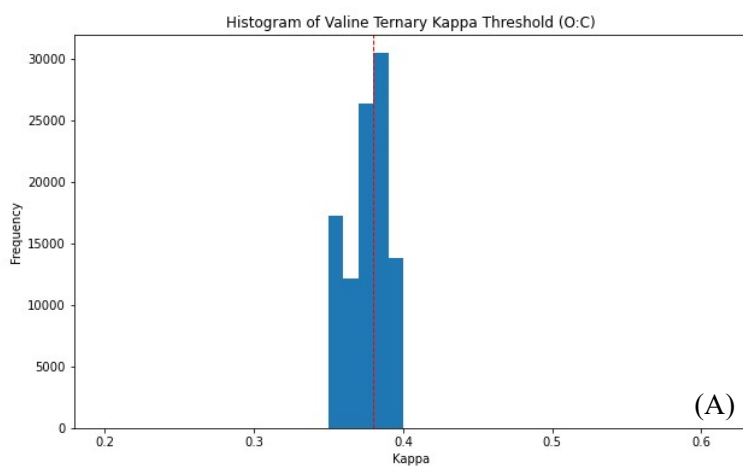


Figure S6. Previous studies account for O:C ratio to parameterize solubility due to the polarity of O-C bonds, but organic compounds such as nitrogen containing organics (e.g.; amino acids) may present solubility influence from N-C bonds. Therefore, the X:C parameterization for solubility is developed for this study by fitting literature solubility values against its X:C ratio. X:C parameterization of solubility for nitrogen containing compounds where (a) X:C vs. solubility (v/v) from Table S9 and (b) Actual solubility (v/v) vs. predicted solubility (v/v) for compounds listed in Table S9, with sold line noting a 1:1 fit and red dashed lines indicating a 10-fold difference from predicted values. Actual solubility (v/v) values are obtained from literature (hmdb.ca) and predicted solubility (v/v) are calculated from Eq. 11.



VII. O:C-LLPS Model Distribution

Figure S7. (A) Histogram Distribution of κ_{thresh} for Val after 100,000 iterations and (B) O:C thresholds vs. κ_{thresh} and corresponding O:C threshold where LLPS is most probable

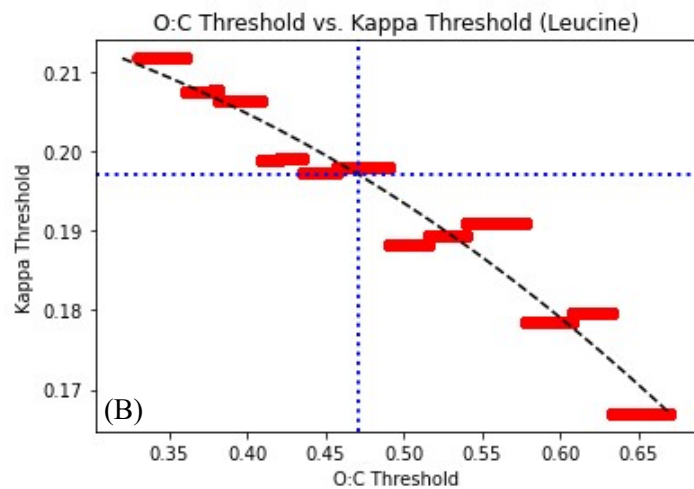
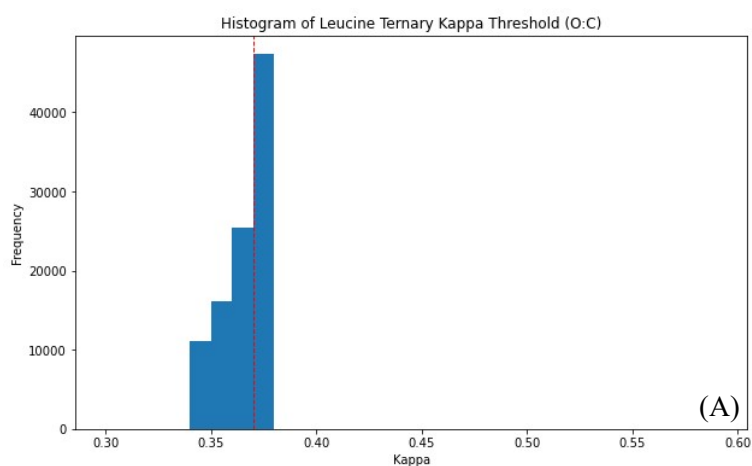
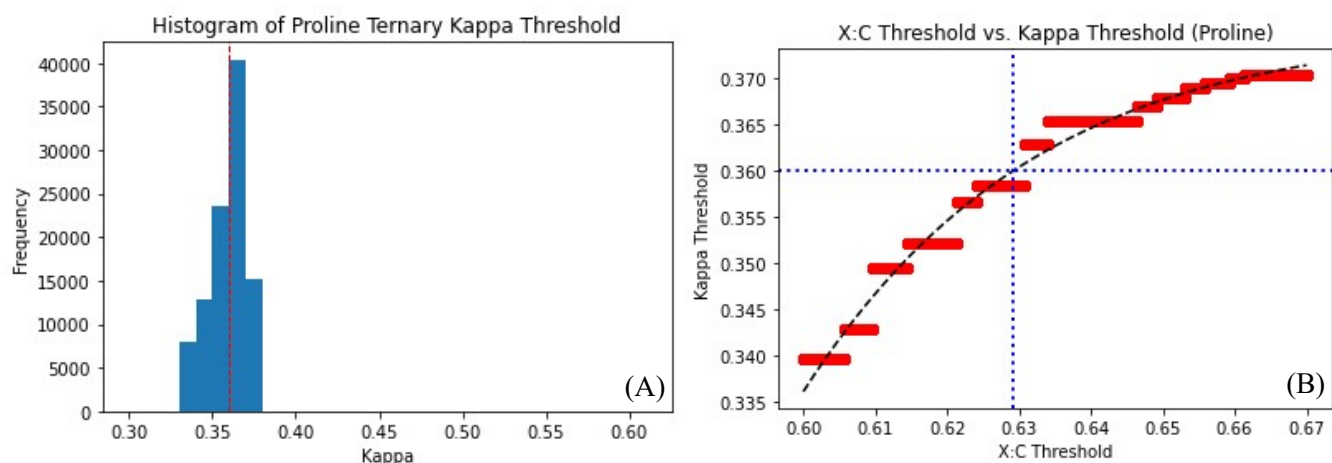


Figure S8. (A) Histogram Distribution of κ_{thresh} for Leu after 100,000 iterations and (B) O:C thresholds vs. κ_{thresh} and corresponding O:C threshold where LLPS is most probable



VIII. X:C-LLPS Model Distributions

Figure S9. (A) Histogram Distribution of κ_{thresh} for Proa after 100,000 iterations and (B) X:C thresholds vs. κ_{thresh} and corresponding X:C threshold where LLPS is most probable

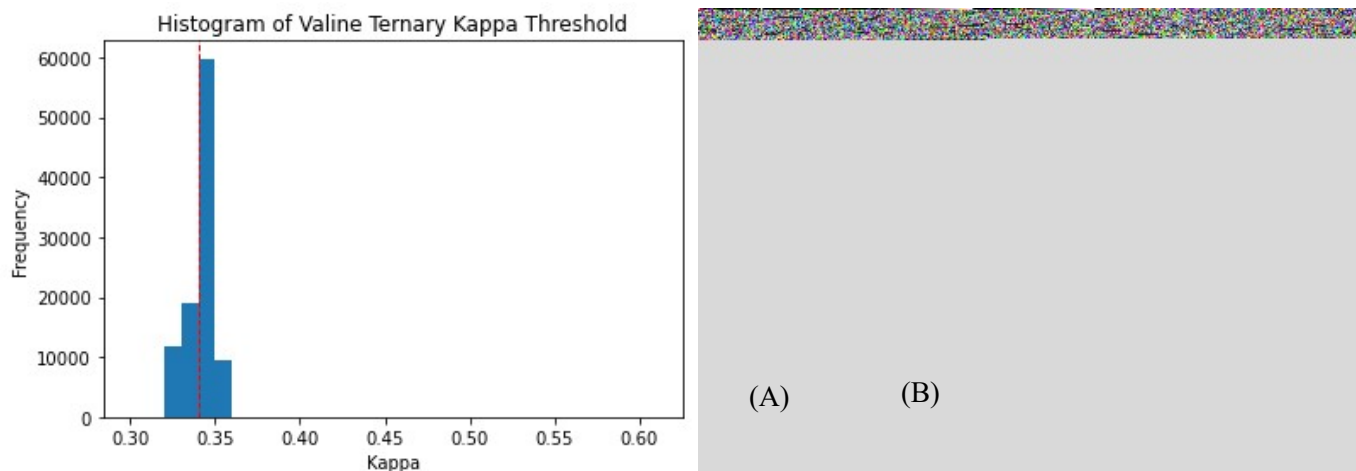


Figure S10. (A) Histogram Distribution of κ_{thresh} for Val after 100,000 iterations and (B) X:C thresholds vs. κ_{thresh} and corresponding X:C threshold where LLPS is most probable

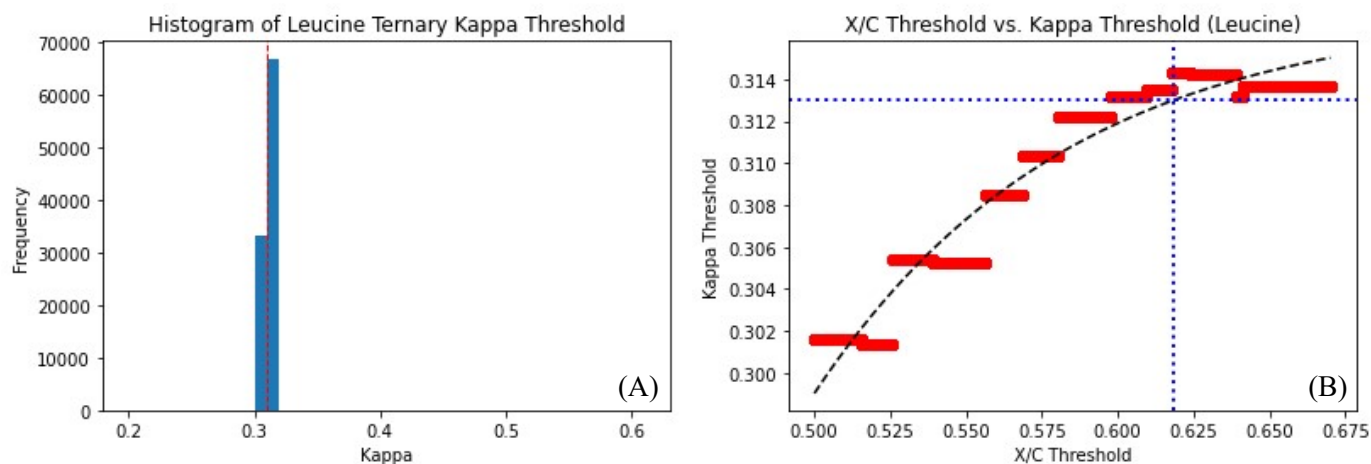


Figure S11. (A) Histogram Distribution of κ_{thresh} for Leu after 100,000 iterations and (B) X:C thresholds vs. κ_{thresh} and corresponding X:C threshold where LLPS is most probable

IX. Surface Tension Measurements

Surface tension measurements of the pure amino acids and binary mixture droplets were taken using a pendant drop goniometer (Biolin Scientific Attention Theta Flex). Solutions were prepared first at the solubility limits of the pure amino acids using Millipore ultra-pure water. The solution compositions are listed in Table S10. A mechanical micro syringe is used to generate a droplet of the solutions $< 10 \mu\text{L}$ at the needle tip. Images were taken at 60 frames/second until the droplet fell. It is assumed as the value plateaus, the surface tension reaches an equilibrium value. The surface tension is determined from fitting the droplet to the Young-Laplace Equation.²⁻⁴ Average surface tension values of the amino acid droplets at its solubility limits are listed in Table S10.

Table S10. Surface Tension of Pure Amino Acids at Solubility Limits and Water

Amino Acid	Water (mL)	Concentration (M)	Surface Tension (mN m^{-1})
Leucine	40	0.171	66.90 ± 0.34
Valine	15	0.569	69.00 ± 0.58
Proline	1	8.686	57.39 ± 0.17
Water			71.33 ± 0.19

Leu and Val were found to have a surface tension value close to the value of pure water (71.3 mN m⁻¹ as measured by the instrument) at its solubility limits. However, Pro is more surface active. Pure Pro is further diluted to assess its surface activity and pure amino acid surface tension is compared against measurements for 2-MGA from Ferdousi-Rokib et al., 2024 (*in review*) (Figure S12).

To assess salting out effects and influence on surface tension, Pro/AS binary mixture surface tension were also measured at several dilutions. The solution concentrations and surface tension results are listed out in Tables S11-S12 and shown in Figure S13.

Table S11. Concentrations of Pro/AS binary mixture dilutions for surface tension measurements

System	Water (mL)	Proline (M)	AS (M)	AS (M)	AS (M)	AS (M)	AS (M)
1	1	8.686	1:9 [§] 0.841	1:4 [§] 2.523	1:1 [§]	4:1 [§]	9:1 [§]
2 [#]	2	4.343	0.420	1.261			
3 [#]	4	2.171	0.210	0.631	1.892		
4 [#]	8	1.086	0.105	0.315	0.946	0.908	
5 [#]	12	0.724	0.070	0.210	0.631	1.892	
6	20	0.434	0.042	0.126	0.378		3.405

[§] Stock solutions were prepared at 1:9, 1:4, 1:1, 4:1 and 9:1 mass weight ratios of Pro/AS, the subsequent molar concentrations of AS are presented in the table.

[#]Stock solutions were prepared at 1:9, 1:4, 1:1, 4:1 and 9:1 mass weight ratios of Pro/AS in 1mL Millipore ultrapure water for System 1. Then System 2 through 5 are then generated by diluting concentrations of System 1.

Table S12. Pure Pro and Pro/AS binary mixture surface tension results

Pro (wt%)	AS (wt%)	Water	Pro (M)	Surface Tension (mN m ⁻¹)	Std Dev
100	0	1	8.686	57.39	0.17
90	10	1	8.686	58.91	0.69
75	25	1	8.686	54.36	0.26
100	0	2	4.343	60.72	0.07
90	10	2	4.343	60.65	0.31
75	25	2	4.343	58.80	0.21
100	0	4	2.171	66.40	0.27
90	10	4	2.171	65.75	0.36
75	25	4	2.171	63.20	0.18
50	50	4	2.171	62.58	0.54

100	0	8	1.086	68.12	0.21
90	10	8	1.086	66.86	0.23
75	25	8	1.086	66.69	0.22
50	50	8	1.086	65.71	0.65
25	75	8	1.086	59.92	0.19
100	0	12	0.724	68.55	0.26
90	10	12	0.724	66.68	0.13
75	25	12	0.724	66.98	0.14
50	50	12	0.724	65.28	0.40
25	75	12	0.724	62.01	0.52
100	0	20	0.434	70.92	0.23
90	10	20	0.434	71.24	0.38
75	25	20	0.434	71.66	0.56
50	50	20	0.434	68.99	0.36
10	90	20	0.434	65.61	0.75

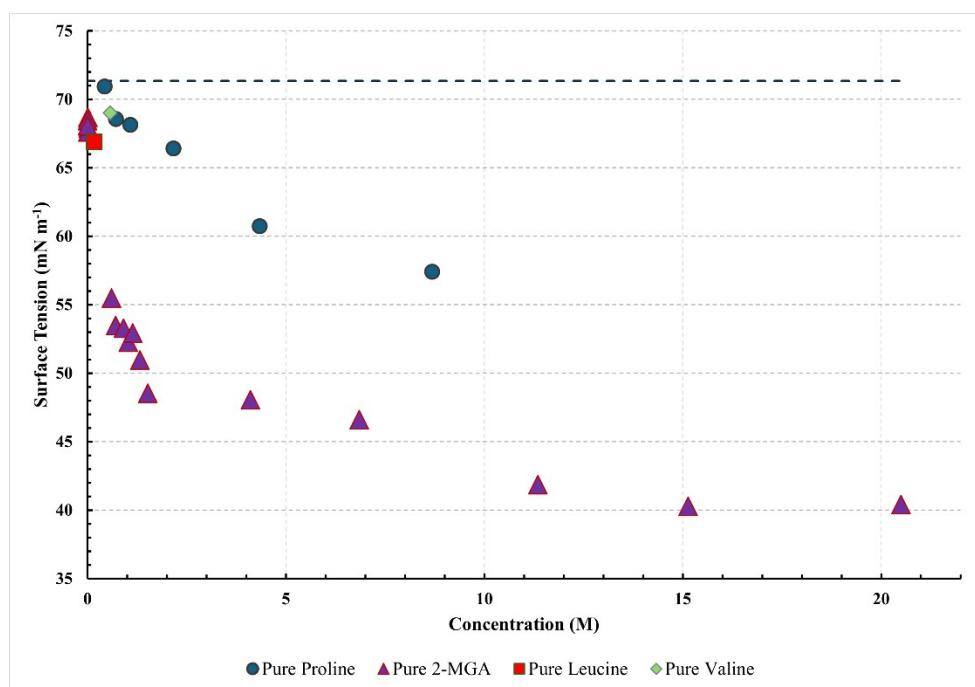


Figure S12. Surface tension measurements for pure amino acids (Leu, Val, Pro) and pure 2-MGA versus concentration. Pure 2-MGA surface tension results are from Ferdousi-Rokib et al., 2024 (*in review*)

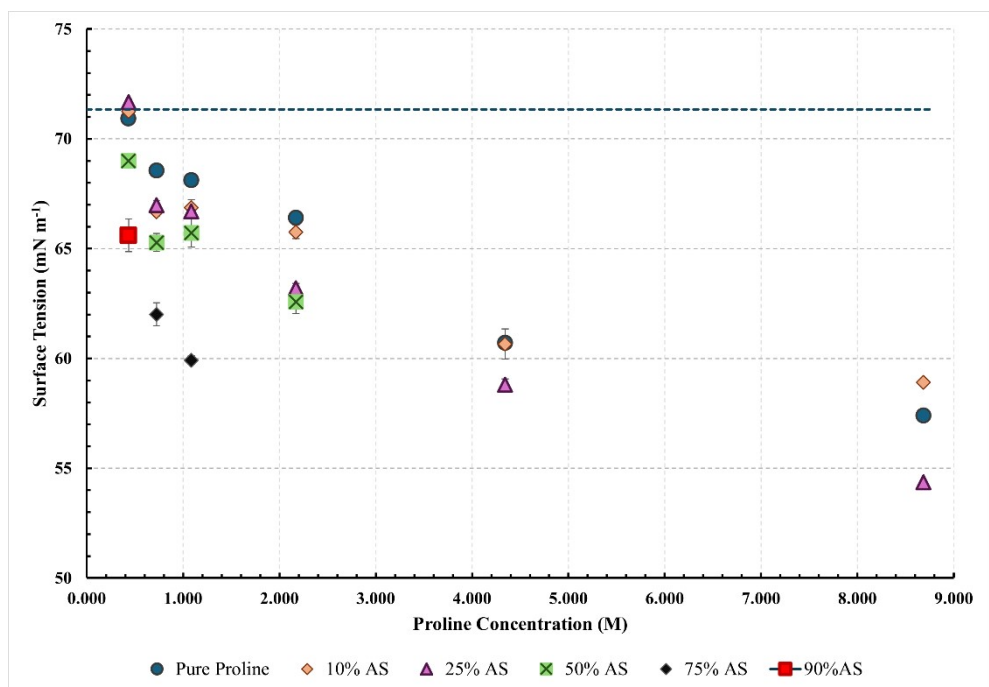


Figure S13. Surface tension measurements for Pro/AS binary system vs. Pro concentration

X. Experimental κ_{CCN} Results

Table S13. Experimental κ_{CCN} for Pro/AS/2-MGA System

Experiment	AS (wt%)	Proline (wt%)	2-MGA (wt%)	κ_{CCN}
1	100	0	0	0.61
2	0	100	0	0.43±0.09
3 [#]	0	0	100	0.14±0.02
4	0	10	90	0.17±0.01
5	0	25	75	0.20±0.02
6	0	50	50	0.23±0.02
7	0	75	25	0.25±0.02
8	0	90	10	0.27±0.03
9	10	90	0	0.25±0.02
10	25	75	0	0.26±0.02
11	50	50	0	0.34±0.03
12	75	25	0	0.44±0.04
13	90	10	0	0.54±0.04
14 [#]	95	0	5	0.62±0.03
15 [#]	90	0	10	0.60±0.02
16 [#]	75	0	25	0.60±0.03
17 [#]	50	0	50	0.50±0.02
18 [#]	25	0	75	0.40±0.03
19 [#]	10	0	90	0.24±0.02
20	45	30	25	0.37±0.03
21	75	10	15	0.52±0.04
22	25	60	15	0.29±0.03
23	60	35	5	0.38±0.03
24	50	25	25	0.34±0.05
25	30	25	45	0.34±0.04
26	30	50	20	0.26±0.03
27	10	75	15	0.24±0.04
28	10	15	75	0.24±0.02

[#]Data points from Ferdousi-Rokib et al (*in review*)

Table S14. Experimental κ_{CCN} for Val/AS/2-MGA System

Experiment	AS (wt%)	Valine (wt%)	2-MGA (wt%)	κ_{CCN}
1	100	0	0	0.61
2	0	100	0	0.06±0.02
3 [#]	0	0	100	0.14±0.02
4	0	10	90	0.18±0.01
5	0	25	75	0.18±0.02
6	0	50	50	0.16±0.02
7	0	75	25	0.14±0.03
8	0	90	10	0.07±0.04
9	10	90	0	0.18±0.01
10	25	75	0	0.23±0.02
11	50	50	0	0.39±0.05
12	75	25	0	0.50±0.03
13	90	10	0	0.55±0.03
14 [#]	95	0	5	0.62±0.03
15 [#]	90	0	10	0.60±0.02
16 [#]	75	0	25	0.60±0.03
17 [#]	50	0	50	0.50±0.02
18 [#]	25	0	75	0.40±0.03
19 [#]	10	0	90	0.24±0.02
20	10	57.5	32.5	0.22±0.02
21	50	37.5	12.5	0.34±0.02
22	10	20	70	0.22±0.02
23	75	10	15	0.55±0.05
24	30	65	5	0.25±0.02
25	60	35	5	0.41±0.04
26	50	25	25	0.53±0.05
27	35	35	30	0.24±0.03
28	30	25	45	0.23±0.03
29	30	10	60	0.31±0.04
30	10	75	15	0.14±0.01

[#]Data points from Ferdousi-Rokib et al (*in review*)

Table S15. Experimental κ_{CCN} for Leu/AS/2-MGA System

Experiment	AS (wt%)	Leucine (wt%)	2-MGA (wt%)	κ_{CCN}
1	100	0	0	0.61
2	0	100	0	0.01±0.00
3 [#]	0	0	100	0.14±0.02
4	0	10	90	0.09±0.02
5	0	25	75	0.10±0.02
6	0	50	50	0.03±0.01
7	0	75	25	0.02±0.02
8	0	90	10	0.02±0.02
9	10	90	0	0.11±0.01
10	25	75	0	0.14±0.01
11	50	50	0	0.14±0.01
12	75	25	0	0.47±0.03
13	90	10	0	0.56±0.03
14 [#]	95	0	5	0.62±0.03
15 [#]	90	0	10	0.60±0.02
16 [#]	75	0	25	0.60±0.03
17 [#]	50	0	50	0.50±0.02
18 [#]	25	0	75	0.40±0.03
19 [#]	10	0	90	0.24±0.02
20	10	60	30	0.12±0.01
21	75	15	10	0.50±0.03
22	65	15	20	0.50±0.03
23	40	55	10	0.22±0.02
24	50	25	25	0.42±0.05
25	30	25	45	0.29±0.04
26	30	50	20	0.19±0.03
27	10	15	75	0.20±0.01
28	10	75	15	0.15±0.03

[#]Data points from Ferdousi-Rokib et al (*in review*)

XI. Model Results

Table S16. Predicted κ values for traditional Köhler, O:C Solubility, X:C Solubility, O:C-LLPS, X:C-LLPS Models for Proline/2-MGA/AS System

Experiment	AS (wt%)	Proline (wt%)	2- MGA (wt%)	κ_{ZSR}	$\kappa_{\text{O:C}}$	$\kappa_{\text{X:C}}$	$\kappa_{\text{O:C-LLPS}}$	$\kappa_{\text{X:C-LLPS}}$
1	100	0	0	0.61	0.61	0.61	0.61	0.61
2	0	100	0	0.22	0.02	0.22	0.02	0.22
3 [#]	0	0	100	0.15	0.15	0.15	0.14	0.14
4	0	10	90	0.15	0.16	0.16	0.15	0.22
5	0	25	75	0.16	0.17	0.17	0.13	0.22
6	0	50	50	0.18	0.19	0.19	0.10	0.22
7	0	75	25	0.20	0.21	0.21	0.02	0.20
8	0	90	10	0.21	0.21	0.21	0.02	0.21
9	10	90	0	0.25	0.24	0.24	0.07	0.38
10	25	75	0	0.30	0.29	0.29	0.15	0.41
11	50	50	0	0.39	0.38	0.38	0.28	0.47
12	75	25	0	0.49	0.49	0.49	0.44	0.54
13	90	10	0	0.56	0.56	0.56	0.54	0.58
14 [#]	95	0	5	0.58	0.59	0.59	0.57	0.43
15 [#]	90	0	10	0.54	0.57	0.57	0.54	0.55
16 [#]	75	0	25	0.46	0.50	0.50	0.54	0.29
17 [#]	50	0	50	0.33	0.39	0.39	0.53	0.43
18 [#]	25	0	75	0.23	0.27	0.27	0.41	0.46
19 [#]	10	0	90	0.18	0.20	0.20	0.26	0.41
20	45	30	25	0.34	0.36	0.36	0.29	0.30
21	75	10	15	0.47	0.50	0.50	0.46	0.24
22	25	60	15	0.29	0.28	0.28	0.17	0.35
23	60	35	5	0.42	0.43	0.43	0.36	0.61
24	50	25	25	0.36	0.39	0.39	0.32	0.22
25	30	25	45	0.27	0.30	0.30	0.24	0.14
26	30	50	20	0.30	0.30	0.30	0.21	0.22
27	10	75	15	0.24	0.24	0.24	0.08	0.22
28	10	15	75	0.19	0.61	0.20	0.18	0.22

[#]Data points from Ferdousi-Rokib et al (*in review*)

Table S17. Predicted κ values for traditional Köhler, O:C Solubility, X:C Solubility, O:C-LLPS, X:C-LLPS, and Weighted Average Models for Valine/2-MGA/AS System

Experiment	AS (wt%)	Valine (wt%)	2- MGA (wt%)	κ_{ZSR}	$\kappa_{\text{O:C}}$	$\kappa_{\text{X:C}}$	$\kappa_{\text{O:C-LLPS}}$	$\kappa_{\text{X:C-LLPS}}$	κ_{WA}
1	100	0	0	0.61	0.61	0.61	0.61	0.61	0.61
2	0	100	0	0.20	0.02	0.20	0.02	0.20	0.15
3 [#]	0	0	100	0.15	0.16	0.16	0.14	0.15	0.14
4	0	10	90	0.15	0.17	0.17	0.15	0.20	0.18
5	0	25	75	0.16	0.17	0.17	0.13	0.20	0.18
6	0	50	50	0.17	0.18	0.18	0.09	0.20	0.17
7	0	75	25	0.19	0.10	0.19	0.02	0.19	0.14
8	0	90	10	0.20	0.06	0.20	0.02	0.20	0.14
9	10	90	0	0.23	0.20	0.23	0.06	0.36	0.27
10	25	75	0	0.28	0.28	0.29	0.13	0.39	0.31
11	50	50	0	0.38	0.39	0.39	0.26	0.45	0.39
12	75	25	0	0.48	0.50	0.50	0.42	0.52	0.49
13	90	10	0	0.56	0.56	0.56	0.53	0.57	0.56
14 [#]	95	0	5	0.58	0.59	0.59	0.57	0.57	0.57
15 [#]	90	0	10	0.54	0.56	0.56	0.54	0.54	0.54
16 [#]	75	0	25	0.46	0.49	0.49	0.54	0.54	0.54
17 [#]	50	0	50	0.33	0.38	0.38	0.53	0.53	0.53
18 [#]	25	0	75	0.23	0.27	0.27	0.41	0.41	0.41
19 [#]	10	0	90	0.18	0.20	0.20	0.26	0.26	0.26
20	10	57.5	32.5	0.21	0.22	0.22	0.08	0.22	0.18
21	50	37.5	12.5	0.37	0.38	0.38	0.30	0.37	0.35
22	10	20	70	0.19	0.21	0.21	0.17	0.30	0.26
23	75	10	15	0.47	0.50	0.50	0.45	0.54	0.52
24	30	65	5	0.30	0.30	0.30	0.16	0.29	0.25
25	60	35	5	0.41	0.43	0.43	0.34	0.41	0.39
26	50	25	25	0.36	0.38	0.38	0.31	0.44	0.40
27	35	35	30	0.30	0.32	0.32	0.26	0.37	0.34
28	30	25	45	0.27	0.29	0.29	0.23	0.39	0.34
29	30	10	60	0.26	0.29	0.29	0.26	0.48	0.41
30	10	75	15	0.22	0.23	0.23	0.07	0.22	0.18

[#]Data points from Ferdousi-Rokib et al (*in review*)

Table S18. Predicted κ values for traditional Köhler, O:C Solubility, X:C Solubility, O:C-LLPS, X:C-LLPS, and Weighted Average Models for Leucine/2-MGA/AS System

Experiment	AS (wt%)	Leucine (wt%)	2- MGA (wt%)	κ_{ZSR}	$\kappa_{\text{O:C}}$	$\kappa_{\text{X:C}}$	$\kappa_{\text{O:C-LLPS}}$	$\kappa_{\text{X:C-LLPS}}$	κ_{WA}
1	100	0	0	0.61	0.61	0.61	0.61	0.61	0.61
2	0	100	0	0.16	0.004	0.16	0.004	0.16	0.02
3 [#]	0	0	100	0.15	0.16	0.16	0.14	0.12	0.14
4	0	10	90	0.15	0.16	0.16	0.15	0.16	0.15
5	0	25	75	0.15	0.16	0.16	0.12	0.16	0.13
6	0	50	50	0.15	0.10	0.16	0.00	0.16	0.09
7	0	75	25	0.16	0.03	0.16	0.00	0.16	0.02
8	0	90	10	0.16	0.02	0.16	0.00	0.16	0.02
9	10	90	0	0.19	0.12	0.21	0.05	0.32	0.08
10	25	75	0	0.24	0.23	0.28	0.11	0.36	0.15
11	50	50	0	0.34	0.38	0.39	0.25	0.43	0.28
12	75	25	0	0.46	0.51	0.51	0.41	0.51	0.43
13	90	10	0	0.55	0.57	0.57	0.52	0.57	0.53
14 [#]	95	0	5	0.58	0.59	0.59	0.56	0.56	0.56
15 [#]	90	0	10	0.54	0.56	0.56	0.56	0.56	0.56
16 [#]	75	0	25	0.46	0.49	0.49	0.54	0.54	0.54
17 [#]	50	0	50	0.33	0.38	0.38	0.44	0.44	0.44
18 [#]	25	0	75	0.23	0.27	0.27	0.26	0.26	0.26
19 [#]	10	0	90	0.18	0.20	0.20	0.19	0.19	0.19
20	10	60	30	0.19	0.20	0.20	0.06	0.20	0.08
21	75	15	10	0.46	0.50	0.50	0.47	0.51	0.48
22	65	15	20	0.41	0.45	0.45	0.39	0.50	0.40
23	40	55	10	0.29	0.34	0.34	0.20	0.30	0.22
24	50	25	25	0.34	0.39	0.39	0.35	0.43	0.32
25	30	25	45	0.25	0.29	0.29	0.22	0.37	0.24
26	30	50	20	0.26	0.30	0.30	0.18	0.29	0.20
27	10	15	75	0.18	0.20	0.20	0.17	0.30	0.19
28	10	75	15	0.19	0.17	0.20	0.05	0.18	0.07

[#]Data points from Ferdousi-Rokib et al (*in review*)

Table S19. χ^2 goodness fits for all models

Ternary System	χ^2						
	Kohler	O:C	X:C	O:C-LLPS	X:C-LLPS	Weighted Average	Best Fit
Leucine	4.90E+11	8.19	4.90E+11	6.29	4.90E+11	2.3	2.3
Valine	10.59	10.4	2.50E+11	19.11	19.12	2.85	2.85
Proline	8.29	12.39	14.95	8.93	5.23		5.23

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Figure S16. Comparison of predicted κ from hygroscopicity models to experimental κ results of :ei ternary mixture; a 1:1 correlation is represented by the black line, and 10% error is outlined in grey dashed lines.