Supporting information: Structural and thermodynamic properties of the bulk triglycerides and triglyceride/water mixture reproduced by a polarizable coarse-grained model

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Table S1 Accuracy tests of three different AA FFs for predicting ρ and ΔH_{vap} of n-octane (C04-C04), (Z)-oct-2-ene (B03-C04), (Z)-hept-2-ene (B03-C03), and methyl butanoate (B06-C03)

Molecules	CG names	Force field	ρ (kg/m3)			ΔH_{vap} (kJ/mol)		
			Sim	Exp	|δ|	Sim	Exp	|δ|
n-octane	C04-C04	GROMOS54a7-UA	707		1.2%	43.7		5.3%
		GROMOS54a7-AA	734	699	5.1%	47.8	41.5	15%
		OPLS-AA	694		0.1%	46.1		11%
(Z)-oct-2-ene	B03-C04	GROMOS54a7-UA	723		0.4%	42.3		5.3%
		GROMOS54a7-AA	752	720	4.4%	44.7	40.2	11%
		OPLS-AA	710		1.4%	44.8		11%
(Z)-hept-2-ene	B03-C03	GROMOS54a7-UA	707		0.6%	37.2		2.5%
		GROMOS54a7-AA	735	703	4.6%	40.0	36.3	10%
		OPLS-AA	691		1.7%	38.7		6.6%
methyl butanoate	B06-C03	GROMOS54a7-UA	884		0.7%	44.7		11%
		GROMOS54a7-AA	971	890	9.2%	54.1	40.3	34%
		OPLS-AA	879		1.2%	44.6		11%

Sim, Exp and |δ| refer to results obtained from simulations, experiments and corresponding relative deviations. Experimental ρ are from ref. 1, 2 and experimental ΔH_{vap} are from ref. 1, 3, 4.

Table S2 Accuracy tests of OPLS-AA FF for predicting ρ and ΔH_{vap} of methyl formate (B06) and (Z)-but-2-ene (B03)

Molecules	CG names	ρ (kg/m3)			ΔH_{vap} (kJ/mol)		
		Sim	Exp	δ	Sim	Exp	δ
methyl formate	B06	931	967	3.8%	28.2	28.4	0.7%
(Z)-but-2-ene	B03	600	617	2.8%	22.9	22.2	3.0%

Experimental ρ are from ref. 1, 2 and experimental ΔHvap are from ref. 1, 3, 4

Table S3 Nonbonded parameters of TG and PCGW

Nonbonded parameters	ε (kJ/mol)	R_0 (nm)	α	β
B06~B06[a]	5.207	0.504	7.638	17.538
B06~B03[a]	4.791	0.534	9.195	16.399
B06~C04[a]	4.087	0.548	9.887	15.811
B06~C03	3.047	0.549	10.639	14.451
B03~B03[a]	4.409	0.565	11.070	15.334
B03~C04	4.127	0.560	11.903	14.784
B03~C03	3.480	0.548	11.254	13.512
C04~C04	4.357	0.578	12.798	14.254
C04~C03[a]	3.671	0.556	12.101	13.028
C03~C03[a]	3.091	0.534	11.442	11.907
B06~WO	5.078	0.570	7.566	12.809
B03~WO	4.050	0.580	9.108	11.977
C04~WO[b]	3.448	0.554	9.793	11.548
C03~WO[b]	2.797	0.539	9.260	10.554
WO~WO[b]	5.209	0.594	7.494	9.355

[a] Non-bonded parameters are directly obtained from ref.5, [b] Non-bonded parameters are directly obtained from ref. 6.

Table S4 Bonded parameters of TG

Bond	L_0 (nm)	k_b (kJ/mol/nm2)	Angle	ϑ_0 (°)	k_a (kJ/mol)
B06-B06[a]	0.400	1250	B03-C04-C04	164	25
C04-C04	0.460	2500	C03-B03-C04	132	25
B03-C04	0.447	2500	B06-C03-B03	163	25
B03-C03	0.400	2500			
B06-C03	0.360	1250			

[a] Bonded parameters are directly obtained from ref. 5.

Table S5 Effects of the thickness of TG layer on the proportions of various TG conformations inside the bulk phase and at the TG/water interfaces, water content penetrating into the TG-rich phase, and interfacial tension

	256 TGs + 6081 PCGWs		512 TGs+6081 PCGWs		1024 TGs + 6081 PCGWs	
Conformations (%)	In bulk (~116 TGs)	At surface (~140 TGs)	In bulk (~372 TGs)	At surface (~140 TGs)	In bulk (~887 TGs)	At surface (~137 TGs)
Propeller	24.56	8.70	24.53	8.74	25.28	8.52
Tuning fork	18.71	9.66	17.68	10.49	18.96	9.17
Chair	22.09	12.84	24.98	12.19	23.41	13.61
Trident	34.64	68.80	32.81	68.58	32.35	68.70
	256 TGs + 6081 PCGWs		512 TGs + 6081 PCGWs		1024 TGs + 6081 PCGWs	
Number of PCGW in bulk TG phase	2.5		7.5		16.0	
ΔG_p (k_BT)	7.6		6.6		7.1	
$\gamma_{TG/W}$ (mN/m)	33.6		32.9		31.5	

Simulation settings of three TG/water systems are strictly identical except for the thickness of TG bulk phase.

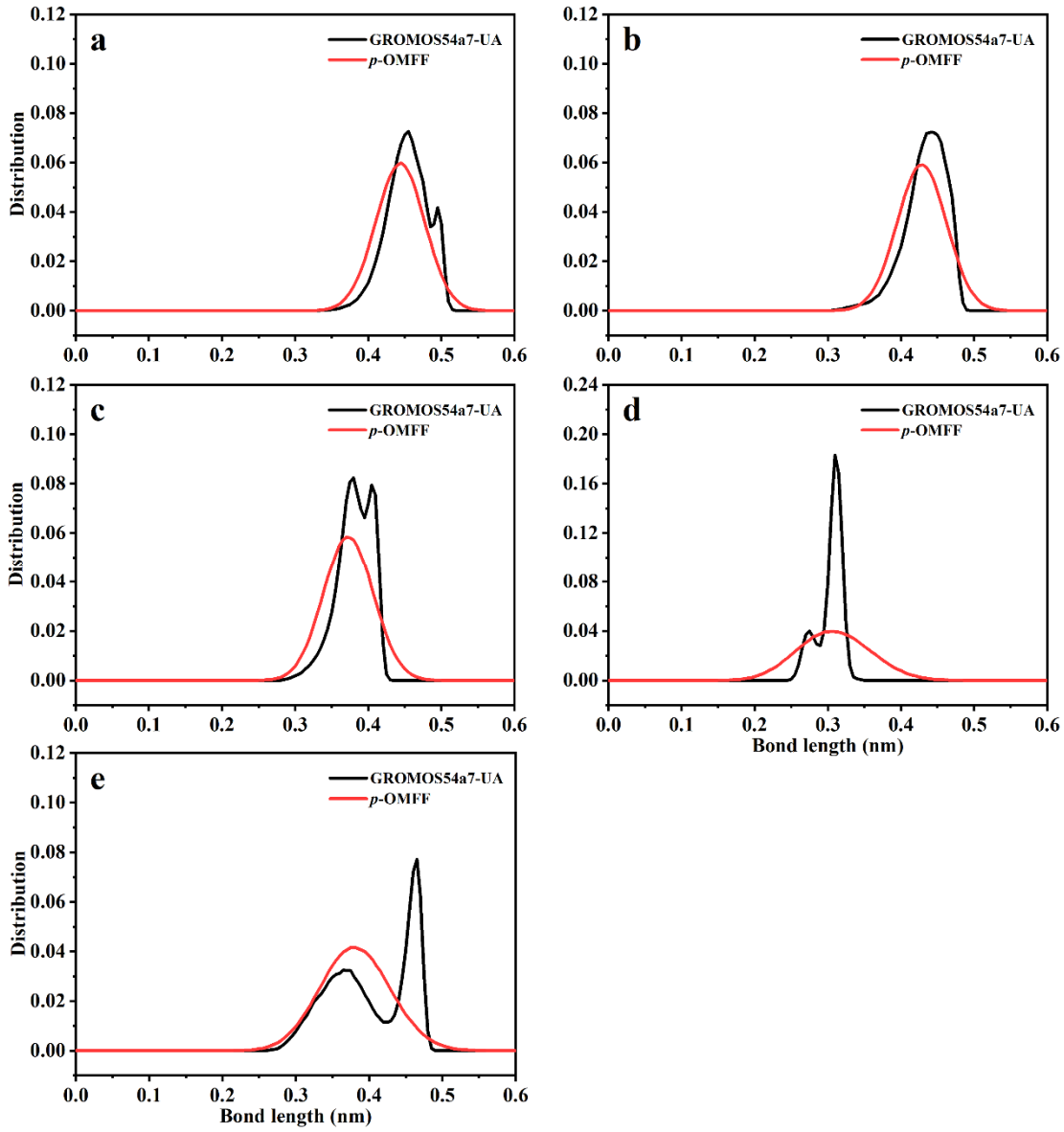

Fig. S1 Bond length distributions of C04-C04 (a), B03-C04 (b), B03-C03 (c), B06-C03 (d), and B06-B06 (e) of TG obtained from AA and CG MD simulations.

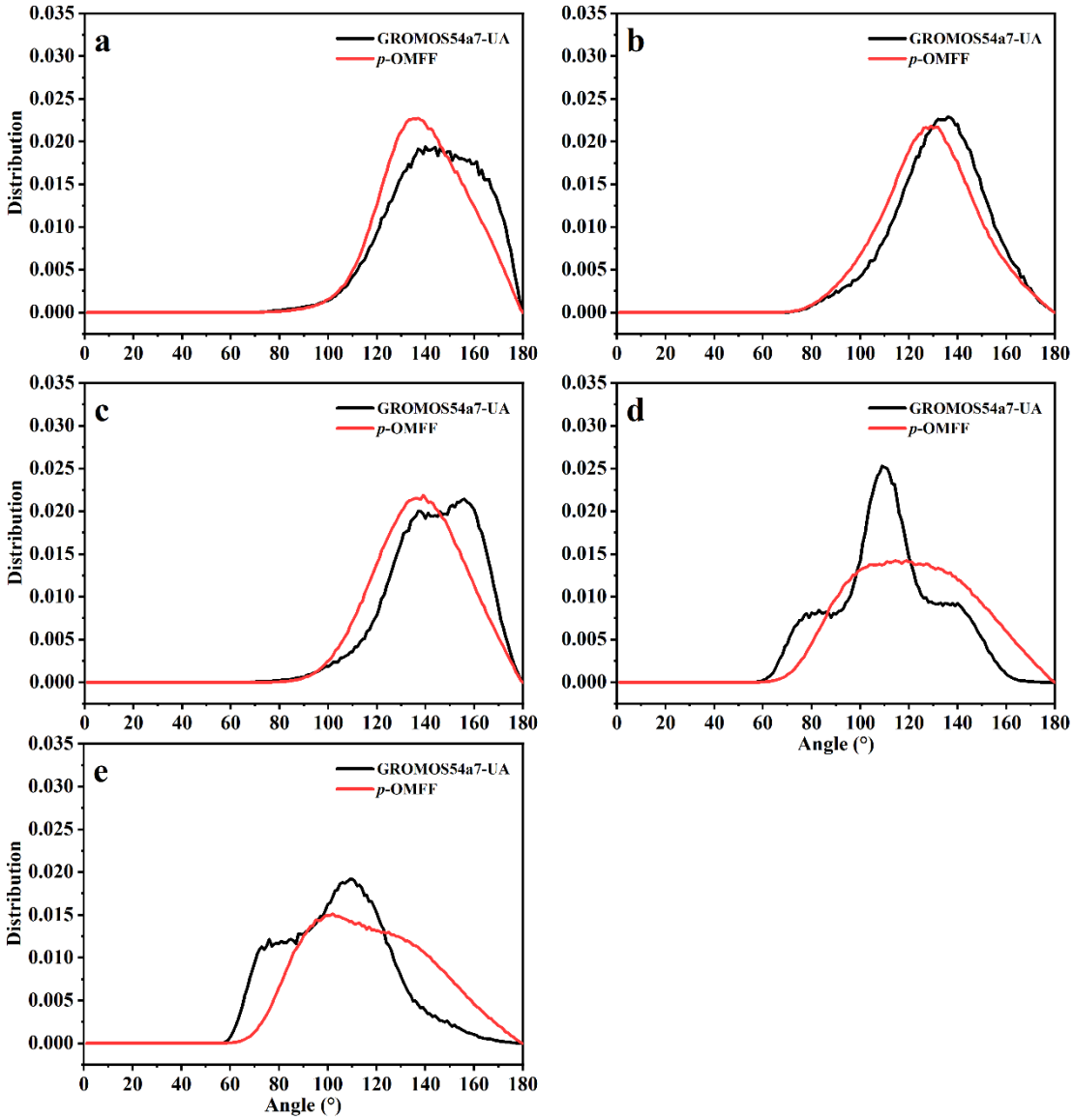

Fig. S2 Angle distributions of B03-C04-C04 (a), C03-B03-C04 (b), B06-C03-B03 (c), B06-B06-C03 (d), and C03-B06-B06 (e) of TG obtained from AA and CG MD simulations. In (d), C03 bead locates in the first or the third fatty acyl chain; in (e), C03 bead locates in the second fatty acyl chain. See Figure 2 for the definition of the order of the chain

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