

Supporting Information for:

Simulations of Interfacial Tension of Liquid-Liquid Ternary Mixtures Using Optimized Parametrization for Coarse-Grained Models

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S1. Workflow

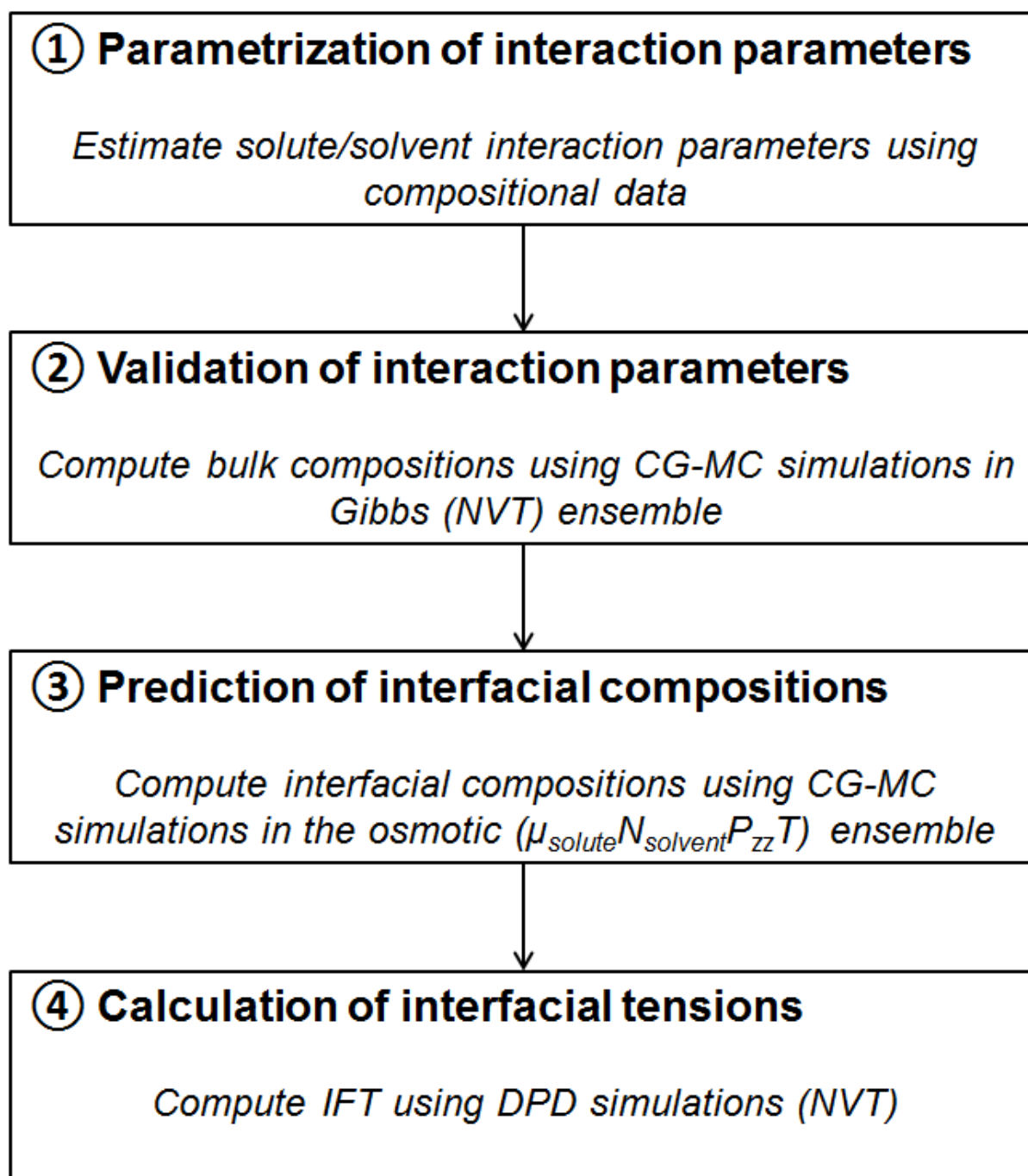


Figure S1. Workflow representing the steps of the methodology used in this work to compute interfacial tension of ternary mixtures from bulk compositions.

S2. Determination of solute/solvent interaction parameters following the approaches #3 and #4.

Using the parametrization approaches #3 and #4, one of the solute/solvent parameter is calculated with Hildebrand solubility parameters following the model developed by Travis *et al.*¹ (see equations 15 and 16 in the main paper), and the second solute/solvent parameter is obtained with a direct fitting performed on available compositional data in order to respect the distribution constant of the solute in both bulk phases. Distribution constant can be expressed according to the following equation:

$$K^{exp} = \frac{x_{solute}^{org.}}{x_{solute}^{aq.}}$$

where $x_{solute}^{org.}$ is the experimental molar fraction of solute in the organic phase and $x_{solute}^{aq.}$ is the experimental molar fraction of solute in the aqueous phase.

As an example, approach #3 is applied to the water/benzene/1,4-dioxane system. The interaction parameter between 1,4-dioxane and benzene is calculated using Hildebrand solubility parameter ($a_{1,4-dioxane/benzene} = 67.45 k_B T$). Then, composition of bulk phases is calculated for different values of parameter $a_{1,4-dioxane/water}$ using several Coarse-grained Monte Carlo (CG-MC) simulations in the Gibbs (NVT) ensemble. Variation of $\log(K^{exp})$ as a function of the $a_{1,4-dioxane/water}$ is presented in Figure S2. Interaction parameter between 1,4-dioxane and water is obtained by reading the Figure S2 ($a_{1,4-dioxane/water} = 43.1 k_B T$).

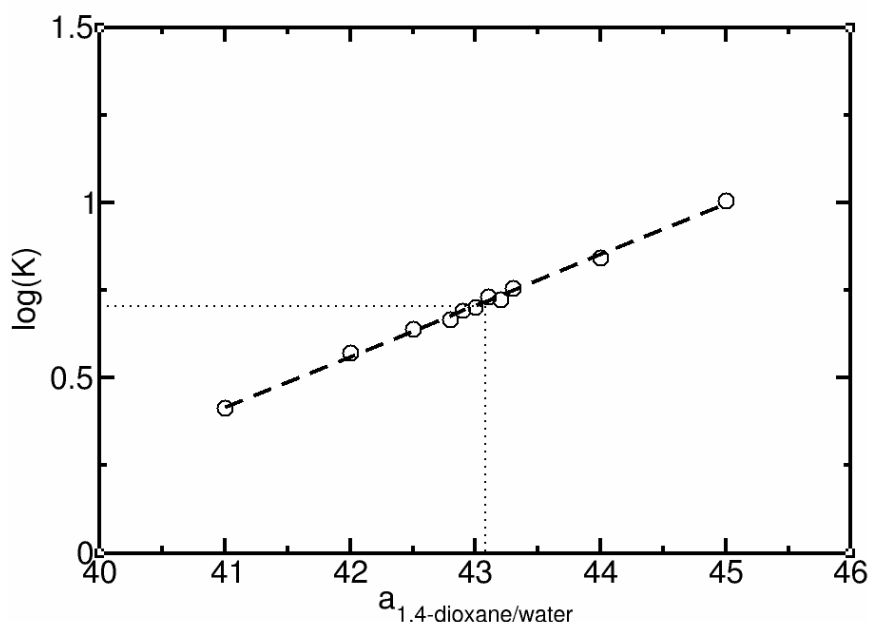


Figure S2. Application of the parametrization approach #3 on the water/benzene/1,4-dioxane system. Composition used is the less concentrated in 1,4-dioxane: $K^{exp} \approx 0.70$.

S3. Experimental data from the literature for the studied systems

Table S1. Experimental data for the water/benzene/1,4-dioxane system at 298.15 K extracted from the work by Backes *et al.*²

Aqueous phase				Organic phase				IFT
Molar fraction			Density	Molar fraction			Density	
x'_1 (%)	x'_2 (%)	x'_3 (%)	ρ' (g/cm ³)	x''_1 (%)	x''_2 (%)	x''_3 (%)	ρ'' (g/cm ³)	σ (mN/m)
98.015	0.025	1.960	1.0042	0.872	89.228	9.900	0.889	18.3
95.586	0.054	4.360	1.0120	1.320	78.280	20.400	0.9061	12.2
93.322	0.058	6.620	1.0178	2.650	67.150	30.200	0.9222	8.7
90.537	0.063	9.400	1.0234	4.410	55.090	40.500	0.9398	5.6
84.990	0.110	14.900	1.0301	10.300	37.100	52.600	0.9660	2.6

Interfacial tension between water and benzene in the absence of solute is 33.8 mN/m at 298.15 K.

Table S2. Experimental data for the water/chloroform/acetone system at 298.15 K extracted from the work of Backes *et al.*²

Aqueous phase				Organic phase				IFT
Molar fraction			Density	Molar fraction			Density	
x'_1 (%)	x'_2 (%)	x'_3 (%)	ρ' (g/cm ³)	x''_1 (%)	x''_2 (%)	x''_3 (%)	ρ'' (g/cm ³)	σ (mN/m)
99.042	0.140	0.818	0.9939	1.820	84.280	13.900	1.3870	21.0
97.957	0.143	1.900	0.9892	2.830	73.070	24.100	1.3152	16.3
95.222	0.168	4.610	0.9792	5.500	55.400	39.100	1.2024	9.8
91.960	0.180	7.860	0.9682	9.320	41.180	49.500	1.1131	5.7
87.787	0.213	12.000	0.9554	14.60	29.600	55.800	1.0395	3.0

Interfacial tension between water and chloroform in the absence of solute is 30.8 mN/m at 298.15 K.

Table S3. Experimental data for the water/benzene/acetic acid system at 298.15 K extracted from the work of Backes *et al.*²

Aqueous phase				Organic phase				IFT
Molar fraction			Density	Molar fraction			Density	
x'_1 (%)	x'_2 (%)	x'_3 (%)	ρ' (g/cm ³)	x''_1 (%)	x''_2 (%)	x''_3 (%)	ρ'' (g/cm ³)	σ (mN/m)
94.041	0.079	5.88	1.0198	0.43	97.89	1.68	0.8750	17.3
87.852	0.148	12.0	1.0358	0.852	94.288	4.86	0.8778	11
79.608	0.392	20.0	1.0463	1.05	89.49	9.46	0.8820	7.0
68.77	1.43	29.8	1.0499	1.85	82.15	16.0	0.8889	4.0
56.35	3.75	39.9	1.0784	4.14	69.96	25.9	0.9013	2.0

Interfacial tension between water and benzene in the absence of solute is 33.8 mN/m at 298.15 K.

Table S4. Experimental data for the water/benzene/2-propanol system at 298.15 K extracted from the work of Sada *et al.*³. Data were converted from mass fractions to molar fractions.

Aqueous phase				Organic phase				IFT
Molar fraction			Density	Molar fraction			Density	
x'_1 (%)	x'_2 (%)	x'_3 (%)	ρ' (g/cm ³)	x''_1 (%)	x''_2 (%)	x''_3 (%)	ρ'' (g/cm ³)	σ (mN/m)
98.5	0.1	1.4	0.9869	0.0	98.6	1.4	0.8722	19.99
97.0	0.0	3.0	0.9803	0.0	96.8	3.2	0.8706	13.27
94.2	0.1	5.7	0.9699	0.4	90.0	9.6	0.8647	5.92
93.1	0.1	6.8	0.9656	0.8	84.3	14.8	0.8604	4.42
92.2	0.2	7.6	0.9620	2.4	77.4	20.2	0.8564	2.84
90.3	0.2	9.5	0.9530	12.2	53.8	34.0	0.8474	1.47
89.2	0.3	10.5	0.9481	17.8	43.7	38.5	0.8454	1.05

Interfacial tension between water and benzene in the absence of solute is 33.8 mN/m at 298.15 K.

Table S5. Experimental data for the water/hexane/acetone system at 298.15 K extracted from the work of Paul and Chazal⁴.

Aqueous phase				Organic phase				IFT
Molar fraction			Density	Molar fraction			Density	
x'_1 (%)	x'_2 (%)	x'_3 (%)	ρ' (g/cm ³)	x''_1 (%)	x''_2 (%)	x''_3 (%)	ρ'' (g/cm ³)	σ (mN/m)
99.8	0.0	0.2	0.9950	0.06	99.0	0.9	0.6623	41.6
97.5	0.0	2.5	0.9865	0.06	96.5	3.4	0.6625	20.5
94.9	0.0	5.1	0.9860	0.06	92.4	7.5	0.6641	12.8
89.4	0.0	10.6	0.9584	0.06	86.2	13.7	0.6671	6.3
85.7	0.0	14.3	0.9444	0.06	79.2	20.7	0.6698	4.7
77.8	0.0	22.2	0.9155	1.20	65.1	33.7	0.6790	2.9
73.3	0.1	26.6	0.9040	2.40	58.9	38.7	0.6856	2.2
69.2	0.4	30.4	0.8931	3.70	51.5	44.8	0.6904	1.5
65.2	0.6	34.2	0.8787	4.60	44.9	50.5	0.6991	1.2
60.5	0.9	38.6	0.8674	5.50	39.0	55.5	0.7092	1.0
54.1	1.5	44.4	0.8464	8.50	31.6	59.9	0.7231	0.9

Interfacial tension between water and hexane in the absence of solute is 50.1 mN/m at 298.15 K.

Table S6. Experimental data for the water/hexane/2-propanol system at 298.15 K extracted from the work of Paul and Chazal⁴.

Aqueous phase				Organic phase				IFT
Molar fraction			Density	Molar fraction			Density	
x'_1 (%)	x'_2 (%)	x'_3 (%)	ρ' (g/cm ³)	x''_1 (%)	x''_2 (%)	x''_3 (%)	ρ'' (g/cm ³)	σ (mN/m)
99.8	0.0	0.2	0.9954	0.06	99.6	0.3	0.6623	35.1
99.1	0.0	0.9	0.9901	0.06	98.9	1.0	0.6627	19.1
97.7	0.0	2.3	0.9850	0.06	97.4	2.5	0.6634	12.4
95.0	0.0	5.0	0.9739	0.06	95.0	4.9	0.6645	8.4
92.3	0.0	7.7	0.9643	0.06	92.8	7.1	0.6655	5.6
89.4	0.0	10.6	0.9527	0.40	89.4	10.2	0.6690	4.5
87.0	0.0	13.0	0.9402	0.90	84.4	14.7	0.6723	3.0
83.2	0.2	16.6	0.9241	1.70	80.8	17.5	0.6770	2.4
76.7	1.0	22.3	0.8995	3.00	73.9	23.1	0.6834	1.7
63.8	4.0	32.2	0.8450	8.00	59.3	32.7	0.6973	0.9
51.7	8.5	39.8	0.7922	17.60	41.7	40.7	0.7211	0.7

Interfacial tension between water and hexane in the absence of solute is 50.1 mN/m at 298.15 K.

S4. Calculation of volume fractions from molar fractions.

Volume fractions are calculated from molar fractions according to the coarse-grained model. In this work, all beads have the same volume.

$$\theta_{water} = \frac{\frac{x_{water}}{N_m}}{\frac{x_{water}}{N_m} + x_{solute}} \text{ and } \phi_{org.} = \frac{x_{org.} \times N_{org.}}{x_{org.} \times N_{org.} + x_{solute}},$$

Where x_{water} and θ_{water} are the molar and the volume fraction of water representing the aqueous phase, respectively, while $x_{org.}$ and $\phi_{org.}$ are the molar and the volume fraction of molecules representing the organic phase, respectively. N_m is the degree of coarse-graining (i.e. the number of water molecules in a bead of water) and $N_{org.}$ is the number of beads constituting a molecule of the organic solvent ($N_{org.} = 1$ for benzene and chloroform, and $N_{org.} = 2$ for hexane). Note that miscibility between solvents is completely neglected.

S5. Dependence of solute/solvent Flory-Huggins parameters with compositional data.

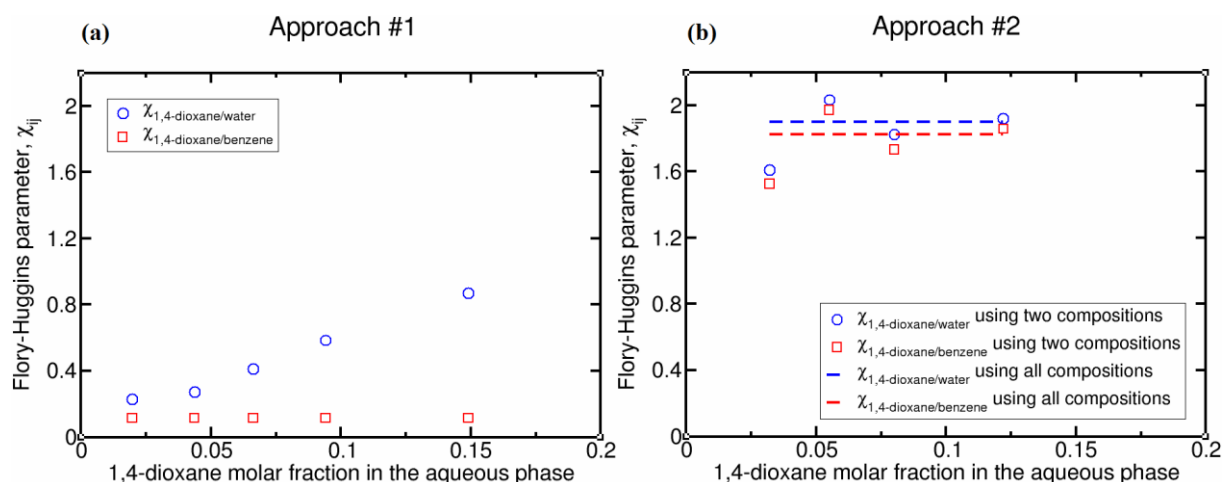


Figure S3. Variation of $\chi_{\text{solute/solvent}}$ parameters, as a function of each experimental composition for the water/benzene/1,4-dioxane system (obtained using approach #1). The composition is expressed using the molar fraction of solute in the aqueous phase. (b) Variation of $\chi_{\text{solute/solvent}}$ parameters as a function of the average solute molar fractions in the aqueous phase for each pair of compositions for the water/benzene/1,4-dioxane system (obtained using approach #2).

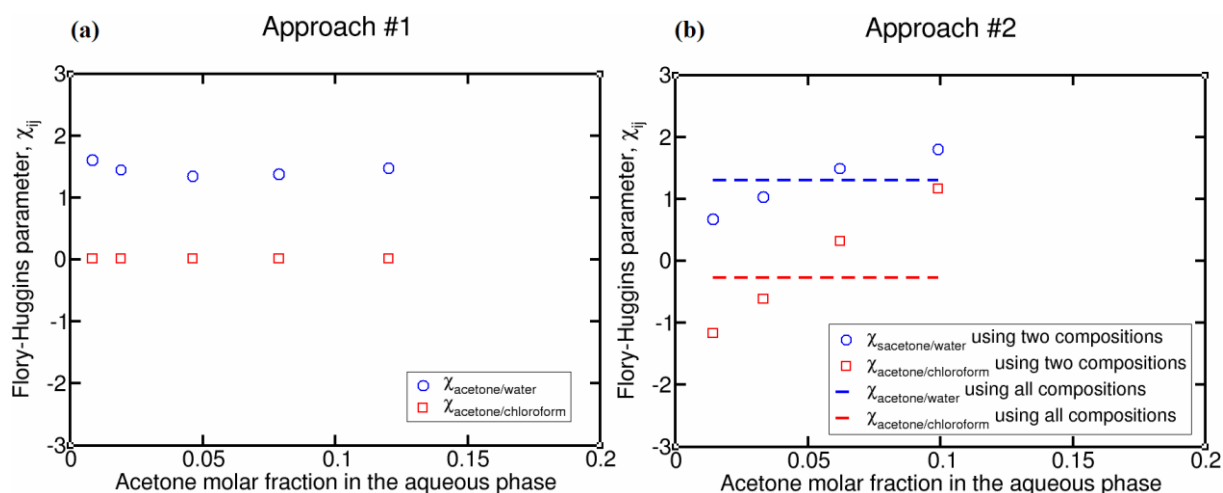


Figure S4. Variation of $\chi_{\text{solute/solvent}}$ parameters, as a function of each experimental composition for the water/chloroform/acetone system (obtained using approach #1). The composition is expressed using the molar fraction of solute in the aqueous phase. (b) Variation of $\chi_{\text{solute/solvent}}$ parameters as a function of the average solute molar fractions in the aqueous phase for each pair of compositions for the water/chloroform/acetone system (obtained using approach #2).

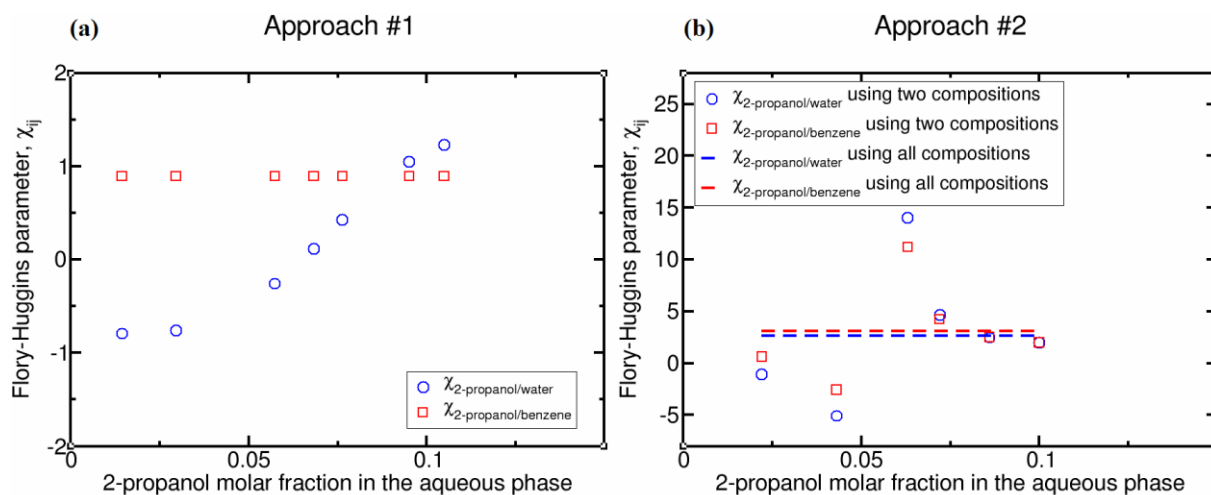


Figure S5. Variation of $\chi_{solute/solvent}$ parameters, as a function of each experimental composition for the water/benzene/2-propanol system (obtained using approach #1). The composition is expressed using the molar fraction of solute in the aqueous phase. (b) Variation of $\chi_{solute/solvent}$ parameters as a function of the average solute molar fractions in the aqueous phase for each pair of compositions for the water/benzene/2-propanol system (obtained using approach #2).

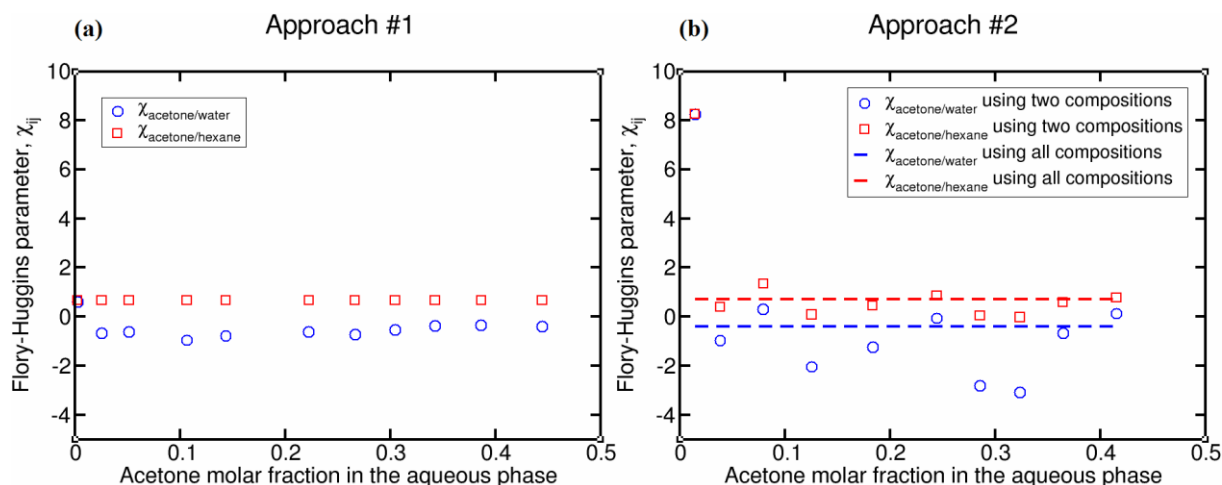


Figure S6. Variation of $\chi_{solute/solvent}$ parameters, as a function of each experimental composition for the water/hexane/acetone system (obtained using approach #1). The composition is expressed using the molar fraction of solute in the aqueous phase. (b) Variation of $\chi_{solute/solvent}$ parameters as a function of the average solute molar fractions in the aqueous phase for each pair of compositions for the water/hexane/acetone system (obtained using approach #2).

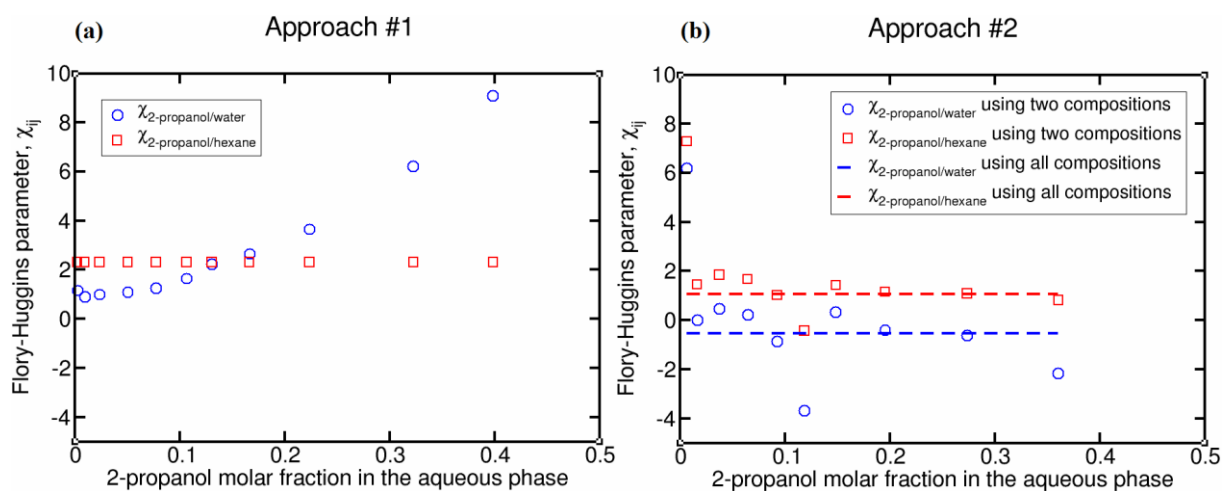


Figure S7. Variation of $\chi_{solute/solvent}$ parameters, as a function of each experimental composition for the water/hexane/2-propanol system (obtained using approach #1). The composition is expressed using the molar fraction of solute in the aqueous phase. (b) Variation of $\chi_{solute/solvent}$ parameters as a function of the average solute molar fractions in the aqueous phase for each pair of compositions for the water/hexane/2-propanol system (obtained using approach #2).

S6. Results of CG-MC simulations in the Gibbs ensemble (NVT).

1. Liquid-liquid equilibrium ternary diagrams for water/chloroform/acetone system at 298.15K

Parametrization approaches #1, #3 and #4 were carried out with the less concentrated acetone composition. Approach #2 is based on the three compositions less concentrated in acetone.

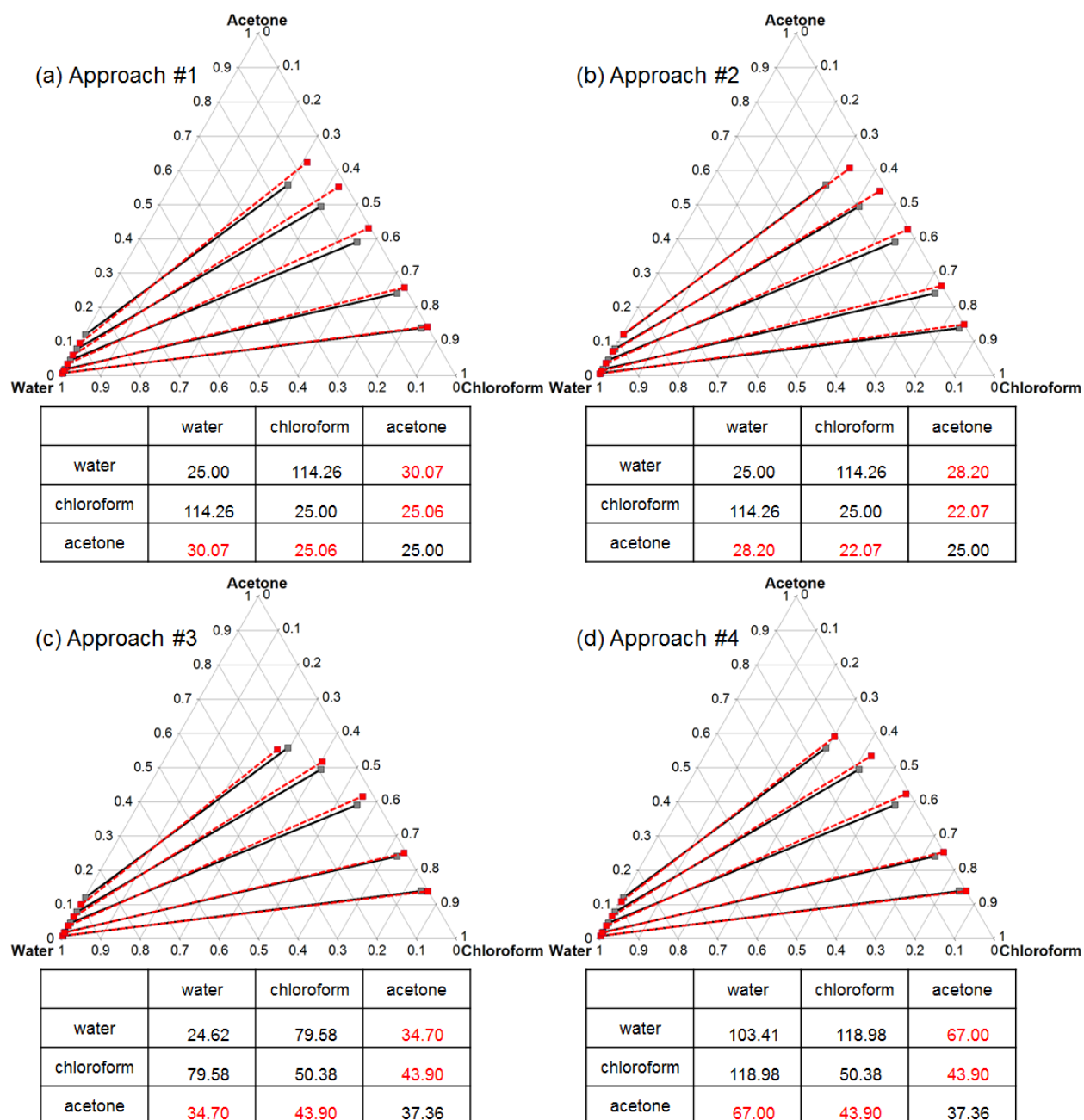


Figure S8. Liquid-liquid equilibrium ternary diagrams for the water/chloroform/acetone system at 298.15K for (a) approach #1, (b) approach #2, (3) approach #3 and (4) approach #4. Experimental data are plotted in black and the results from the CG-MC simulations are in red. DPD interaction parameters used to compute LLE are located below each ternary plot. Numbers in red represent the solute/solvent parameters obtained for each approach.

2. Liquid-liquid equilibrium ternary diagram for water/benzene/acetic acid system at 298.15K.

Parametrization approaches #1, #3 and #4 were carried out with the second composition the less concentrated in acetic acid. Approach #2 is based on the five compositions.

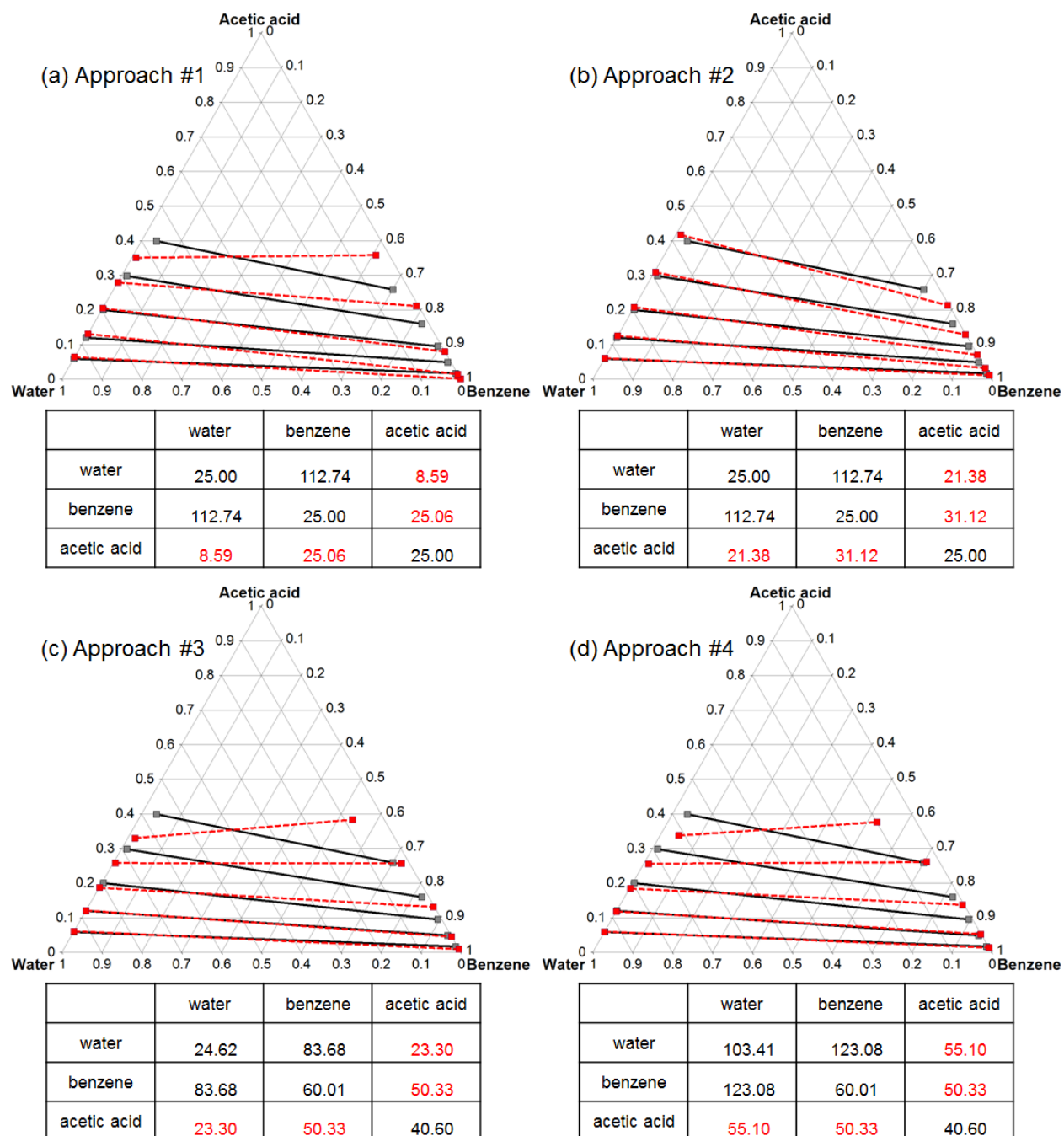


Figure S9. Liquid-liquid equilibrium ternary diagrams for the water/benzene/acetic acid system at 298.15K for (a) approach #1, (b) approach #2, (3) approach #3 and (4) approach #4. Experimental data are plotted in black and the results from the CG-MC simulations are in red. DPD interaction parameters used to compute LLE are located below each ternary plot. Numbers in red represent the solute/solvent parameters obtained for each approach.

3. Liquid-liquid equilibrium ternary diagram for water/benzene/2-propanol system at 298.15K.

Parametrization approaches #1, #3 and #4 were carried out with the fourth composition the less concentrated in 2-propanol. Approach #2 is based on the five compositions less concentrated in 2-propanol.

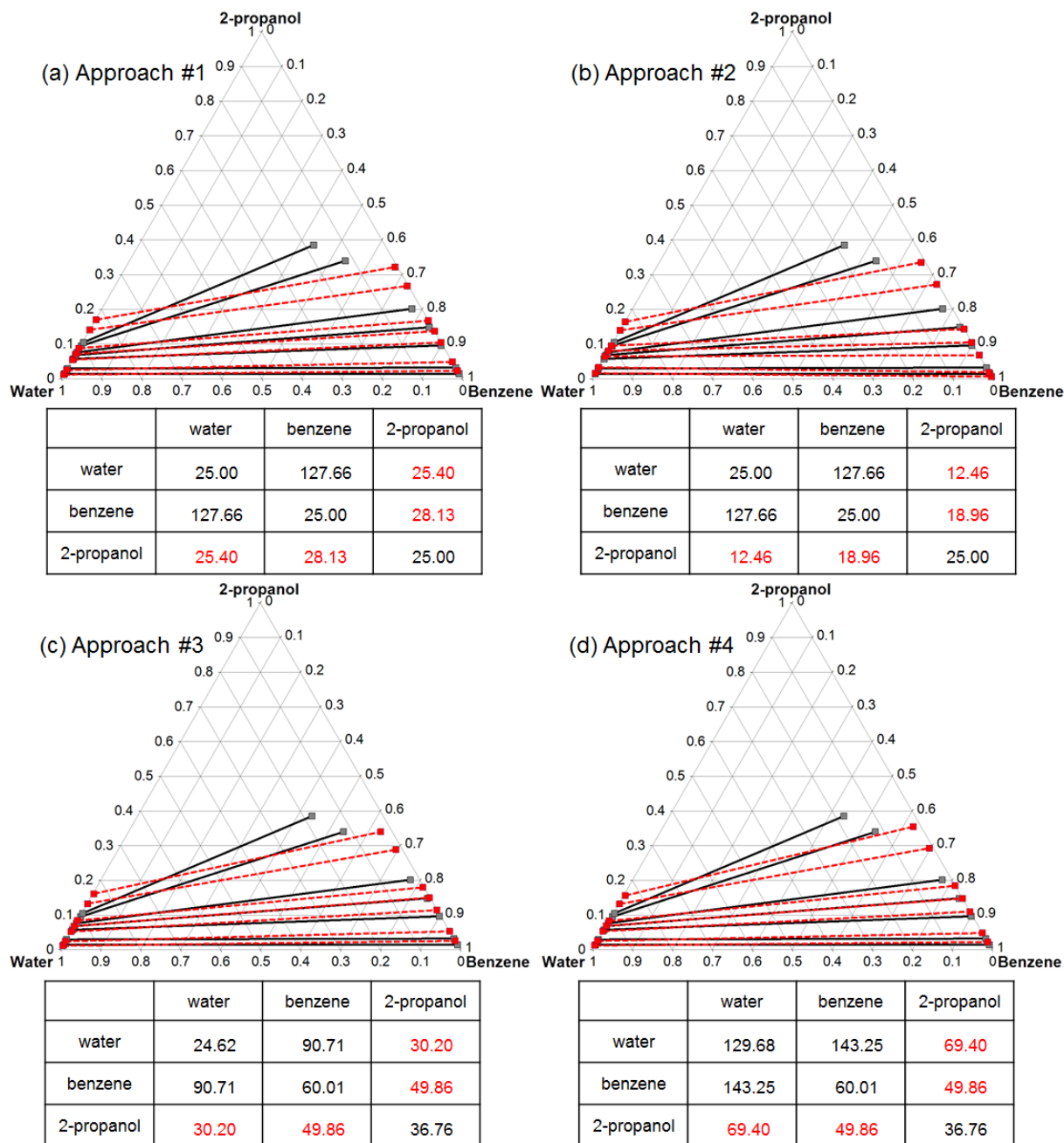


Figure S10. Liquid-liquid equilibrium ternary diagrams for the water/benzene/2-propanol system at 298.15K for (a) approach #1, (b) approach #2, (3) approach #3 and (4) approach #4. Experimental data are plotted in black and the results from the CG-MC simulations are in red. DPD interaction parameters used to compute LLE are located below each ternary plot. Numbers in red represent the solute/solvent parameters obtained for each approach.

4. Liquid-liquid equilibrium ternary diagram for water/hexane/acetone system at 298.15K.

Transferability approach and approaches #3 and #4 were carried out with the fifth composition the less concentrated in acetone. Approach #2 is based on the eleven compositions. Note that approach #1 provides similar interaction parameters than approach #2 regardless of the composition used.

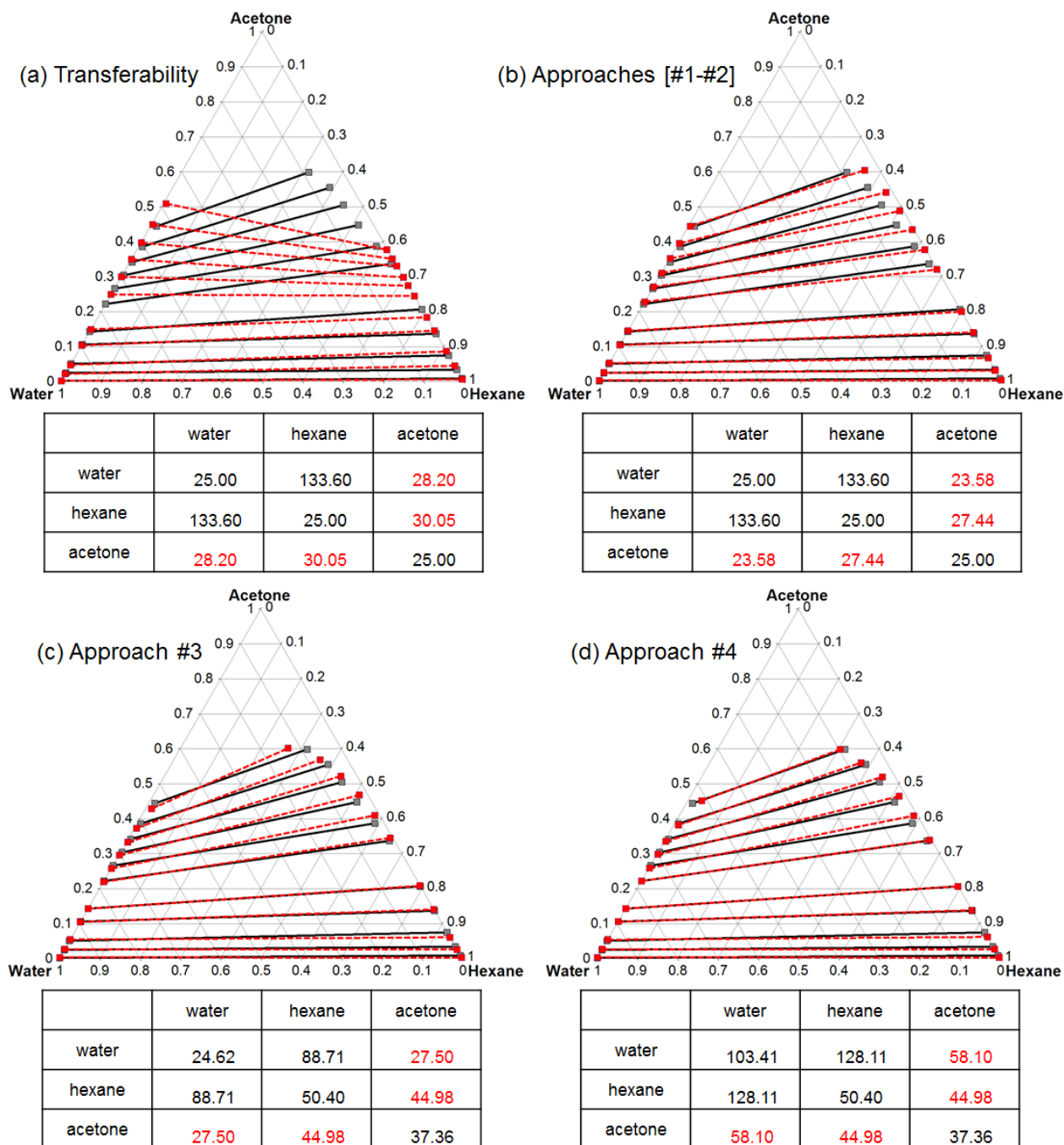


Figure S11. Liquid-liquid equilibrium ternary diagrams for the water/hexane/acetone system at 298.15K for (a) approach #1, (b) approach #2, (3) approach #3 and (4) approach #4. Experimental data are plotted in black and the results from the CG-MC simulations are in red. DPD interaction parameters used to compute LLE are located below each ternary plot. Numbers in red represent the solute/solvent parameters obtained for each approach.

5. Liquid-liquid equilibrium ternary diagram for water/hexane/2-propanol system at 298.15K.

Parametrization approaches #1, #3 and #4 were carried out with the seventh composition the less concentrated in 2-propanol. Approach #2 is based on the ten compositions less concentrated in 2-propanol.

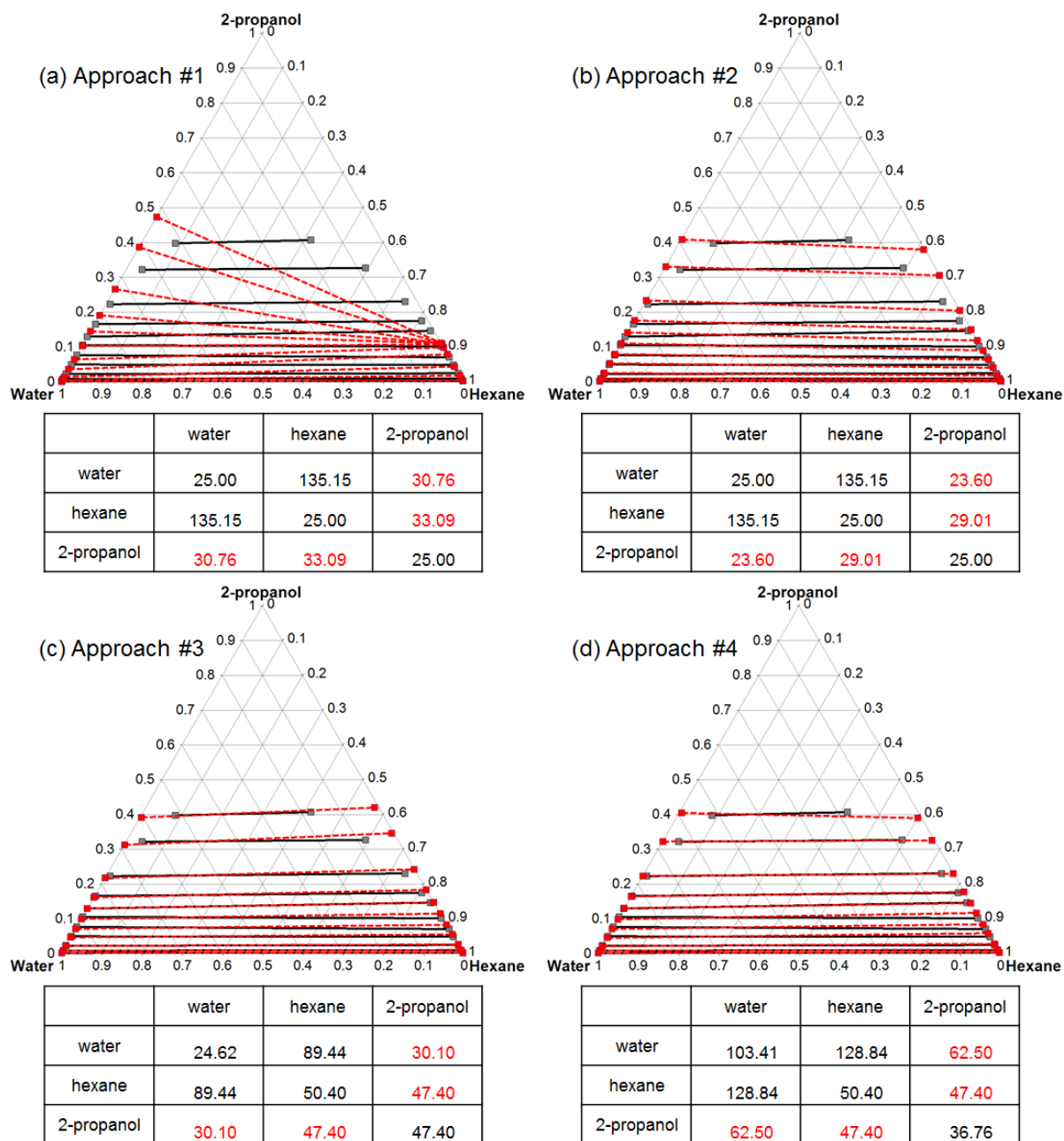


Figure S12. Liquid-liquid equilibrium ternary diagrams for the water/hexane/2-propanol system at 298.15K for (a) approach #1, (b) approach #2, (c) approach #3 and (d) approach #4. Experimental data are plotted in black and the results from the CG-MC simulations are in red. DPD interaction parameters used to compute LLE are located below each ternary plot. Numbers in red represent the solute/solvent parameters obtained for each approach.

S7. Coarse-grained compositions of the systems studied for the CG-MC simulations in the Gibbs ensemble (NVT).

These compositions were calculated from experimental compositional data of liquid-liquid equilibrium and correspond to a heterogeneous system that decants following the conodal lines. The total number of beads for each system is set at 6000.

Table S7. Compositions used in CG-MC simulations in the Gibbs (NVT) ensemble for the water/benzene/1,4-dioxane system.

Number of water beads	Number of benzene beads	Number of 1,4-dioxane beads
2411	3014	575
2141	2677	1182
1898	2372	1730
1632	2041	2327
1242	1553	3205

Table S8. Compositions used in CG-MC simulations in the Gibbs (NVT) ensemble for the water/chloroform/acetone system.

Number of water beads	Number of chloroform beads	Number of acetone beads
3236	2284	480
2979	2103	918
2511	1772	1717
2088	1474	2438
1705	1203	3092

Table S9. Compositions used in CG-MC simulations in the Gibbs (NVT) ensemble for the water/benzene/acetic acid system.

Number of water beads	Number of benzene beads	Number of acetic acid beads
2223	3168	609
1896	2922	1182
1536	2652	1812
1171	2378	2451
845	2133	3022

Table S10. Compositions used in CG-MC simulations in the Gibbs (NVT) ensemble for the water/benzene/2-propanol system.

Number of water beads	Number of benzene beads	Number of 2-propanol beads
2564	3204	232
2455	3069	476
2233	2791	976
2116	2645	1239
2011	2513	1476
1700	2125	2175
1551	1938	2511

Table S11. Compositions used in CG-MC simulations in the Gibbs (NVT) ensemble for the water/hexane/acetone system.

Number of water beads	Number of hexane beads*	Number of acetone beads
1988	3978	34
1912	3824	264
1821	3640	539
1651	3304	1045
1527	3054	1419
1289	2578	2133
1178	2358	2464
1076	2152	2772
978	1958	3064
881	1762	3357
771	1542	3687

* Hexane molecules are represented by two beads

Table S12. Compositions used in CG-MC simulations in the Gibbs (NVT) ensemble for the water/hexane/2-propanol system.

Number of water beads	Number of hexane beads	Number of 2-propanol beads
1351	4632	17
1338	4590	72
1314	4504	182
1269	4352	379
1226	4204	570
1177	4036	787
1127	3866	1007
1074	3680	1246
985	3376	1639
838	2872	2290
728	2494	2778

* Hexane molecules are represented by two beads

S8. Chemical potentials of the solute and pressures of the system.

These data are calculated at equilibrium using CG-MC simulation in the Gibbs ensemble (NVT) following the four parametrization approaches.

Table S13. Chemical potentials of 1,4-dioxane and pressures for the water/benzene/1,4-dioxane system. P = pressure

#1 approach		#2 approach		#3 approach		#4 approach	
$\mu_{1,4-dioxane}$ (DPD unit)	P (DPD unit)	$\mu_{1,4-dioxane}$ (DPD unit)	P (DPD unit)	$\mu_{1,4-dioxane}$ (DPD unit)	P (DPD unit)	$\mu_{1,4-dioxane}$ (DPD unit)	P (DPD unit)
11.13	23.74	12.86	24.48	24.18	40.50	37.41	76.85
11.83	23.80	13.37	25.05	25.84	43.04	37.50	75.64
12.22	23.86	13.56	25.39	27.16	45.43	37.46	74.56
12.48	23.91	13.67	25.58	28.45	48.14	37.34	73.44
12.77	24.02	13.73	25.70	30.30	52.36	37.09	71.94

Table S14. Chemical potentials of acetone and pressures for the water/chloroform/acetone system. P = pressure

#1 approach		#2 approach		#3 approach		#4 approach	
$\mu_{acetone}$ (DPD unit)	P (DPD unit)	$\mu_{acetone}$ (DPD unit)	P (DPD unit)	$\mu_{acetone}$ (DPD unit)	P (DPD unit)	$\mu_{acetone}$ (DPD unit)	P (DPD unit)
11.40	23.79	11.49	23.42	16.26	31.66	26.37	67.15
12.02	23.89	11.19	23.31	16.93	32.02	26.20	64.12
12.59	24.08	11.92	23.27	17.63	32.65	25.32	58.76
12.88	24.25	12.34	23.40	18.03	33.21	24.27	54.18
13.07	24.41	12.62	23.60	18.33	33.73	23.37	50.31

Table S15. Chemical potentials of acetic acid and pressures for the water/benzene/acetic acid system. P = pressure

#1 approach		#2 approach		#3 approach		#4 approach	
$\mu_{acetic\ acid}$ (DPD unit)	P (DPD unit)	$\mu_{acetic\ acid}$ (DPD unit)	P (DPD unit)	$\mu_{acetic\ acid}$ (DPD unit)	P (DPD unit)	$\mu_{acetic\ acid}$ (DPD unit)	P (DPD unit)
5.73	21.09	10.64	23.22	16.25	37.40	24.07	62.82
7.66	19.65	11.48	23.05	17.65	36.94	24.12	58.06
9.21	19.24	12.07	23.09	18.71	37.13	23.84	53.78
10.34	19.71	12.52	23.35	19.52	37.83	23.37	50.29
11.17	20.63	12.88	23.78	20.14	38.74	22.88	47.69

Table S16. Chemical potentials of 2-propanol and pressures for the water/benzene/2-propanol system. P = pressure

#1 approach		#2 approach		#3 approach		#4 approach	
$\mu_{2\text{-propanol}}$ (DPD unit)	P (DPD unit)	$\mu_{2\text{-propanol}}$ (DPD unit)	P (DPD unit)	$\mu_{2\text{-propanol}}$ (DPD unit)	P (DPD unit)	$\mu_{2\text{-propanol}}$ (DPD unit)	P (DPD unit)
10.57	23.74	5.69	22.87	17.29	38.05	27.54	75.78
11.27	23.81	6.62	22.16	17.92	37.92	27.69	73.50
11.97	23.95	7.72	20.99	18.50	37.63	27.19	69.00
12.20	24.02	8.14	20.52	18.65	37.48	26.90	66.72
12.36	24.08	8.47	20.16	18.73	37.33	26.54	64.73
12.70	24.22	9.30	19.46	18.94	36.94	25.39	59.14
12.82	24.29	9.66	19.32	18.95	36.75	24.79	56.60

Table S17. Chemical potentials of acetone and pressures for the water/hexane/acetone system. P = pressure

<i>Transferability</i>		[#1-#2] approach		#3 approach		#4 approach	
μ_{acetone} (DPD unit)	P (DPD unit)	μ_{acetone} (DPD unit)	P (DPD unit)	μ_{acetone} (DPD unit)	P (DPD unit)	μ_{acetone} (DPD unit)	P (DPD unit)
9.48	22.67	8.18	22.64	14.03	35.90	20.48	57.59
11.44	22.94	10.17	22.64	16.10	35.66	22.26	56.25
12.06	23.20	10.90	22.67	16.85	35.42	22.69	54.75
12.59	23.58	11.59	22.76	17.55	35.12	22.75	52.22
12.80	23.78	11.91	22.84	17.88	34.96	22.63	50.48
13.04	24.05	12.35	23.02	18.27	34.76	22.20	47.44
13.12	24.13	12.49	23.12	18.40	34.71	21.96	46.14
13.17	24.19	12.61	23.22	18.48	34.67	21.70	44.99
13.22	24.23	12.72	23.31	18.57	34.67	21.51	43.93
13.26	24.27	12.81	23.42	18.65	34.67	21.26	42.99
13.29	24.32	12.90	23.55	18.71	34.72	20.98	41.92

Table S18. Chemical potentials of 2-propanol and pressures for the water/hexane/2-propanol system. P = pressure

#1 approach		#2 approach		#3 approach		#4 approach	
$\mu_{2\text{-propanol}}$ (DPD unit)	P (DPD unit)	$\mu_{2\text{-propanol}}$ (DPD unit)	P (DPD unit)	$\mu_{2\text{-propanol}}$ (DPD unit)	P (DPD unit)	$\mu_{2\text{-propanol}}$ (DPD unit)	P (DPD unit)
10.39	22.81	7.82	22.76	14.09	34.16	22.33	61.40
11.73	22.99	9.21	22.75	15.47	34.18	23.57	60.89
12.51	23.29	10.16	22.74	16.40	34.25	24.15	59.91
13.01	23.71	10.90	22.74	17.13	34.36	24.38	58.19
13.20	23.98	11.32	22.76	17.54	34.45	24.29	56.61
13.30	24.18	11.63	22.79	17.81	34.55	24.08	55.02
13.36	24.31	11.86	22.83	18.03	34.64	23.87	53.62
13.41	24.42	12.07	22.88	18.23	34.75	23.53	52.02
13.44	24.52	12.33	22.99	18.46	34.91	23.05	49.71
13.46	24.57	12.64	23.19	18.73	35.15	22.26	46.41
13.46	24.56	12.81	23.37	18.89	35.33	21.73	44.38

S9. Coarse-grained compositions of the systems studied using the CG-MC simulations in the osmotic ensemble ($\mu_{\text{solute}}N_{\text{solvent}}P_{zz}T$).

Compositions at equilibrium obtained using CG-MC simulations in the osmotic ensemble. These compositions are used in DPD simulations to calculate IFT.

Table S19. Compositions used in DPD simulations for the water/benzene/1,4-dioxane system and for the four parametrization approaches.

		#1 approach	#2 approach	#3 approach	#4 approach
Water beads	Benzene beads	1,4-dioxane beads	1,4-dioxane beads	1,4-dioxane beads	1,4-dioxane beads
7234	9042	1964	2186	2195	2700
6424	8029	3818	4479	4106	4312
5693	7116	5668	6054	6082	6045
4898	6122	7261	7817	7850	8107
3727	4659	9566	8416	11599	10873

Table S20. Compositions used in DPD simulations for the water/chloroform/acetone system and for the four parametrization approaches.

		#1 approach	#2 approach	#3 approach	#4 approach
Water beads	Chloroform beads	Acetone beads	Acetone beads	Acetone beads	Acetone beads
9707	6852	1668	1572	1629	1654
8938	6309	3061	2978	2979	2998
7533	5317	5546	5410	5541	5526
6264	4421	7662	7745	7651	6949
5114	3610	9958	9898	9947	9408

Table S21. Compositions used in DPD simulations for the water/benzene/acetic acid system and for the four parametrization approaches.

		#1 approach	#2 approach	#3 approach	#4 approach
Water beads	Benzene beads	Acetic acid beads	Acetic acid beads	Acetic acid beads	Acetic acid beads
6670	9502	1862	1952	1945	1921
5687	8766	3646	3709	3727	3659
4607	7955	5580	5734	5666	5695
3512	7134	7548	7619	7629	7492
2534	6401	9049	9382	9378	8665

Table S22. Compositions used in DPD simulations for the water/benzene/2-propanol system and for the four parametrization approaches.

		#1 approach	#2 approach	#3 approach	#4 approach
Water beads	Benzene beads	2-propanol beads	2-propanol beads	2-propanol beads	2-propanol beads
7691	9613	881	740	869	877
7366	9208	1667	1512	1676	1688
6699	8374	3243	3042	3231	3017
6348	7936	4121	3847	4005	4134
6032	7541	4876	4560	4628	4822
5100	6374	6997	6653	7064	6965
4652	5815	7867	7668	7527	7635

Table S23. Compositions used in DPD simulations for the water/hexane/acetone system and for the four parametrization approaches.

		<i>Transferability</i>	[#1-#2] approach	#3 approach	#4 approach
Water beads	Hexane beads*	Acetone beads	Acetone beads	Acetone beads	Acetone beads
5966	11932	177	163	150	132
5737	11474	1051	976	954	912
5461	10922	1953	1845	1831	1847
4955	9910	3666	3422	3347	3316
4582	9164	4843	4543	4546	4609
3868	7736	6921	6772	6770	6784
3536	7072	7908	7823	7736	7742
3228	6456	8450	8633	8529	8151
2936	5872	9710	9551	9384	9831
2644	5288	10691	10188	10503	10053
2313	4626	10398	11191	10939	10654

* Hexane molecules are represented by two beads.

Table S24. Compositions used in DPD simulations for the water/hexane/2-propanol system and for the four parametrization approaches.

		#1 approach	#2 approach	#3 approach	#4 approach
Water beads	Hexane beads*	2-propanol beads	2-propanol beads	2-propanol beads	2-propanol beads
7426	10498	157	112	116	114
7306	10376	555	403	419	417
7069	10136	1243	952	950	954
6646	9704	2349	1836	1824	1897
6253	9304	3614	2683	2707	2757
5841	8884	Phase separation	3484	3476	3610
5476	8516		4369	4260	4426
5047	8078		5223	5200	4970
4405	7424		6507	6385	6354
3446	6448		8507	8291	8241
2809	5802		9534	9355	9149

* Hexane molecules are represented by two beads.

S10. Interfacial concentration in solute and thickness of the interface.

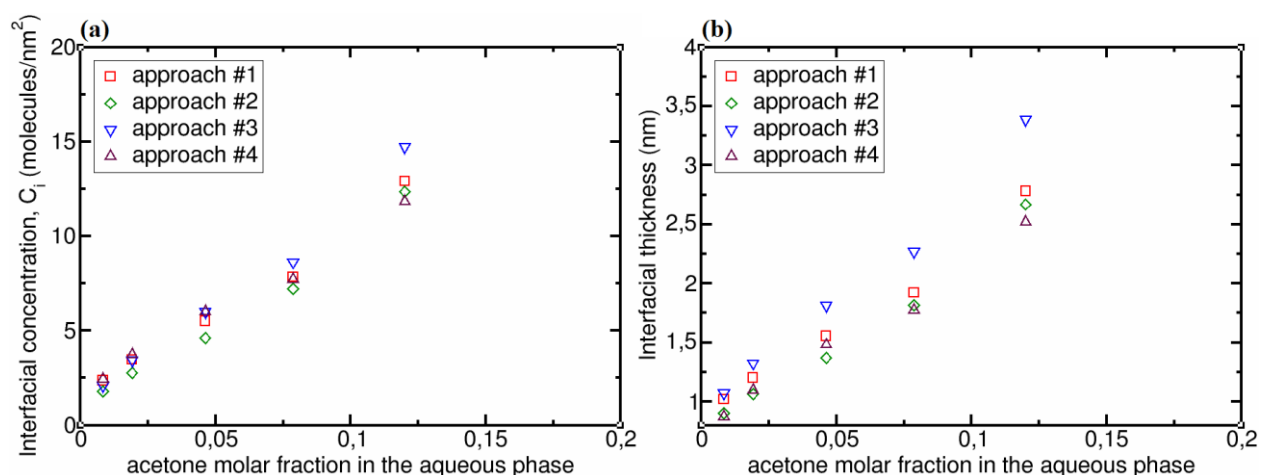


Figure S13. Water/chloroform/acetone system at 298.15K (a) Variation of the acetone interfacial concentration, as a function of the molar fraction of acetone in the aqueous phase. (b) Variation of the water/chloroform interface thickness, as a function of the molar fraction of acetone in the aqueous phase.

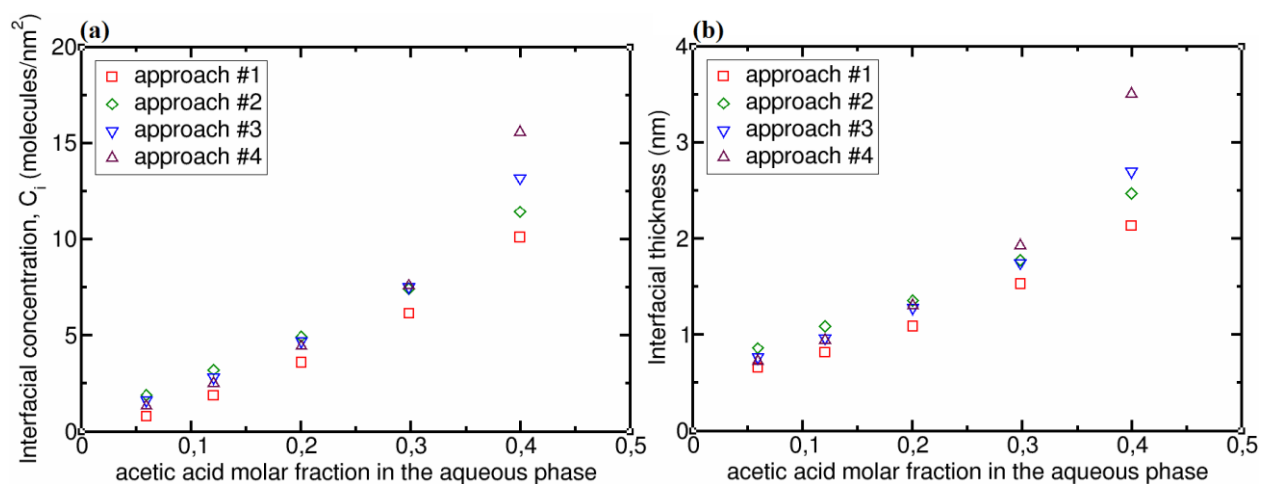


Figure S14. Water/benzene/acetic acid system at 298.15K (a) Variation of the acetic acid interfacial concentration, as a function of the molar fraction of acetic acid in the aqueous phase. (b) Variation of the water/benzene interface thickness, as a function of the molar fraction of acetic acid in the aqueous phase.

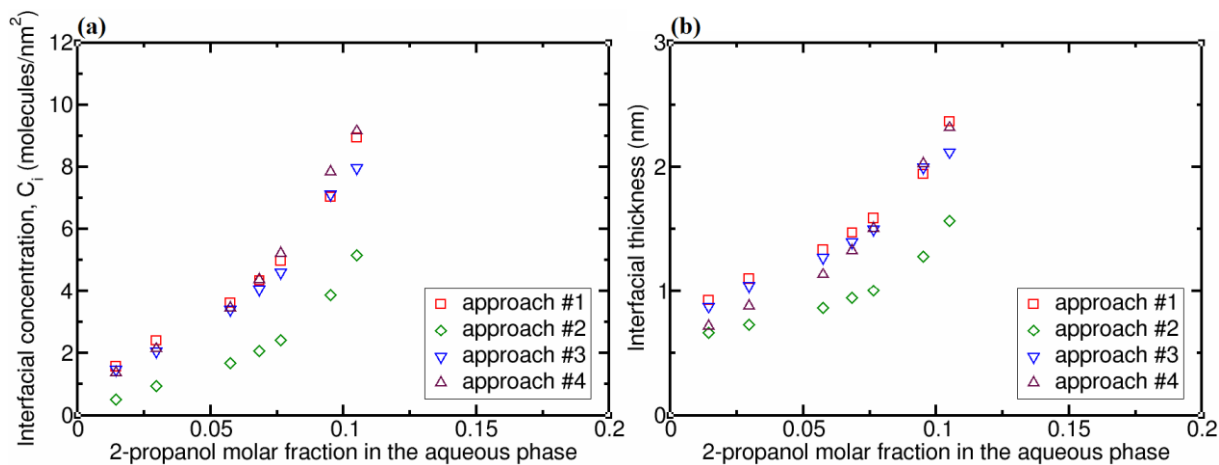


Figure S15. Water/benzene/2-propanol system at 298.15K (a) Variation of the 2-propanol interfacial concentration, as a function of the molar fraction of 2-propanol in the aqueous phase. (b) Variation of the water/benzene interface thickness, as a function of the molar fraction of 2-propanol in the aqueous phase.

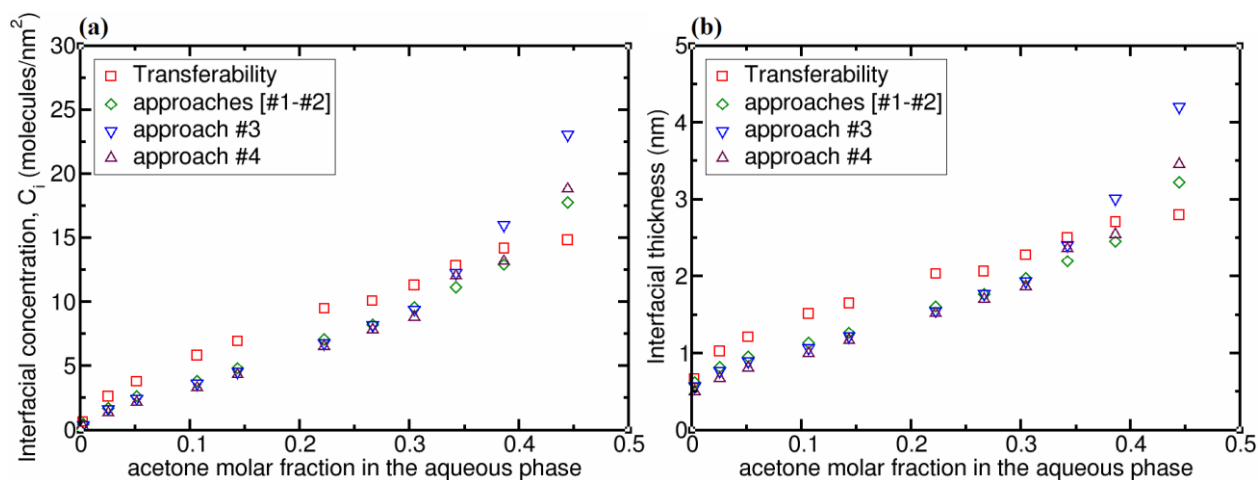


Figure S16. Water/hexane/acetone system at 298.15K (a) Variation of the acetone interfacial concentration, as a function of the molar fraction of acetone in the aqueous phase. (b) Variation of the water/hexane interface thickness, as a function of the molar fraction of acetone in the aqueous phase.

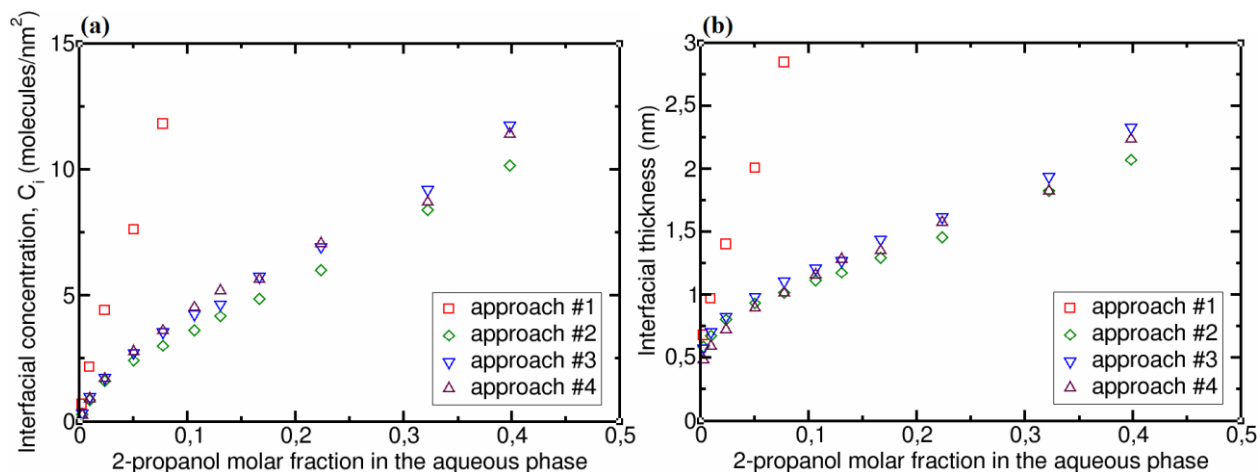


Figure S17. Water/hexane/2-propanol system at 298.15K (a) Variation of the 2-propanol interfacial concentration, as a function of the molar fraction of 2-propanol in the aqueous phase. (b) Variation of the water/hexane interface thickness, as a function of the molar fraction of 2-propanol in the aqueous phase.

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