

FIG. 1: Pressure composition plot for chloroform(1)- acetone(2) mixture at  $T=450$  K. CHELPG charges of acetone-chloroform aggregate (circle), CHELPG charges of isolated acetone and chloroform at the MP2/6-31++g(3df,3dp) (square). Dashed lines are guide to eye.

TABLE I: Selected coexistence points for the pressure composition plot for chloroform(1)-acetone(2) mixture at T=450 K (a) using CHELPG charges of acetone-chloroform aggregate, (b) using CHELPG charges of isolated acetone and chloroform at the MP2/6-31++g(3df,3dp)

P (bar)	$x_{\text{CHCl}_3}$	$y_{\text{CHCl}_3}$
15.05	0.992	0.995
15.30	0.956	0.968
15.70	0.895	0.921
16.44	0.750	0.806
16.70	0.684	0.752
17.02	0.598	0.678
17.42	0.488	0.581
17.86	0.360	0.444
18.32	0.212	0.289
19.02	0.0267	0.046
19.18	0.00321	0.0043
P (bar)	$x_{\text{CHCl}_3}$	$y_{\text{CHCl}_3}$
15.00	0.990	0.991
14.98	0.966	0.966
14.97	0.948	0.948
14.98	0.910	0.901
14.99	0.90	0.899
14.99	0.890	0.887
15.04	0.832	0.825
15.13	0.772	0.757
15.71	0.590	0.547
16.15	0.504	0.447
17.85	0.266	0.210
18.55	0.190	0.142
19.36	0.094	0.07
19.61	0.008	0.009

TABLE II: Selected coexistence points for the pressure composition plot for chloroform(1)-acetone(2) mixture at T=400 K.

P (bar)	$x_{\text{CHCl}_3}$	$y_{\text{CHCl}_3}$
5.51	0.996	0.996
5.49	0.970	0.972
5.47	0.950	0.954
5.47	0.944	0.949
5.46	0.937	0.934
5.45	0.900	0.904
5.45	0.875	0.877
5.45	0.844	0.842
5.45	0.825	0.822
5.46	0.805	0.798
5.48	0.757	0.743
5.50	0.731	0.710
5.52	0.701	0.674
5.56	0.668	0.633
5.61	0.633	0.589
5.87	0.503	0.431
6.00	0.444	0.370
6.16	0.380	0.304
6.36	0.310	0.236
6.61	0.232	0.166
6.92	0.141	0.094
8.10	0.0007	0.00078

TABLE III: Selected coexistence points for the pressure composition plot for chloroform(1)-acetone(2) mixture at T=350 K.

P (bar)	$x_{\text{CHCl}_3}$	$y_{\text{CHCl}_3}$
1.47	0.998	0.999
1.46	0.970	0.977
1.452	0.941	0.952
1.45	0.933	0.944
1.447	0.923	0.936
1.44	0.913	0.926
1.437	0.889	0.901
1.434	0.874	0.885
1.432	0.858	0.868
1.43	0.840	0.847
1.429	0.820	0.824
1.433	0.749	0.731
1.440	0.718	0.691
1.45	0.682	0.647
1.46	0.642	0.597
1.48	0.60	0.541
1.51	0.551	0.480
1.54	0.499	0.414
1.59	0.441	0.344
1.65	0.379	0.271
1.73	0.303	0.198
1.83	0.210	0.124
1.96	0.111	0.062
2.21	0.0008	0.0007

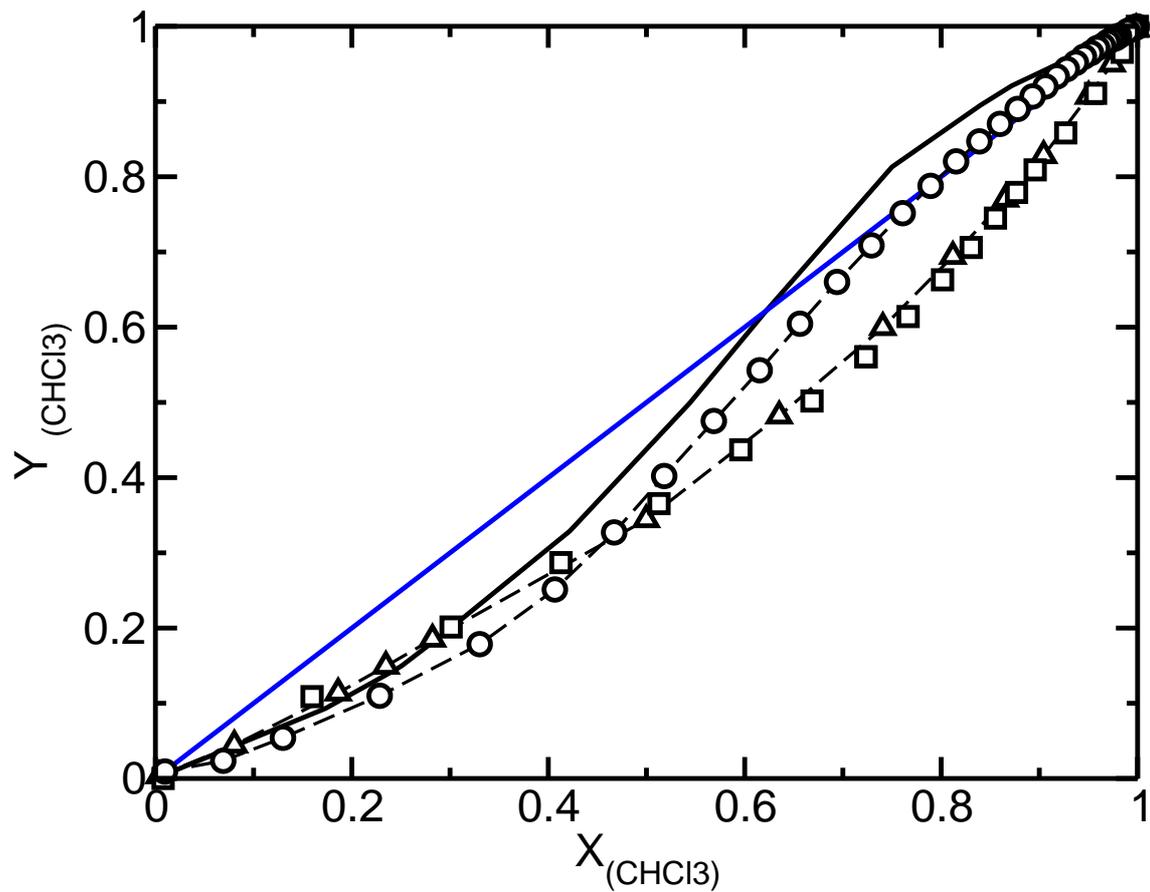


FIG. 2: Composition plot for chloroform(1)- acetone(2) mixture at  $T=308.32$  K. New (1)/New (2) (Circle), CDP(1)/TraPPE(2) (square) and OPLS-UA (1)/OPLS-UA (2) (Triangle) and expt (line). Equilibrium line is shown in blue.

TABLE IV: Selected coexistence points predicted by simulation using the new, OPLS-UA, and TraPPE force fields for acetone.

model	T (K)	P (bar)	$\rho_{gas}$ (kg/m <sup>3</sup> )	$\rho_{liq}$ (kg/m <sup>3</sup> )
new force field	470	27.35±0.1	64.02±1.4	524.24±1.2
	450	19.62±0.06	43.47 ±0.4	573.17 ±0.9
	430	13.64±0.04	28.85±0.1	611.14±0.3
	400	7.41±0.01	15.22 ±0.4	656.44 ±1.0
	370	3.63±0.04	7.54 ±0.3	697.15 ±0.8
	350	2.09±0.06	4.45 ±0.1	723.18±0.9
	330	1.12 ±0.05	2.46 ±0.3	747.40 ±0.9
	300	0.36±0.03	0.86 ±0.2	775.49 ±0.8
OPLS-UA	480	26.88	98.17	520.80
	460	21.15	64.02	563.90
	440	14.43	44.24	597.84
	420	10.07	27.39	627.38
	400	6.81	18.20	657.54
	380	4.13	12.53	684.60
	360	2.38	8.32	711.38
	340	1.36	4.57	733.90
	320	0.67	2.81	756.22
	300	0.36	1.46	777.90
TraPPE-UA	300	0.423	1.1	773.3
	340	1.82	4.36	729
	380	5.56	12.32	681.1
	420	13.17	28.22	626.62
	460	26.44	59.74	556.1
	500	47.7	134.35	435.9

TABLE V: Selected coexistence points predicted by simulation using the new, OPLS-UA, and CDP force fields for chloroform.

model	T (K)	P (bar)	$\rho_{gas}$ (kg/m <sup>3</sup> )	$\rho_{liq}$ (kg/m <sup>3</sup> )
new force field	500	33.08±1.0	149.96±0.1	956.72±3.0
	470	20.85 ±0.2	87.47 ±0.1	1081.52±0.8
	450	14.82 ±0.2	59.78±0.1	1146.4±0.1
	430	10.21±0.2	40.49±0.1	1203.1±0.1
	400	5.41 ±0.4	21.62 ±0.1	1268.95±1.0
	370	2.57 ±0.4	10.63±0.1	1336.71±1.5
	350	1.47±0.1	6.271±0.1	1380.71±1.2
	330	0.78 ±0.1	3.50 ±0.1	1418.51±1.5
	300	0.27 ±0.03	1.53 ±0.1	1475.59±1.2
OPLS-UA	470	18.15	82.47	1121.51
	440	11.02	43.34	1206.13
	420	7.39	28.32	1251.64
	400	4.75	18.62	1297.55
	380	2.8	11.31	1336.08
	360	1.52	6.297	1379.31
	340	0.93	4.042	1415.67
	320	0.45	2.111	1451.30
CDP	500	33.96	142.16	955.89
	470	21.45	82.46	1089.51
	430	11.66	46.77	1200.59
	410	7.33	29.52	1247.28
	390	4.84	19.86	1292.08
	370	3.20	13.15	1337.06
	350	1.61	7.04	1377.12
	330	0.88	3.90	1420.84
	310	0.29	1.39	1455.38

TABLE VI: Selected coexistence points for the pressure composition plot for chloroform(1)-acetone(2) mixture at T=308.32 K using the new force fields.

P (bar)	$x_{\text{CHCl}_3}$	$y_{\text{CHCl}_3}$
0.335±0.004	0.999	0.999
0.331±0.003	0.960	0.971
0.327±0.004	0.920	0.933
0.324±0.005	0.878	0.890
0.323±0.004	0.839	0.847
0.322±0.003	0.789	0.788
0.335±0.004	0.615	0.543
0.391±0.005	0.407	0.252
0.454±0.003	0.229	0.110
0.558±0.007	0.009	0.009

TABLE VII: Selected coexistence points for the pressure composition plot for chloroform(1)-acetone(2) mixture at T=308.32 K using OPLS-UA force field.

P (bar)	$x_{\text{CHCl}_3}$	$y_{\text{CHCl}_3}$
1.194	0.998	0.996
1.201	0.974	0.950
1.207	0.950	0.907
1.265	0.741	0.60
1.292	0.635	0.482
1.392	0.235	0.150
1.450	0.003	0.003

TABLE VIII: Selected coexistence points for the pressure composition plot for chloroform(1)-acetone(2) mixture at T=308.32 K using CDP(1)/TraPPE(2) force fields.

P (bar)	$x_{\text{CHCl}_3}$	$y_{\text{CHCl}_3}$
0.363	0.999	1
0.40	0.927	0.858
0.457	0.801	0.663
0.50	0.724	0.561
0.538	0.596	0.437
0.566	0.513	0.366
0.665	0.160	0.109
0.697	0.007	0

TABLE IX: Selected coexistence points for the pressure composition plot for acetone(1)-methanol(2) mixture at T=323.15 K using new(1)/TraPPE (2) force fields.

P (bar)	$x_{\text{acetone}}$	$y_{\text{acetone}}$
3.91	0.998	0.998
4.00	0.935	0.956
4.101	0.875	0.910
4.188	0.816	0.85
4.25	0.762	0.80
4.28	0.729	0.767
4.331	0.675	0.705
4.342	0.657	0.684
4.352	0.639	0.660
4.359	0.619	0.63
4.364	0.598	0.605
4.364	0.576	0.571
4.363	0.570	0.570
4.359	0.551	0.534
4.346	0.525	0.494
4.32	0.496	0.45
4.295	0.464	0.408
4.25	0.428	0.358
4.188	0.388	0.302
4.104	0.342	0.252
4.008	0.29	0.208
3.903	0.238	0.159
3.751	0.18	0.108
3.572	0.134	0.065
3.217	0.064	0.022
3.015	0.024	0.008
2.92365	0.00793	0.00758

TABLE X: Selected coexistence points for the pressure composition plot for acetone(1)-methanol(2) mixture at T=323.15 K using TraPPE force field.

P (bar)	$x_{\text{acetone}}$	$y_{\text{acetone}}$
4.493	0.998	0.999
4.71	0.941	0.977
4.877	0.902	0.957
5.053	0.85	0.926
5.199	0.81	0.888
5.276	0.789	0.858
5.335	0.765	0.828
5.362	0.752	0.809
5.388	0.738	0.789
5.411	0.724	0.766
5.443	0.691	0.708
5.445	0.680	0.680
5.447	0.67	0.669
5.437	0.651	0.620
5.404	0.62	0.558
5.334	0.602	0.48
5.217	0.570	0.413
5.043	0.531	0.34
4.806	0.48	0.274
4.493	0.424	0.207
4.096	0.346	0.142
3.564	0.234	0.071
2.807	0.016	0.011

TABLE XI: Selected coexistence points for the pressure composition plot for chloroform(1)-methanol(2) mixture at T=323.15 K using new (1)/TraPPE (2) force fields.

P (bar)	$x_{\text{CHCl}_3}$	$y_{\text{CHCl}_3}$
0.674	0.999	0.99994
0.755	0.865	0.97994
0.847	0.73	0.93365
0.878	0.696	0.90058
0.891	0.680	0.87962
0.903	0.664	0.84472
0.914	0.646	0.7829
0.921	0.62	0.68223
0.921	0.601	0.56139
0.913	0.589	0.48705
0.898	0.546	0.38585
0.877	0.49	0.31054
0.842	0.45	0.23575
0.606	0.230	0.06107
0.440	0.126	0.01871
0.372	0	1e-05

TABLE XII: Selected coexistence points for the pressure composition plot for chloroform(1)-methanol(2) mixture at T=323.15 K using CDP(1)/TraPPE (2) force fields.

P (bar)	$x_{\text{CHCl}_3}$	$y_{\text{CHCl}_3}$
0.69095	0.999	0.999
0.79519	0.868	0.96
0.94235	0.728	0.906
0.99419	0.687	0.873
1.01314	0.672	0.854
1.03321	0.6579	0.825
1.05384	0.6417	0.783
1.07446	0.625	0.741
1.08896	0.6153	0.626
1.0622	0.5892	0.487
0.98527	0.5464	0.386
0.90667	0.495	0.281
0.68069	0.3453	0.130
0.41181	0.129	0.011
0.37924		1e-05

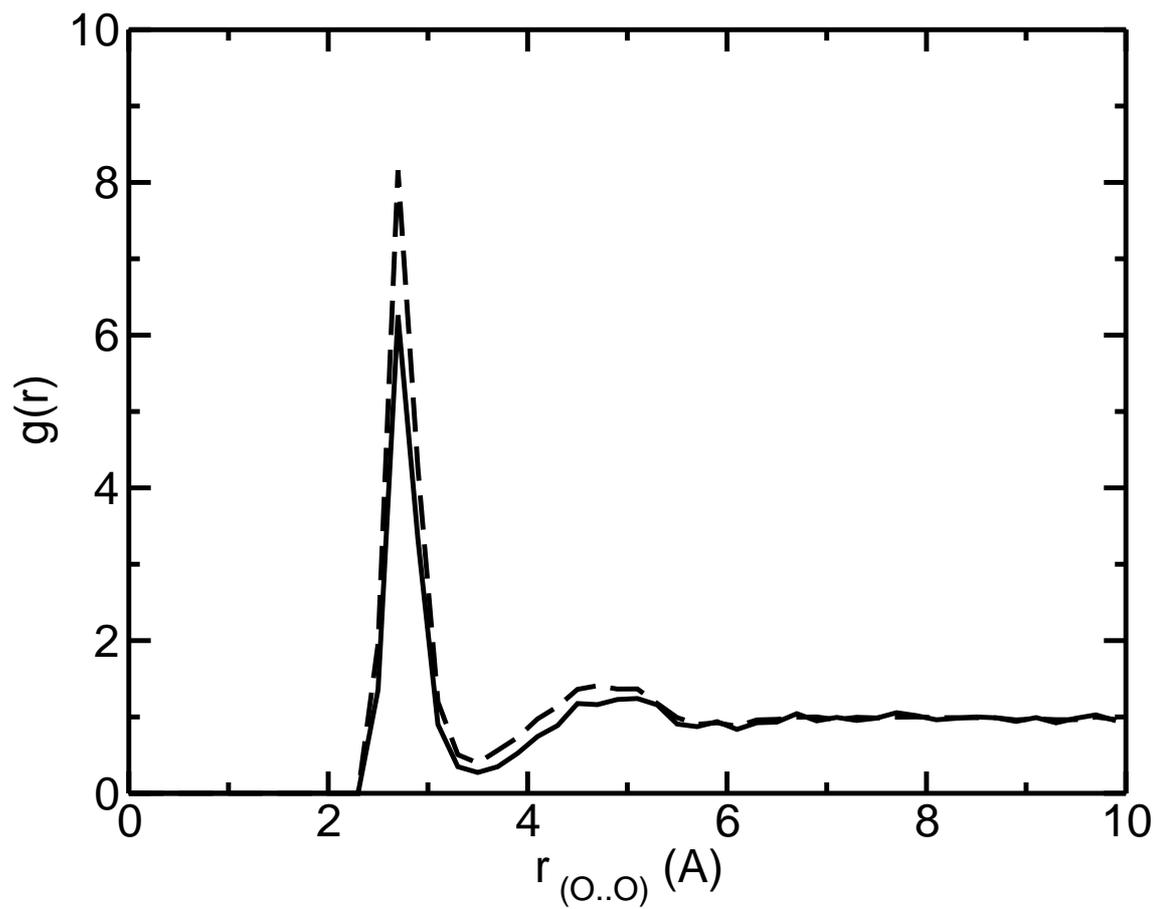


FIG. 3: Radial Distribution function for the  $O_{\text{MeOH}}-O_{\text{MeOH}}$  pair interaction in acetone methanol mixture at  $T=300$  K and  $P=1$  bar. TraPPE(1)/TraPPE(2) (square), new(1)/TraPPE (2) (circle).

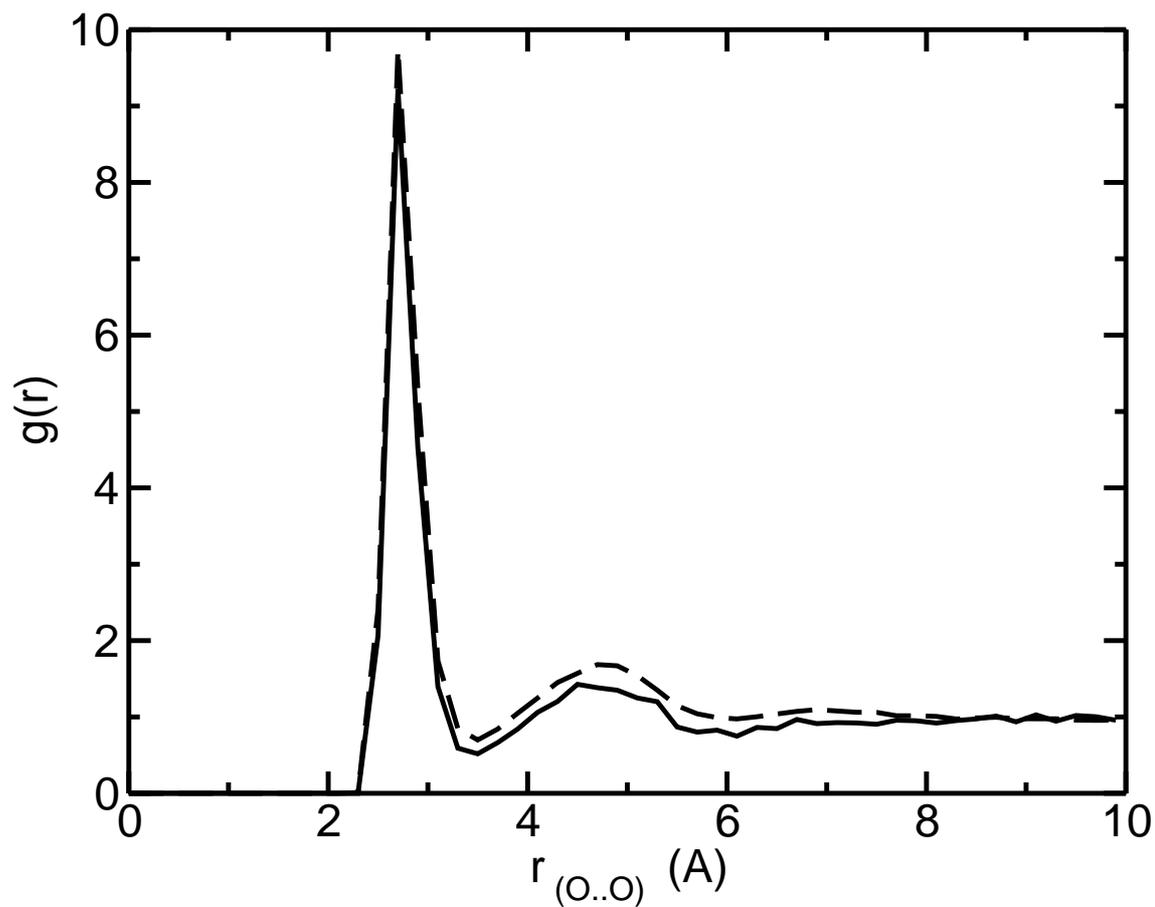


FIG. 4: Radial Distribution function for the pair  $O_{\text{MeOH}}-O_{\text{MeOH}}$  interaction in methanol chloroform mixture at  $T=300 \text{ K}$  and  $P=1 \text{ bar}$ . TraPPE(1)/TraPPE(2) (square), new(1)/TraPPE (2) (circle).