Supporting Information for: Optimizing Non-bonded Interactions of the OPLS Force Field for Aqueous Solutions of Carbohydrates: How to Capture Both Thermodynamics and Dynamics

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S1 OPLS force field parameters

In force field-based molecular simulation, the interactions between atoms are due to non-bonded and bonded interaction potentials:[1]

$$U = U_{\text{non-bonded}} + U_{\text{bonded}} \tag{S1}$$

The non-bonded interactions between two atoms i and j, separated by a distance r_{ij}, consist of two parts:

$$U_{\text{non-bonded}}\left(r_{ij}\right) = U_{\text{LJ}}\left(r_{ij}\right) + U_{\text{Coulomb}}\left(r_{ij}\right) = 4\epsilon\left[\left(\frac{\sigma}{r_{ij}}\right)^{12} - \left(\frac{\sigma}{r_{ij}}\right)^{6}\right] + \frac{q_i q_j}{4\pi\varepsilon_0 r_{ij}} \tag{S2}$$

in which q_i is the partial atomic charge of atom i, ε_0 is the permittivity of vacuum, and ϵ and σ are the Lennard-Jones parameters. The interaction parameters of dissimilar atoms are calculated from the standard OPLS force field mixing rules:[2,3]

$$\begin{aligned} \sigma_{ij} &= \sqrt{\sigma_{ii} \cdot \sigma_{jj}} \\ \epsilon_{ij} &= \sqrt{\epsilon_{ii} \cdot \epsilon_{jj}} \end{aligned} \tag{S3}$$

In this study, the Lennard-Jones (LJ) parameters and partial atomic charges of the OPLS force field for carbohydrates[3] are rescaled so that thermodynamic and transport properties of water-sucrose mixtures are reproduced. The optimum scaling factors for the LJ energy parameter (ϵ) and partial atomic charges (q) are 0.80 and 0.95, respectively. The interaction sites for sucrose and glucose are shown in Figure S1. The non-bonded interaction parameters of the original and refined OPLS force fields for sucrose and glucose molecules are listed in Tables S1 and S2, respectively.

The bonded contribution to the total energy consists of three parts due to bond-stretching,

bond-angle bending, and dihedral angle torsion:[2,3]

$$U_{\text{bonded}} = U_{\text{bond}} + U_{\text{angle}} + U_{\text{torsion}} \tag{S4}$$

These terms are defined as follows:[2,3]

$$U_{\text{bond}} = \sum_{\text{bonds}} K_b(r - r_0)^2 \tag{S5}$$

$$U_{\text{angle}} = \sum_{\text{angles}} K_\theta(\theta - \theta_0)^2 \tag{S6}$$

$$U_{\text{torsion}} = \sum_{\text{dihedrals}} \left[\frac{V_1}{2}(1 + \cos\phi) + \frac{V_2}{2}(1 - \cos 2\phi) + \frac{V_3}{2}(1 + \cos 3\phi) \right] \tag{S7}$$

where r, θ, ϕ are the bond lengths, bond angles, and dihedral angles in a configuration of the molecule. r_0 and θ_0 are the equilibrium bond lengths and bond angles, respectively. K_r, K_θ, V_i are the coefficients of these bonded potentials. For sucrose and glucose, the bonded parameters are provided in Tables S3 to S5 and Tables S6 to S8, respectively.

S2 Results

Thermodynamic and transport properties of water-sucrose and water-glucose mixtures were computed based on the scaled non-bonded parameters of the OPLS force field. Five mass fractions of sucrose-water (w_{sucrose} = 20%, 30%, 40%, 50%, and 60%) and four mass fractions of glucose-water (w_{glucose} = 20%, 30%, 40%, and 50%) were considered. Temperature and pressure in all MD simulations are 298 K and 1 atm, respectively. Computed thermodynamic factors and shear viscosities used for optimizing the OPLS[2,3] force field parameters and comparing the original[4] and modified[5] GLYCAM06 force fields are reported in Tables S9 and S10. These computed properties are shown in Figures 2 to 4 of the main text. The computed liquid densities, thermodynamic factors, self-diffusivities of water, self-diffusivities of sucrose, and mutual diffusion coefficients of water-sucrose mixtures are reported in Tables S11 to S16, respectively. Similarly, these properties are listed in Tables S17 to S22 for water-glucose mixtures. For both mixtures, the computed properties are shown in Figures 5 and 6 of the main text.

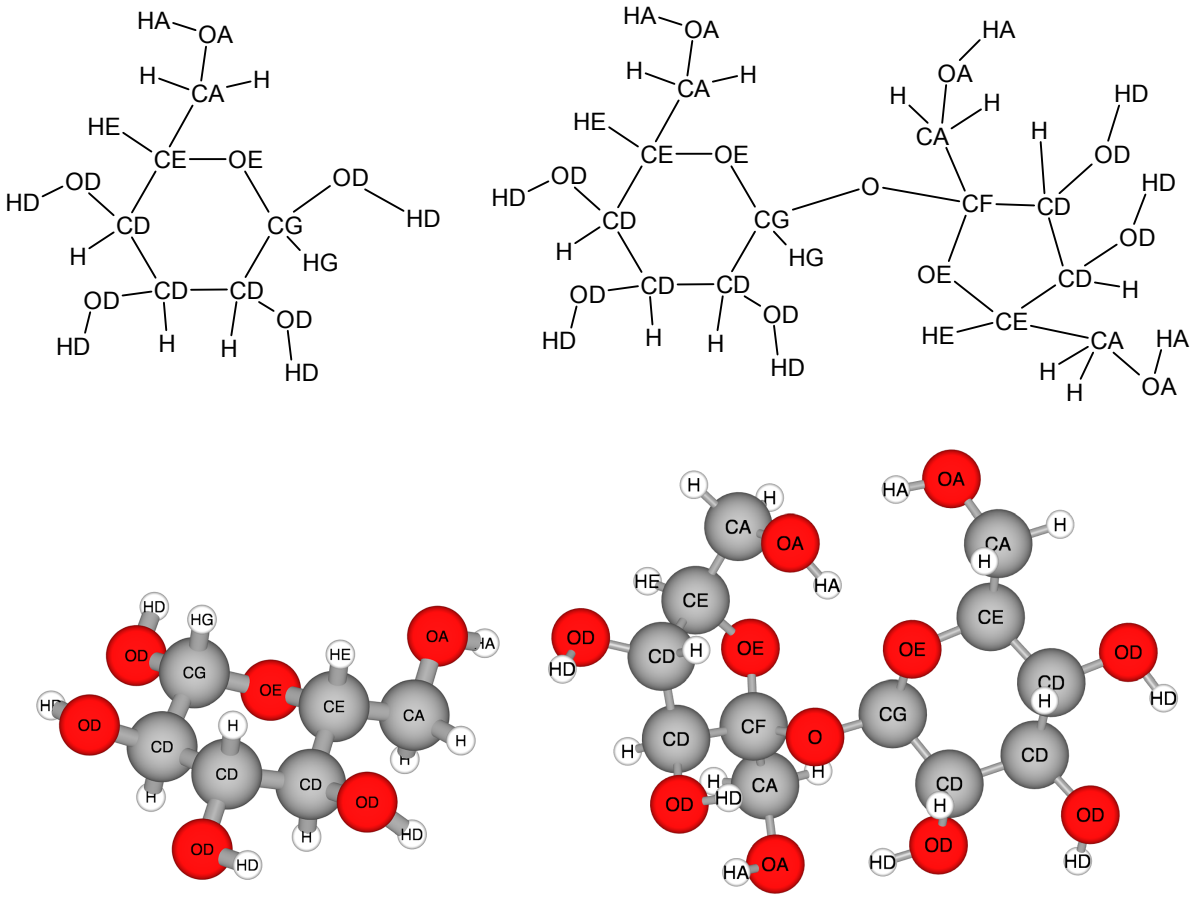

Figure S1: Schematic and atomistic representations of glucose and sucrose with the presentation of all interaction sites. The atomistic representation was created by using iRASPA.[6] The non-bonded and bonded interaction parameters for both molecules are listed in Tables S1 and S2, and Tables S3 to S8, respectively.

Table S1: Non-bonded interaction parameters (Equation (S2)) of the original and refined OPLS[3] force fields for sucrose. LJ energy parameters and partial atomic charges are scaled with factors of 0.8 and 0.95, respectively.

	original OPLS			refined OPLS		
Atom	ϵ (kcal/mol)	σ (Å)	q (e)	ϵ (kcal/mol)	σ (Å)	q (e)
CA	0.066	3.5	0.145	0.0528	3.5	0.13775
CD	0.066	3.5	0.205	0.0528	3.5	0.19475
CE	0.066	3.5	0.17	0.0528	3.5	0.1615
CF	0.066	3.5	0.4	0.0528	3.5	0.38
CG	0.066	3.5	0.3	0.0528	3.5	0.285
H	0.03	2.5	0.06	0.024	2.5	0.057
HA	0	0	0.418	0	0	0.3971
HD	0	0	0.435	0	0	0.41325
HE	0.03	2.5	0.03	0.024	2.5	0.0285
HG	0.03	2.5	0.1	0.024	2.5	0.095
O	0.14	2.9	-0.4	0.112	2.9	-0.38
OA	0.17	3.12	-0.683	0.136	3.12	-0.64885
OD	0.17	3.07	-0.7	0.136	3.07	-0.665
OE	0.14	2.9	-0.4	0.112	2.9	-0.38

Table S2: Non-bonded interaction parameters (Equation (S2)) of the original and refined OPLS[3] force fields for glucose. LJ energy parameters and partial atomic charges are scaled with factors of 0.8 and 0.95, respectively.

	original OPLS			refined OPLS		
Atom	ϵ (kcal/mol)	σ (Å)	q (e)	ϵ (kcal/mol)	σ (Å)	q (e)
CA	0.066	3.5	0.145	0.0528	3.5	0.13775
CD	0.066	3.5	0.205	0.0528	3.5	0.19475
CE	0.066	3.5	0.17	0.0528	3.5	0.1615
CG	0.066	3.5	0.365	0.0528	3.5	0.34675
H	0.03	2.5	0.06	0.024	2.5	0.057
HA	0	0	0.418	0	0	0.3971
HD	0	0	0.435	0	0	0.41325
HE	0.03	2.5	0.03	0.024	2.5	0.0285
HG	0.03	2.5	0.1	0.024	2.5	0.095
OA	0.17	3.12	-0.683	0.136	3.12	-0.64885
OD	0.17	3.07	-0.7	0.136	3.07	-0.665
OE	0.14	2.9	-0.4	0.112	2.9	-0.38

Table S3: bond-stretching parameters (Equation (S5)) of the OPLS force field for sucrose.[3] Interaction sites for sucrose are shown in Figure S1.

Bond	K_b (kcal/mol/Å^2)	r_0 (Å)
CA-CE	268	1.529
CA-CF	268	1.529
CA-H	340	1.09
CA-OA	320	1.41
CD-CD	268	1.529
CD-CE	268	1.529
CD-CF	268	1.529
CD-CG	268	1.529
CD-H	340	1.09
CD-OD	320	1.41
CE-HE	340	1.09
CE-OE	320	1.41
CF-O	320	1.38
CF-OE	320	1.38
CG-HG	340	1.09
CG-O	320	1.38
CG-OE	320	1.38
HA-OA	553	0.945
HD-OD	553	0.945

Table S4: bond-angle bending parameters (Equation (S6)) of the OPLS force field for sucrose.[3] Interaction sites for sucrose are shown in Figure S1.

Bond	K_θ (kcal/mol/rad^2)	θ_0 (°)
CA-CE-CD	58.35	112.7
CA-CE-HE	37.5	110.7
CA-CE-OE	50	109.5
CA-CF-CD	58.35	112.7
CA-CF-O	50	109.5
CA-CF-OE	50	109.5
CA-OA-HA	55	108.5
CD-CD-CD	58.35	112.7
CD-CD-CE	58.35	112.7
CD-CD-CF	58.35	112.7
CD-CD-CG	58.35	112.7
CD-CD-H	37.5	110.7
CD-CD-OD	50	109.5
CD-CE-HE	37.5	110.7
CD-CE-OE	50	109.5
CD-CF-O	50	109.5
CD-CF-OE	50	109.5
CD-CG-HG	37.5	110.7
CD-CG-O	50	109.5
CD-CG-OE	50	109.5
CD-OD-HD	55	108.5
CE-CA-H	37.5	110.7
CE-CA-OA	50	109.5
CE-CD-H	37.5	110.7
CE-CD-OD	50	109.5
CE-OE-CF	60	109.5
CE-OE-CG	60	109.5
CF-CA-H	37.5	110.7
CF-CA-OA	50	109.5
CF-CD-H	37.5	110.7
CF-CD-OD	50	109.5
CF-O-CG	60	109.5
CG-CD-H	37.5	110.7
CG-CD-OD	50	109.5
H-CA-H	33	107.8
H-CA-OA	35	109.5
H-CD-OD	35	109.5
HE-CE-OE	35	109.5
HG-CG-O	35	109.5
HG-CG-OE	35	109.5
O-CF-OE	92.6	111.55
O-CG-OE	92.6	111.55

Table S5: Dihedral angle torsion parameters (Equation (S7)) of the OPLS force field for sucrose.[3] All V_1, V_2, and V_3 coefficients are reported in units of kcal/mol. Interaction sites for sucrose are shown in Figure S1.

Dihedral angle	V_1	V_2	V_3	Dihedral angle	V_1	V_2	V_3
CA-CE-OE-CF	0.65	-0.25	0.67	H-CA-CF-O	0	0	0.468
CA-CE-OE-CG	0.65	-0.25	0.67	H-CA-CF-OE	0	0	0.468
CA-CF-O-CG	0.65	-0.25	0.67	H-CA-OA-HA	0	0	0.45
CA-CF-OE-CE	0.65	-0.25	0.67	H-CD-CD-H	0	0	0.318
CD-CD-CD-CE	1.74	-0.157	0.279	H-CD-CD-OD	0	0	0.468
CD-CD-CD-CG	1.74	-0.157	0.279	H-CD-CE-CA	0	0	0.366
CD-CD-CD-H	0	0	0.366	H-CD-CE-HE	0	0	0.318
CD-CD-CD-OD	-1.336	0	0	H-CD-CE-OE	0	0	0.468
CD-CD-CE-CA	1.74	-0.157	0.279	H-CD-CF-CA	0	0	0.366
CD-CD-CE-HE	0	0	0.366	H-CD-CF-O	0	0	0.468
CD-CD-CE-OE	-1.336	0	0	H-CD-CF-OE	0	0	0.468
CD-CD-CF-CA	1.74	-0.157	0.279	H-CD-CG-HG	0	0	0.318
CD-CD-CF-O	-1.336	0	0	H-CD-CG-O	0	0	0.468
CD-CD-CF-OE	-1.336	0	0	H-CD-CG-OE	0	0	0.468
CD-CD-CG-HG	0	0	0.366	H-CD-OD-HD	0	0	0.45
CD-CD-CG-O	-1.336	0	0	HE-CE-OE-CF	0	0	0.76
CD-CD-CG-OE	-1.336	0	0	HE-CE-OE-CG	0	0	0.76
CD-CD-OD-HD	2.674	-2.883	1.026	HG-CG-O-CF	0	0	0.76
CD-CE-OE-CF	0.65	-0.25	0.67	HG-CG-OE-CE	0	0	0.76
CD-CE-OE-CG	0.65	-0.25	0.67	O-CF-OE-CE	-0.375	-1.358	0.004
CD-CF-O-CG	0.65	-0.25	0.67	O-CG-OE-CE	-0.375	-1.358	0.004
CD-CF-OE-CE	0.65	-0.25	0.67	OA-CA-CE-CD	-1.336	0	0
CD-CG-O-CF	0.65	-0.25	0.67	OA-CA-CE-HE	0	0	0.468
CD-CG-OE-CE	0.65	-0.25	0.67	OA-CA-CE-OE	4.319	0	0
CE-CA-OA-HA	2.674	-2.883	1.026	OA-CA-CF-CD	-1.336	0	0
CE-CD-CD-CF	1.74	-0.157	0.279	OA-CA-CF-O	4.319	0	0
CE-CD-CD-H	0	0	0.366	OA-CA-CF-OE	4.319	0	0
CE-CD-CD-OD	-1.336	0	0	OD-CD-CD-OD	9.066	0	0
CE-CD-OD-HD	2.674	-2.883	1.026	OD-CD-CE-CA	-1.336	0	0
CF-CA-OA-HA	2.674	-2.883	1.026	OD-CD-CE-HE	0	0	0.468
CF-CD-CD-H	0	0	0.366	OD-CD-CE-OE	4.319	0	0
CF-CD-CD-OD	-1.336	0	0	OD-CD-CF-CA	-1.336	0	0
CF-CD-OD-HD	2.674	-2.883	1.026	OD-CD-CF-O	4.319	0	0
CG-CD-CD-H	0	0	0.366	OD-CD-CF-OE	4.319	0	0
CA-CE-OE-CF	-1.336	0	0	OD-CD-CG-HG	0	0	0.468
CG-CD-OD-HD	2.674	-2.883	1.026	OD-CD-CG-O	4.319	0	0
H-CA-CE-CD	0	0	0.366	OD-CD-CG-OE	4.319	0	0
H-CA-CE-HE	0	0	0.318	OE-CF-O-CG	-0.375	-1.358	0.004
H-CA-CE-OE	0	0	0.468	OE-CG-O-CF	-0.375	-1.358	0.004
H-CA-CF-CD	0	0	0.366				

Table S6: bond-stretching parameters (Equation (S5)) of the OPLS force field for glucose.[3] Interaction sites for glucose are shown in Figure S1.

Bond	K_b (kcal/mol/Å^2)	r_0 (Å)
CA-CE	268	1.529
CA-H	340	1.09
CA-OA	320	1.41
CD-CD	268	1.529
CD-CE	268	1.529
CD-CG	268	1.529
CD-H	340	1.09
CD-OD	320	1.41
CE-HE	340	1.09
CE-OE	320	1.41
CG-HG	340	1.09
CG-OD	320	1.38
CG-OE	320	1.38
HA-OA	553	0.945
HD-OD	553	0.945

Table S7: bond-angle bending parameters (Equation (S6)) of the OPLS force field for glucose.[3] Interaction sites for glucose are shown in Figure S1.

Bond	K_θ (kcal/mol/rad^2)	θ_0 (°)
CA-CE-CD	58.35	112.7
CA-CE-HE	37.5	110.7
CA-CE-OE	50	109.5
CA-OA-HA	55	108.5
CD-CD-CD	58.35	112.7
CD-CD-CE	58.35	112.7
CD-CD-CG	58.35	112.7
CD-CD-H	37.5	110.7
CD-CD-OD	50	109.5
CD-CE-HE	37.5	110.7
CD-CE-OE	50	109.5
CD-CG-HG	37.5	110.7
CD-CG-OD	50	109.5
CD-CG-OE	50	109.5
CD-OD-HD	55	108.5
CE-CA-H	37.5	110.7
CE-CA-OA	50	109.5
CE-CD-H	37.5	110.7
CE-CD-OD	50	109.5
CE-OE-CG	60	109.5
CG-CD-H	37.5	110.7
CG-CD-OD	50	109.5
CG-OD-HD	55	108.5
H-CA-H	33	107.8
H-CA-OA	35	109.5
H-CD-OD	35	109.5
HE-CE-OE	35	109.5
HG-CG-OD	35	109.5
HG-CG-OE	35	109.5
OD-CG-OE	92.6	111.55

Table S8: Dihedral angle torsion parameters (Equation (S7)) of the OPLS force field for glucose.[3] All V_1, V_2, and V_3 coefficients are reported in units of kcal/mol. Interaction sites for glucose are shown in Figure S1.

Dihedral angle	V_1	V_2	V_3	Dihedral angle	V_1	V_2	V_3
CA-CE-OE-CG	0.65	-0.25	0.67	H-CA-OA-HA	0	0	0.45
CD-CD-CD-CE	1.74	-0.157	0.279	H-CD-CD-H	0	0	0.318
CD-CD-CD-CG	1.74	-0.157	0.279	H-CD-CD-OD	0	0	0.468
CD-CD-CD-H	0	0	0.366	H-CD-CE-CA	0	0	0.366
CD-CD-CD-OD	-1.336	0	0	H-CD-CE-HE	0	0	0.318
CD-CD-CE-CA	1.74	-0.157	0.279	H-CD-CE-OE	0	0	0.468
CD-CD-CE-HE	0	0	0.366	H-CD-CG-HG	0	0	0.318
CD-CD-CE-OE	-1.336	0	0	H-CD-CG-OD	0	0	0.468
CD-CD-CG-HG	0	0	0.366	H-CD-CG-OE	0	0	0.468
CD-CD-CG-OD	-1.336	0	0	H-CD-OD-HD	0	0	0.45
CD-CD-CG-OE	-1.336	0	0	HE-CE-OE-CG	0	0	0.76
CD-CD-OD-HD	2.674	-2.883	1.026	HG-CG-OD-HD	0	0	0.45
CD-CE-OE-CG	0.65	-0.25	0.67	HG-CG-OE-CE	0	0	0.76
CD-CG-OD-HD	2.674	-2.883	1.026	OA-CA-CE-CD	-1.336	0	0
CD-CG-OE-CE	0.65	-0.25	0.67	OA-CA-CE-HE	0	0	0.468
CE-CA-OA-HA	2.674	-2.883	1.026	OA-CA-CE-OE	4.319	0	0
CE-CD-CD-H	0	0	0.366	OD-CD-CD-OD	9.066	0	0
CE-CD-CD-OD	-1.336	0	0	OD-CD-CE-CA	-1.336	0	0
CE-CD-OD-HD	2.674	-2.883	1.026	OD-CD-CE-HE	0	0	0.468
CG-CD-CD-H	0	0	0.366	OD-CD-CE-OE	4.319	0	0
CG-CD-CD-OD	-1.336	0	0	OD-CD-CG-HG	0	0	0.468
CG-CD-OD-HD	2.674	-2.883	1.026	OD-CD-CG-OD	4.319	0	0
H-CA-CE-CD	0	0	0.366	OD-CD-CG-OE	4.319	0	0
H-CA-CE-HE	0	0	0.318	OD-CG-OE-CE	-0.375	-1.358	0.004
H-CA-CE-OE	0	0	0.468	OE-CG-OD-HD	-1.257	-1.806	0.003

Table S9: Computed thermodynamic factors (Γ) of water-sucrose mixtures reported in Figures 2 to 4 of the main text. Temperature and pressure are 298 K and 1 atm, respectively. LJ energy parameters (ϵ) and partial atomic charges (q) are scaled by factors either 0.8 or 1. No additional independent simulations were performed to compute 95% confidence intervals.

force field	OPLS[2,3]	OPLS[2,3]	OPLS[2,3]	OPLS[2,3]	GLYCAM06[4]	GLYCAM06[5]
scaling factor for ϵ	0.8	0.8	1	1	original	modified
scaling factor for q	0.8	1	0.8	1	-	-
w_{sucrose}	Γ	Γ	Γ	Γ	Γ	Γ
0.2	0.73	0.38	1.16	0.99	0.33	1.35
0.3	0.58	0.30	1.31	0.89	0.29	1.63
0.4	0.56	0.29	1.61	1.08	0.28	1.99
0.5	0.52	0.29	1.88	1.08	0.27	2.54
0.6	0.53	0.26	1.83	1.13	0.42	3.18

Table S10: Computed shear viscosities (η) of water-sucrose mixtures reported in Figures 2 to 4 of the main text. Temperature and pressure are 298 K and 1 atm, respectively. LJ energy parameters (ϵ) and partial atomic charges (q) are scaled by factors either 0.8 or 1. All shear viscosities and 95% confidence intervals are reported in units of cP. 95% confidence intervals are computed from 5 independent simulations.

force field	OPLS[2,3]		OPLS[2,3]		OPLS[2,3]		OPLS[2,3]		GLYCAM06[4]		GLYCAM06[5]	
scaling factor for ϵ	0.8		0.8		1		1		original		modified	
scaling factor for q	0.8		1		0.8		1		-		-	
w_{sucrose}	$\bar{\eta}$	95%	$\bar{\eta}$	95%	$\bar{\eta}$	95%	$\bar{\eta}$	95%	$\bar{\eta}$	95%	$\bar{\eta}$	95%
0.2	1.12	0.04	1.15	0.07	1.48	0.14	1.45	0.08	1.13	0.05	1.19	0.03
0.3	1.49	0.08	1.55	0.10	2.50	0.35	2.45	0.11	1.69	0.16	1.80	0.13
0.4	2.27	0.27	2.50	0.40	5.41	0.42	5.59	0.31	10.28	4.06	3.31	0.34
0.5	4.10	0.23	7.07	0.68	20.20	2.86	23.00	4.60	226.76	105.70	8.84	0.75
0.6	10.69	0.79	24.16	7.20	124.40	40.39	179.21	63.60	732.07	230.93	43.80	4.67

Table S11: Computed liquid densities (ρ) of water-sucrose mixtures reported in Figure 5 of the main text. Temperature and pressure are 298 K and 1 atm, respectively. For the refined OPLS force field, LJ energy parameters (ϵ) and partial atomic charges (q) of the original OPLS force field[2,3] are scaled by factors 0.8 and 0.95, respectively. All densities are reported in units of kg/m^3. 95% confidence intervals are computed from block averages.

	original OPLS[3]		refined OPLS	
w_{sucrose}	$\bar{\rho}$	95%	$\bar{\rho}$	95%
0.2	1093.3	0.8	1090.3	0.7
0.3	1138.4	0.5	1133.3	0.7
0.4	1186.5	1.2	1180.0	0.5
0.5	1238.6	1.9	1229.8	0.8
0.6	1294.7	2.5	1282.6	1.4

Table S12: Computed thermodynamic factors (Γ) of water-sucrose mixtures reported in units of Figure 5 of the main text. Temperature and pressure are 298 K and 1 atm, respectively. For the refined OPLS force field, LJ energy parameters (ϵ) and partial atomic charges (q) of the original OPLS force field[2,3] are scaled by factors 0.8 and 0.95, respectively. 95% confidence intervals are computed from 5 independent simulations. No additional independent simulations were performed for the calculation of thermodynamic factors based on the original OPLS force field.

	original OPLS[3]		refined OPLS	
w_{sucrose}	$\bar{\Gamma}$	95%	$\bar{\Gamma}$	95%
0.2	0.99	-	1.09	0.04
0.3	0.89	-	1.18	0.06
0.4	1.08	-	1.36	0.05
0.5	1.08	-	1.47	0.04
0.6	1.13	-	1.90	0.15

Table S13: Computed shear viscosities (η) of water-sucrose mixtures reported in Figure 5 of the main text. Temperature and pressure are 298 K and 1 atm, respectively. For the refined OPLS force field, LJ energy parameters (ϵ) and partial atomic charges (q) of the original OPLS force field[2,3] are scaled by factors 0.8 and 0.95, respectively. All shear viscosities are reported in units of cP. 95% confidence intervals are computed from 5 independent simulations.

	original OPLS[3]		refined OPLS	
w_{sucrose}	$\bar{\eta}$	95%	$\bar{\eta}$	95%
0.2	1.45	0.08	1.38	0.07
0.3	2.45	0.11	2.16	0.10
0.4	5.59	0.31	4.18	0.14
0.5	23.00	4.60	11.83	1.53
0.6	179.21	63.60	57.63	16.00

Table S14: Computed self-diffusion coefficients of sucrose ($D_{\text{sucrose,self}}$) in water-sucrose mixtures reported in Figure 5 of the main text. Temperature and pressure are 298 K and 1 atm, respectively. For the refined OPLS force field, LJ energy parameters (ϵ) and partial atomic charges (q) of the original OPLS force field[2,3] are scaled by factors 0.8 and 0.95, respectively. All self-diffusivities are reported in units of 10^{-12} m^2/s. 95% confidence intervals are computed from 5 independent simulations.

	original OPLS[3]		refined OPLS	
w_{sucrose}	$\bar{D}_{\text{sucrose,self}}$	95%	$\bar{D}_{\text{sucrose,self}}$	95%
0.2	337.72	5.19	377.85	5.20
0.3	198.34	3.52	245.29	4.51
0.4	93.12	1.70	133.44	1.83
0.5	25.67	1.72	50.15	2.53
0.6	4.73	0.27	12.85	0.52

Table S15: Computed self-diffusion coefficients of water ($D_{\text{water,self}}$) in water-sucrose mixtures reported in Figure 5 of the main text. Temperature and pressure are 298 K and 1 atm, respectively. For the refined OPLS force field, LJ energy parameters (ϵ) and partial atomic charges (q) of the original OPLS force field[2,3] are scaled by factors 0.8 and 0.95, respectively. All self-diffusivities are reported in units of 10^{-11} m^2/s. 95% confidence intervals are computed from 5 independent simulations.

	original OPLS[3]		refined OPLS	
w_{sucrose}	$\bar{D}_{\text{water,self}}$	95%	$\bar{D}_{\text{water,self}}$	95%
0.2	176.51	0.75	181.03	0.70
0.3	131.09	0.58	137.37	0.18
0.4	89.20	0.36	96.06	0.88
0.5	50.82	1.01	56.55	0.56
0.6	23.10	0.79	27.84	0.49

Table S16: Computed Fick diffusion coefficients (D_{Fick}) of water-sucrose mixtures reported in Figure 5 of the main text. Temperature and pressure are 298 K and 1 atm, respectively. For the refined OPLS force field, LJ energy parameters (ϵ) and partial atomic charges (q) of the original OPLS force field[2,3] are scaled by factors 0.8 and 0.95, respectively. All diffusivities are reported in units of 10^{-11} m^2/s. 95% confidence intervals are computed from error propagation analysis based on 5 independent simulations for computed Maxwell-Stefan diffusion coefficients and thermodynamic factors.

	original OPLS[3]		refined OPLS	
w_{sucrose}	$\bar{D}_{\text{Fick}}$	95%	$\bar{D}_{\text{Fick}}$	95%
0.2	43.46	2.15	47.28	3.01
0.3	29.14	2.34	37.09	2.27
0.4	21.45	1.32	27.88	1.55
0.5	12.61	1.02	17.35	1.81
0.6	5.75	1.24	10.70	3.11

Table S17: Computed liquid densities (ρ) of water-glucose mixtures reported in Figure 6 of the main text. Temperature and pressure are 298 K and 1 atm, respectively. For the refined OPLS force field, LJ energy parameters (ϵ) and partial atomic charges (q) of the original OPLS force field[2,3] are scaled by factors 0.8 and 0.95, respectively. All densities are reported in units of kg/m^3. 95% confidence intervals are computed from block averages.

	original OPLS[3]		refined OPLS	
w_{glucose}	$\bar{\rho}$	95%	$\bar{\rho}$	95%
0.2	1094.8	0.8	1091.1	0.8
0.3	1140.8	0.8	1135.4	1.0
0.4	1189.5	1.1	1182.4	1.1
0.5	1241.6	1.6	1232.4	1.6

Table S18: Computed thermodynamic factors (Γ) of water-glucose mixtures reported in Figure 6 of the main text. Temperature and pressure are 298 K and 1 atm, respectively. For the refined OPLS force field, LJ energy parameters (ϵ) and partial atomic charges (q) of the original OPLS force field[2,3] are scaled by factors 0.8 and 0.95, respectively. 95% confidence intervals are computed from 5 independent simulations.

	original OPLS[3]		refined OPLS	
w_{glucose}	$\bar{\Gamma}$	95%	$\bar{\Gamma}$	95%
0.2	0.91	0.01	1.06	0.01
0.3	0.90	0.03	1.12	0.01
0.4	0.95	0.02	1.21	0.03
0.5	0.97	0.04	1.34	0.03

Table S19: Computed shear viscosities (η) of water-glucose mixtures reported in Figure 6 of the main text. Temperature and pressure are 298 K and 1 atm, respectively. For the refined OPLS force field, LJ energy parameters (ϵ) and partial atomic charges (q) of the original OPLS force field[2,3] are scaled by factors 0.8 and 0.95, respectively. All shear viscosities are reported in cP. 95% confidence intervals are computed from 5 independent simulations.

	original OPLS[3]		refined OPLS	
w_{glucose}	$\bar{\eta}$	95%	$\bar{\eta}$	95%
0.2	1.35	0.02	1.29	0.08
0.3	2.23	0.06	2.01	0.15
0.4	4.68	0.23	3.61	0.27
0.5	13.38	0.46	8.45	0.43

Table S20: Computed self-diffusion coefficients of glucose ($D_{\mathrm{glucose,self}}$) in water-glucose mixtures reported in Figure 6 of the main text. Temperature and pressure are 298 K and 1 atm, respectively. For the refined OPLS force field, LJ energy parameters (ϵ) and partial atomic charges (q) of the original OPLS force field[2,3] are scaled by factors 0.8 and 0.95, respectively. All self-diffusivities are reported in units of 10^{-12} m^2/s. 95% confidence intervals are computed from 5 independent simulations.

	original OPLS[3]		refined OPLS	
w_{glucose}	$\bar{D}_{\mathrm{s,sucrose}}$	95%	$\bar{D}_{\mathrm{s,sucrose}}$	95%
0.2	463.13	4.43	517.54	3.67
0.3	283.99	4.58	346.76	1.16
0.4	146.87	0.26	204.08	0.91
0.5	57.84	0.68	95.73	1.63

Table S21: Computed self-diffusion coefficients of water ($D_{\text{water,self}}$) in water-glucose mixtures reported in units of Figure 6 of the main text. Temperature and pressure are 298 K and 1 atm, respectively. For the refined OPLS force field, LJ energy parameters (ϵ) and partial atomic charges (q) of the original OPLS force field[2,3] are scaled by factors 0.8 and 0.95, respectively. All self-diffusivities are reported in units of 10^{-11} m^2/s. 95% confidence intervals are computed from 5 independent simulations.

	original OPLS[3]		refined OPLS	
w_{glucose}	$\bar{D}_{\text{s,water}}$	95%	$\bar{D}_{\text{s,water}}$	95%
0.2	179.73	0.29	185.40	0.47
0.3	132.45	0.37	139.80	0.51
0.4	88.89	0.29	97.20	0.27
0.5	52.23	0.44	59.72	0.29

Table S22: Computed Fick diffusion coefficients (D_{Fick}) of water-glucose mixtures reported in Figure 6 of the main text. Temperature and pressure are 298 K and 1 atm, respectively. For the refined OPLS force field, LJ energy parameters (ϵ) and partial atomic charges (q) of the original OPLS force field[2,3] are scaled by factors 0.8 and 0.95, respectively. All diffusivities are reported in units of 10^{-11} m^2/s. 95% confidence intervals are computed from error propagation analysis based on 5 independent simulations for computed Maxwell-Stefan diffusion coefficients and thermodynamic factors.

	original OPLS[3]		refined OPLS	
w_{glucose}	$\bar{D}_{\text{Fick}}$	95%	$\bar{D}_{\text{Fick}}$	95%
0.2	56.50	2.59	64.75	2.70
0.3	41.24	1.72	49.90	3.07
0.4	28.42	2.18	37.70	2.13
0.5	16.91	1.08	25.53	2.19

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