

Supporting Information

Densities, Speeds of Sound, Excess Molar Enthalpies and Heat Capacities of *o*-Chlorotoluene and Cyclic Ethers Mixtures

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Table S1. Measured Excess Molar Enthalpies, H^E Values for the Various (1 + 2) Mixtures as a Function of Mole Fraction, x_1 of Component (1) at $T/K = 308.15$ and $p = 0.1\text{MPa}$

x_1	$H^E / \text{J. mol}^{-1}$	x_1	$H^E / \text{J. mol}^{-1}$
<i>o</i> -Chlorotoluene (1) +1,3-dioxolane(2)			
0.0945	151	0.5432	383
0.1378	211	0.5899	368
0.1987	276	0.6329	343
0.2396	313	0.6923	307
0.2914	352	0.7321	272
0.3467	376	0.8021	211
0.3962	392	0.8492	164
0.4532	400	0.8912	122
0.4991	393	0.9319	77.9
<i>o</i> -Chlorotoluene (1) +1,4 dioxane (2)			
0.0998	162	0.5563	338
0.1643	244	0.6019	315
0.2095	283	0.6532	281
0.2498	315	0.7067	246
0.3012	346	0.7538	206
0.3567	365	0.8093	160
0.4098	368	0.8498	125
0.4597	367	0.8932	87.5
0.5088	356	0.9265	60.2
<i>o</i> -Chlorotoluene (1) +tetrahydropyran (2)			
0.0994	-69.4	0.5267	-220
0.1367	-94.5	0.5894	-214
0.1982	-127	0.6276	-211
0.2467	-154	0.6934	-191
0.3018	-178	0.7368	-175
0.3537	-192	0.7864	-155

0.3986	-204	0.8254	-134
0.4367	-214	0.8765	-99.0
0.4825	-220	0.9078	-76.9

Standard uncertainties u are $u(x_1) = \pm 1 \cdot 10^{-4}$, $u(T) = \pm 0.02\text{K}$, $u(H^E) = 0.1\%$

1 PRIGOGINE-FLORY-PATTERSON (PFP) THEORY

1.1 Excess molar volumes

According to PFP theory, ¹excess molar volumes, V^E is given by

$$V^E = V_{\text{interaction}}^E + V_{\text{free-volume}}^E + V_{\text{p}^* \text{effect}}^E \quad (\text{S1})$$

$$V_{\text{interaction}}^E = \left[\sum_{i=1}^2 x_i v_i^* \right] \left[\frac{\{(\tilde{v}^{1/3} - 1)\tilde{v}^{2/3}\psi_1\theta_2\chi_{12}^{**}\}}{\{(4/3)\tilde{v}^{-1/3} - 1\}\tilde{v}} \right] \quad (\text{S2})$$

$$V_{\text{free-volume}}^E = \left[\sum_{i=1}^2 x_i v_i^* \right] \left[(\tilde{v}_1 - \tilde{v}_2)^2 \{ (14/9)\tilde{v}^{-1/3} - 1 \} \right] \left[\frac{\psi_1\psi_2}{\{(4/3)\tilde{v}^{-1/3} - 1\}\tilde{v}} \right] \quad (\text{S3})$$

$$V_{\text{p}^* \text{effect}}^E = \left[\sum_{i=1}^2 x_i v_i^* \right] \left[\frac{(\tilde{v}_1 - \tilde{v}_2)(P_1^* - P_2^*)\psi_1\psi_2}{(P_1^*\psi_2 - P_2^*\psi_1)} \right] \quad (\text{S4})$$

$V_{\text{interaction}}^E$, $V_{\text{free-volume}}^E$, $V_{\text{p}^* \text{effect}}^E$ represent interaction, free volume and internal pressure contribution respectively and x_i ($i = 1$ or 2) represent mole fraction of component 1 or 2 of the binary mixtures.

$$\theta_2 = \varphi_2 s_2 / (\varphi_1 s_1 + \varphi_2 s_2) \quad (\text{S5})$$

$$\psi_1 = \varphi_1 P_1^* / (\varphi_1 P_1^* + \varphi_2 P_2^*) \quad (\text{S6})$$

$$\varphi_1 = 1 - \varphi_2 = x_1 v_1^* / (x_1 v_1^* + x_2 v_2^*) \quad (\text{S7})$$

$$P_1^* = \alpha_1 T \tilde{v}_1^2 [(\kappa_T)_1]^{-1} \quad (\text{S8})$$

1.2 Excess molar enthalpies

According to this theory H^E is due to contributions, namely, interaction and free volume and is given by²

$$H^E = H_{\text{interaction}}^E + H_{\text{free-volume}}^E \quad (\text{S9})$$

where

$$H_{\text{interaction}}^{\text{E}} = \left(\sum_{i=1}^2 x_i U_i^* \right) \left(\frac{\chi_{12}^{**} \theta_2 \psi_1}{P_1^*} \right) \left(-\tilde{U}(\tilde{T}_U) + \tilde{T}_U \tilde{C}_p(\tilde{T}_U) \right) \quad (\text{S10})$$

$$H_{\text{free-volume}}^{\text{E}} = \left(\sum_{i=1}^2 x_i U_i^* \right) \tilde{C}_p(\tilde{T}_U) \left(\psi_1 \tilde{T}_1 + \psi_2 \tilde{T}_2 - \tilde{T}_U \right) \quad (\text{S11})$$

$$\tilde{T}_U = \left(\tilde{v}_U^{1/3} - 1 \right) / \tilde{v}_U^{4/3} \quad (\text{S12})$$

$$\tilde{v}_U = \psi_1 \tilde{v}_1 + \psi_2 \tilde{v}_2 \quad (\text{S13})$$

$$\tilde{v}_i = \left[\frac{\alpha_i T}{3(1 + \alpha_i T)} + 1 \right] \quad (\text{S14})$$

$$\tilde{U}(\tilde{T}_U) = \sum_{i=1}^2 \psi_i \tilde{U}_i \quad (\text{S15})$$

$$\tilde{U}_i = \frac{-1}{\tilde{v}_i} \quad (\text{S16})$$

$$\tilde{C}_p(\tilde{T}_U) = \sum_{i=1}^2 \psi_i \left(\tilde{C}_p \right)_i \quad (\text{S17})$$

$$\left(\tilde{C}_p \right)_i = 3\tilde{v}_i^{1/3} / 4 - 3\tilde{v}_i^{1/3} \quad (\text{S18})$$

$$U_i^* = P_i^* v_i^* \quad (\text{S19})$$

where $i = 1$ or 2 represents components of the (1 +2) mixture.

1.3 Excess isentropic compressibilities

The isentropic compressibility, κ_s is given³ by

$$\kappa_s = -\frac{1}{V} \left(\frac{\partial V}{\partial P} \right)_s \quad (\text{S20})$$

$$\text{where } \left(\frac{\partial V}{\partial P} \right)_s = \left(\frac{\partial V}{\partial P} \right)_T + T C_p^{-1} \left(\frac{\partial V}{\partial T} \right)_p \quad (\text{S21})$$

$\left(\frac{\partial V}{\partial P}\right)_T$ and $\left(\frac{\partial V}{\partial T}\right)_P$ were expressed by

$$\left(\frac{\partial V}{\partial P}\right)_T = \left[\frac{-\tilde{v}^{7/3} + 2\tilde{v}^2 - \tilde{v}^{5/3}}{4/3 - \tilde{v}^{1/3}} \right] \frac{v^*}{P^*} \frac{1}{\tilde{T}} \quad (\text{S22})$$

$$\left(\frac{\partial V}{\partial T}\right)_P = \left[\frac{\tilde{v}^{4/3} - \tilde{v}}{4/3 - \tilde{v}^{1/3}} \right] \frac{v^*}{T} \quad (\text{S23})$$

where

$$P^* = \sum_{i=1}^2 \varphi_i p_i^* - \sum_{i=1}^2 \varphi_i \theta_2 \chi_{12}^{**} \quad (\text{S24})$$

$$v^* = \sum_{i=1}^2 \varphi_i v_i^* \quad (\text{S25})$$

$$\bar{v} = v / v^* \quad (\text{S26})$$

$$\Psi_1 = \frac{\varphi_1 P_1^*}{\sum_{i=1}^2 \varphi_i P_i^*} \quad (\text{S27})$$

$$\tilde{T} = \left[\sum_{i=1}^2 (\varphi_i P_i^* \bar{T}_i / \sum_{i=1}^2 \varphi_i P_i^*) \right] \left[1 - (\varphi_1 \theta_2 \chi_{12}^{**}) (\sum_{i=1}^2 \varphi_i P_i^*)^{-1} \right]^{-1} \quad (\text{S28})$$

where v^* , U^* , $\tilde{v}$, P^* , C_p , ψ , (κ_T) , α are characteristic volume, characteristic configurational energy, reduced volume, characteristic pressure, heat capacity, contact energy fraction, isothermal compressibility and coefficient of volume expansion of pure component; $\tilde{U}$, $\tilde{T}$, $\tilde{C}_p(\tilde{T}_U)$, $\tilde{U}(\tilde{T}_U)$ are the reduced internal energy, reduced temperature, reduced heat capacity and reduced configurational energy of the mixtures and all the terms have the same significance as described in the literature.^{4,5}

The estimation of V^E , H^E and κ_S^E values by PFP theory require knowledge of interaction energy parameter, χ_{12}^{**} along with Flory's parameters of pure liquids. The parameters characteristic volume, V^* , characteristic pressure, P^* , isothermal compressibility, κ_T and coefficient of volume expansion of pure component, α are recorded in Table S2. The κ_T values for D, D' were taken from literature^{6, 7} and for *o*-CT, THP were calculated by employing ΔH_v values (taken from literature⁷) using the expression;

$$\delta^2 = \alpha T / (\kappa_T)_1 \quad (S29)$$

Solubility parameter, δ was calculated by⁸

$$\delta = [(\Delta H_v - RT) / v]^{0.5} \quad (S30)$$

where v is molar volume.

The χ_{12}^{**} parameters for present mixtures were calculated by employing H^E values of equimolar mixtures using:

$$H^E = \sum_{i=1}^2 x_i P_i^* (U_i^{-1} - U_{cal}^{-1}) + x_1 U_1^* \theta_2 \chi_{12}^{**} U_{cal}^{-1} \quad (S31)$$

Such V^E , H^E and κ_S^E data predicted by PFP theory are reported in Table S3 and deviation between experimental values and values obtained from PFP theory along with χ_{12}^{**} have been reported in Table S4. Examination of data in Table S3 revealed that PFP theory correctly predicts the sign of V^E , H^E and κ_S^E values of the investigated mixtures {except for κ_S^E values of *o*-CT (1) + D or D' (2)}.

Table S2. Parameters of the Pure Components i.e Characteristic Volume, V^* , Characteristic Pressure, P^* , Isothermal Compressibility, κ_T and Coefficient of Thermal Expansion, α used in PFP Theory Calculations at $T / K = 298.15, 303.15, 308.15$.

Components	T / K	$V^* / \text{cm}^3 \cdot \text{mol}^{-1}$	$P^* / \text{J} \cdot \text{cm}^{-3}$	$\kappa_T / \text{TPa}^{-1}$	$\alpha(\times 10^{-3}) / \text{K}^{-1}$
<i>o</i> -Chlorotoluene	298.15	95.69	553.71	735.03	0.905
	303.15	95.78	554.58	755.91	0.911
	308.15	95.90	555.20	776.17	0.915
1, 3-dioxolane	298.15	54.44	771.91	758.10 ^a	1.189
	303.15	54.54	772.62	781.21	1.196
	308.15	54.65	773.69	804.42	1.203
1,4-dioxane	298.15	67.60	713.04	738.01 ^b	1.101
	303.15	67.70	713.75	764.51	1.108
	308.15	67.82	714.25	789.81	1.114
Tetrahydropyran	298.15	76.33	545.0	1051.56	1.170
	303.15	76.45	545.8	1085.00	1.179
	308.15	76.60	546.2	1117.57	1.186

^aRef 6. ^b Ref 7.

Table S3. Comparison of Calculated V^E , κ_S^E at $T/K = 298.15, 303.15, 308.15$ and H^E at $T/K = 308.15$ Values from PFP Theory with Their Corresponding Experimental Values.

Properties	Mole fraction of component								
	0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9
<i>o</i> -Chlorotoluene (1) + 1,3-dioxolane(2)									
T/K = 298.15									
$V^E(\text{Expt.})/$ $\text{cm}^3 \cdot \text{mol}^{-1}$	0.032	0.055	0.066	0.070	0.068	0.059	0.048	0.035	0.019
$V^E(\text{PFP})/$ $\text{cm}^3 \cdot \text{mol}^{-1}$	0.140	0.242	0.310	0.346	0.352	0.331	0.283	0.212	0.117
$\kappa_S^E(\text{Expt.})/$ TPa^{-1}	6.4	11.8	15.5	15.7	14.4	12.7	10.2	7.6	4.4
$\kappa_S^E(\text{PFP})/$ TPa^{-1}	-60.2	-58.8	-57.8	-57.2	-56.8	-56.6	-56.5	-56.5	-56.6
T/K = 303.15									
$V^E(\text{Expt.})/$ $\text{cm}^3 \cdot \text{mol}^{-1}$	0.042	0.068	0.083	0.090	0.086	0.076	0.063	0.047	0.028
$V^E(\text{PFP})/$ $\text{cm}^3 \cdot \text{mol}^{-1}$	0.142	0.245	0.314	0.351	0.357	0.335	0.287	0.214	0.118
$\kappa_S^E(\text{Expt.})/$ TPa^{-1}	7.7	13.0	16.4	16.6	15.2	13.4	11.1	8.3	5.3
$\kappa_S^E(\text{PFP})/$ TPa^{-1}	-62.4	-60.8	-59.8	-59.1	-58.7	-58.5	-58.4	-58.4	-58.5
T/K = 308.15									
$V^E(\text{Expt.})/$ $\text{cm}^3 \cdot \text{mol}^{-1}$	0.045	0.078	0.099	0.109	0.103	0.093	0.077	0.057	0.034
$V^E(\text{PFP})/$ $\text{cm}^3 \cdot \text{mol}^{-1}$	0.144	0.250	0.320	0.357	0.364	0.342	0.293	0.219	0.121

$\kappa_s^E(\text{Expt.})/$ TPa ⁻¹	9.4	14.5	17.2	17.4	16.5	14.6	12.4	9.3	6.1
$\kappa_s^E(\text{PFP})/$ TPa ⁻¹	-63.9	-62.3	-61.3	-60.7	-60.2	-60.0	-59.9	-59.9	-60.0
$H^E(\text{Expt.})/$ J.mol ⁻¹	161	278	356	394	395	362	300	216	111
$H^E(\text{PFP})/$ J.mol ⁻¹	170	288	362	397	397	367	309	227	124

o-Chlorotoluene (1) + 1,4-dioxane(2)

T/K = 298.15K

$V^E(\text{Expt.})/$ cm ³ . mol ⁻¹	0.088	0.153	0.188	0.208	0.208	0.198	0.166	0.118	0.062
$V^E(\text{PFP})/$ cm ³ . mol ⁻¹	0.036	0.063	0.080	0.089	0.090	0.085	0.072	0.054	0.030
$\kappa_s^E(\text{Expt.})/$ TPa ⁻¹	2.3	4.3	5.3	6.0	6.0	5.3	4.5	3.5	1.8
$\kappa_s^E(\text{PFP})/$ TPa ⁻¹	-68.5	-66.5	-64.8	-63.3	-62.0	-60.8	-59.8	-58.8	-58.0

T/K = 303.15

$V^E(\text{Expt.})/$ cm ³ . mol ⁻¹	0.100	0.163	0.198	0.212	0.214	0.203	0.170	0.124	0.068
$V^E(\text{PFP})/$ cm ³ . mol ⁻¹	0.037	0.064	0.081	0.090	0.092	0.086	0.073	0.055	0.030
$\kappa_s^E(\text{Expt.})/$ TPa ⁻¹	2.7	4.4	5.5	6.2	6.2	5.5	4.7	3.7	2.1
$\kappa_s^E(\text{PFP})/$ TPa ⁻¹	-70.8	-68.7	-66.9	-65.3	-63.9	-62.7	-61.7	-60.7	-59.8

T/K = 308.15

$V^E(\text{Expt.})/$	0.116	0.170	0.207	0.215	0.218	0.209	0.178	0.134	0.078
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$\text{cm}^3 \cdot \text{mol}^{-1}$									
$V^E(\text{PFP})/$	0.038	0.065	0.083	0.092	0.093	0.087	0.075	0.056	0.031
$\text{cm}^3 \cdot \text{mol}^{-1}$									
$\kappa_S^E(\text{Expt.})/$	3.0	4.6	5.7	6.4	6.3	5.7	4.8	3.7	2.4
TPa^{-1}									
$\kappa_S^E(\text{PFP})/$	-72.8	-70.7	-68.8	-67.2	-65.8	-64.5	-63.4	-62.4	-61.5
TPa^{-1}									
$H^E(\text{Expt.})/$	164	276	344	370	357	316	250	168	82.4
$\text{J} \cdot \text{mol}^{-1}$									
$H^E(\text{PFP})/$	143	247	316	352	358	335	287	213	117
$\text{J} \cdot \text{mol}^{-1}$									

o-Chlorotoluene (1) +THP (2)

T/K=298.15

$V^E(\text{Expt.})/$	-0.075	-0.134	-0.164	-0.183	-0.189	-0.177	-0.152	-0.110	-0.060
$\text{cm}^3 \cdot \text{mol}^{-1}$									
$V^E(\text{PFP})/$	-0.007	-0.013	-0.017	-0.019	-0.019	-0.018	-0.015	-0.011	-0.006
$\text{cm}^3 \cdot \text{mol}^{-1}$									
$\kappa_S^E(\text{Expt.})/$	-7.8	-12.9	-16.5	-18.7	-19.2	-18.2	-15.3	-11.4	-6.4
TPa^{-1}									
$\kappa_S^E(\text{PFP})/$	-84.6	-81.7	-79.0	-76.5	-74.1	-71.8	-69.7	-67.6	-65.7
TPa^{-1}									

T/K=303.15K

$V^E(\text{Expt.})/$	-0.080	-0.141	-0.173	-0.190	-0.192	-0.178	-0.158	-0.115	-0.065
$\text{cm}^3 \cdot \text{mol}^{-1}$									
$V^E(\text{PFP})/$	-0.008	-0.013	-0.017	-0.019	-0.020	-0.018	-0.016	-0.012	-0.006
$\text{cm}^3 \cdot \text{mol}^{-1}$									
$\kappa_S^E(\text{Expt.})/$	-9.0	-14.6	-18.3	-20.3	-20.8	-19.3	-16.3	-12.2	-6.8
TPa^{-1}									

$\kappa_S^E(\text{PFP})/$ TPa^{-1}	-87.3	-84.3	-81.4	-78.8	-76.3	-73.9	-71.7	-69.6	-67.5
T/K = 308.15									
$V^E(\text{Expt.})/$ $\text{cm}^3 \cdot \text{mol}^{-1}$	-0.086	-0.150	-0.181	-0.193	-0.196	-0.190	-0.160	-0.124	-0.071
$V^E(\text{PFP})/$ $\text{cm}^3 \cdot \text{mol}^{-1}$	-0.008	-0.014	-0.018	-0.020	-0.020	-0.019	-0.016	-0.012	-0.006
$\kappa_S^E(\text{Expt.})/$ TPa^{-1}	-9.2	-15.7	-19.6	-21.8	-22.0	-20.2	-17.1	-12.3	-7.0
$\kappa_S^E(\text{PFP})/$ TPa^{-1}	-89.9	-86.7	-83.8	-81.0	-78.4	-76.0	-73.6	-71.4	-69.3
$H^E(\text{Expt.})/$ $\text{J} \cdot \text{mol}^{-1}$	-70.5	-131	-172	-205	-220	-215	-192	-145	-83.5
$H^E(\text{PFP})/$ $\text{J} \cdot \text{mol}^{-1}$	-85.4	-149	-192	-215	-220	-208	-179	-134	-74.0

Table. S4 Values of Interaction Energy, χ_{12}^{} Parameter Utilized in PFP Theory Along With The Deviations $\sigma(V_{\text{PFP}}^E)$, $\sigma(\kappa_{\text{S PFP}}^E)$, $\sigma(H_{\text{PFP}}^E)$ Between Experimental Values and The Values Obtained From PFP Theory.**

Mixture	T / K	Interaction energy parameter $\chi_{12}^{**}/$ $\text{J}\cdot\text{cm}^{-3}$	Deviations		
			$\sigma(V_{\text{PFP}}^E)/$ $\text{cm}^3\cdot\text{mol}^{-1}$	$\sigma(\kappa_{\text{S PFP}}^E)/$ TPa^{-1}	$\sigma(H_{\text{PFP}}^E)/$ $\text{J}\cdot\text{mol}^{-1}$
<i>o</i> -Chlorotoluene (1) + 1,3- dioxolane (2)	298.15	20.40	0.233	72.7	-
	303.15	20.36	0.221	75.7	-
	308.15	20.33	0.213	78.5	9
<i>o</i> -Chlorotoluene (1) +1,4-dioxane(2)	298.15	16.63	0.098	71.0	-
	303.15	16.60	0.104	73.3	-
	308.15	16.56	0.110	75.5	30
<i>o</i> -Chlorotoluene (1) +tetrahydropyran(2)	298.15	-8.90	0.139	64.6	-
	303.15	-8.85	0.143	65.7	-
	308.15	-8.80	0.150	67.1	14

REFERENCES

- (1) Van, H. T.; Patterson, D. Volume of mixing and P^* effect: Part 1. Hexane isomers with normal and branched hexadecane. *J. Solution Chem.* **1982**, *11*, 793–805.
- (2) Galvao, A. C.; Francesconi, A. Z. Applications of the Prignone-Flory-Patterson model to excess molar enthalpy of binary liquid mixtures containing acetonitrile and alkanols. *J. Mol. Liquids* **2008**, *139*, 110–116.
- (3) Oswal, S. L.; Maisuria, M. M. Speed of sound, isentropic compressibilities and excess molar volumes of cycloalkanes, alkanes and aromatic hydrocarbon at 303.15 K. I. Results for cycloalkane + cycloalkanes, and cycloalkane + alkanes. *J. Mol. Liquids* **2002**, *100*, 91–112.
- (4) Flory, P. J. The statistical thermodynamic of liquid mixtures. *J. Am. Chem. Soc* **1965**, *87*, 1833–1838.
- (5) Flory, P. J. The thermodynamic properties of mixture of small non-polar molecules. *J. Am. Chem. Soc.* **1965**, *87*, 1838–1846.
- (6) Giner, I.; Villares, A.; Haro, M.; Gascon, I.; Lafuente, C. Thermodynamic properties of binary mixtures formed by cyclic ethers and chloroalkanes. *J. Therm. Anal. Calorim.* **2007**, *90*, 587–595.
- (7) Riddick, J. A.; Bunger, W. B.; Sakano, T. K. *Organic solvents, physical properties and method of purification*; 4th ed.: Wiley-Interscience: New York, 1986.
- (8) Hildebrand, J. H.; Prusnitz, J. M.; Scott, R. L. *Regular and Related Solutions: The Solubility of Gases, Liquids, and Solids, Van-Nonstand Reinheld Compound*. New York, 1971.