



SCIREA Journal Of Chemistry

ISSN: 2995-6943

<http://www.scirea.org/journal/Chemistry>

March 11, 2025

Volume 10, Issue 1, February 2025

<https://doi.org/10.54647/chemistry150387>

“Study of thermodynamic and acoustic properties of binary liquid mixtures of some cyclic ether with n-alkanols at 298.15K: By Ultrasonic Velocity Measurements”

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Abstract

Liquid-liquid interactions, also known as solvent-solvent interactions, refer to the behavior of two or more liquid substances when they come into contact with each other. These interactions are of paramount importance in the field of chemistry because they underlie various processes in nature and industry. In the present study, we reported the densities and excess molar volumes and (V_m^E) excess acoustical impedance (Z^E) for the pure organic solvent mixtures such as 1,3-Dioxolane + Pentanol, 1,3-Dioxolane + Hexanol, 1,3-Dioxolane + Heptanol, 1,3-Dioxolane + Octanol, 1,3-Dioxolane + Nonanol and 1,3-Dioxolane + Decanol at 298.15K. Excess molar volumes (V_m^E) excess acoustic impedance (Z^E) for these binary mixtures were calculated from the experimental density and sound velocity data and fitted to the Redlich-Kister polynomial equation to check the accuracy of experimental

data. In addition, the interactions between the unlike molecules in the mixtures were confirmed by using the experimental results. It has been observed that, weak dispersive type intermolecular interactions are confirmed in the systems investigated. Dipole inducement is found to be more predominant in the system. The results are interpreted in terms of molecular interaction between the components of the mixtures.

Keywords: Density, Acoustical impedance, 1,3-dioxolane, binary liquid mixture, Molecular interactions, molar volume and Ultrasonic velocity.

Nomenclature

ρ , Densities of liquid

u , Ultrasonic velocity

V_m , molar volume

V_m^E , excess molar volume

Z , acoustic impedance

Z^E , excess acoustic impedance

X_1 , Mole fraction of 1,3-Dioxolane

Y^E , Thermodynamic excess function

INTRODUCTION

Study of acoustic and thermodynamic properties of liquid mixtures plays an important role and it helps us to understand how it behaves when mixed with other compounds. From the past decades, thermodynamic properties of mixtures have become increasingly important in the academic as well as scientific industries which include pharmaceutical, oil or gas for flow assurance, oil recovery and polymer chemistry for solvent selection and emission, and engineering chemistry for the design of separation processes. Besides, in design engineering thermodynamic data is also mandatory for fluid flow and design of pipelines. In addition, the thermodynamic properties are also required for new correlations/predictive and/or analytical models to test the solution theories for the mixtures as well as to afford information about

molecular interactions such as structural breaking or making properties and nature/characterization behavior of the compounds in the mixture, solute-solute, solute-solvent and solvent-solvent. Also, the excess molar volumes are derived from experimental density results which are very valuable to get the information about intermolecular interactions and geometrical effects between the component molecules. Additionally, the study of liquid mixtures involves investigating the acoustic and thermodynamic properties, which provide insights on molecular interactions among different compounds [1]. This study explains the relationship between thermodynamic parameters and the nature of molecular bonds. Therefore, ultrasonic techniques mainly used to explore and understand the characteristics and dynamics of such molecular interactions [2–5].

Likewise, these excess thermodynamic properties of liquid mixtures play a significant role to understand specific interactions such as differences in the molecular size, shape, structural effects arising from interstitial accommodation and the continuation of solution theories because they depend on the intermolecular interactions such as hydrogen donor or acceptor specifically in binary mixtures dipole-dipole and van der Waal interactions. In the present investigation, the preferred organic solvents with numerous industrial applications have been chosen. 1,3-Dioxolane is used as a solvent and industrial feedstock. 1-Alkanol is a good solvent for plasticizer and as an intermediate for textile and leather finishing agents. The present paper is a part of our ongoing research program in the measurement of thermodynamic and transport properties of binary liquid mixtures. The liquids were chosen in the present investigation on the basis of their industrial importance. 1,3-dioxolane (cyclic diether) have played a major role in the pharmaceutical chemistry. Therefore, the applications of these compounds attract us to study their behavior in alcohols. Alcohols are used as hydraulic fluids in pharmaceutical and cosmetics, in medications for animals, in manufacturing of perfumes, paint removers, flavors and dyestuffs, as defrosting and as an antiseptic agent. The experimental results have been used to discuss the nature of interaction between unlike molecules in terms of hydrogen bonding, dipole-dipole interactions and dispersive forces. It is well known that ethers interact with alcohols by dipole-dipole interaction, formation of new hydrogen bonds or hetero-associations and dispersion forces. Ultrasonic velocity and viscosity measurements have been widely used in the field of interactions and structural aspect evaluations studies. In the present work, an attempt has been made to investigate the behavior of binary solutions of 1,3-dioxolane in alkanols with regard to adiabatic compressibility, specific acoustical impedance (Z) and molar volume (V_m) from

ultrasonic measurements at 298.15K. The results are interpreted in terms of molecular interaction between the components of the mixtures.

2. EXPERIMENTAL PROCEDURE

2.1 Chemicals. The source and purity of the chemical compound are shown in table-1. The substances density, viscosity and ultrasonic velocity is compared with the literature data (Table-2) to ascertain the purity, and a good agreement between the experimental data and literature data [6-13] was observed.

2.2 Apparatus and Procedure: All six binary liquid mixtures were prepared by weighing appropriate amounts of pure liquids on a digital electronic balance (Citizen Scale (I) PVT. LTD. Mumbai, India.) with a precision ± 0.1 . The experimental uncertainty in mole fractions did not exceed ± 0.0005 . All the solutions were prepared by mass ratios and stored in the air-tight stopper measuring flasks.

Table 1. CAS Registry Number, Mass Fraction Purity of the chemicals

Component	Formula	CAS Reg. No.	Supplier	Mass Fraction Purity (%)	Method Purity analysis method
1,3-Dioxolane	C ₃ H ₆ O ₂	646-06-0	CDH Delhi	99.7	Chromatography by the supplier
Pentanol	C ₅ H ₁₂ O	71-41-0	CDH Delhi	99.7	Chromatography by the supplier
Hexanol	C ₆ H ₁₄ O	111-27-3	CDH Delhi	99.5	Chromatography by the supplier
Heptanol	C ₇ H ₁₆ O	111-70-6	CDH Delhi	99	Chromatography by the supplier
Octanol	C ₈ H ₁₈ O	111-87-5	CDH Delhi	99.7	Chromatography by the supplier
Nonanol	C ₉ H ₂₀ O	143-08-8	CDH Delhi	99	Chromatography by the supplier
Decanol	C ₁₀ H ₂₂ O	112-30-1	CDH Delhi	99	Chromatography by the supplier

Table 2. Comparison of Experimental and Literature density (ρ), and sound velocity (u) of pure Components with Available Literature Values at T = 298.15K.

Compound	ρ (g.cm ⁻³)		u (m.s ⁻¹)	
	This work	Literature	This work	Literature
1,3-Dioxolane	1.0616	1.0577 ⁹	1340	1338 ⁹
		1.0586 ⁹		1338 ¹⁰

Pentanol	0.8124	0.8108 ⁶	1198	1197 ⁸
		0.8107 ⁶		1268 ¹²
Hexanol	0.8176	0.8187 ⁶	1306	1304 ⁷
		0.8152 ⁷		1303 ⁷
Heptanol	0.8196	0.8187 ¹⁰	1325	1327 ⁷
		0.8197 ¹¹		1327.37 ¹³
Octanol	0.8236	0.8216 ⁶	1350	1348 ¹³
		0.8218 ⁶		1347 ¹²
Nonanol	0.8248	0.8244 ⁷	1366	1365 ⁷
		0.824224 ⁷		1364 ¹²
Decanol	0.8292	0.8267 ⁷	1378	1380 ⁷
		0.8264 ¹¹		1379 ¹²

2.3 Measurements:

Density: Densities of pure components and liquid-liquid mixtures were measured with a 25-ml specific gravity bottle by relative measurement method with an accuracy of $\pm 0.01 \text{ kg.m}^{-3}$. The specific gravity bottle with the experimental mixture was immersed in the temperature-controlled water bath (MSI Goyal scientific, Meerut, U.P. India.), operating in the temperature range of -10°C to 85°C with an accuracy $\pm 0.1^{\circ}\text{C}$. Double distilled water used for the calibration of the specific gravity bottle. At least three times for each composition in experimental were generally repeated and the results were treatment.

Sound velocity:

The speed of sound (u) was measured at a frequency 3 MHz in these solutions using the interferometric method with a (Model F-80D, Mittal Enterprise, New Delhi, India) at 298.15K. The interferometer cell was filled with the test liquid, and water was circulated around the measuring cell from a water bath. The uncertainty was estimated to be $\pm 0.1\%$. The measured values of ultrasonic velocities of pure 1,3-dioxolane with pentanol, Hexanol, heptanol, octanol, nonanol and decanol compare well with the corresponding literature values.

Theoretical:

The experimentally measured density (ρ) and ultrasonic velocity (u), are used to evaluate derived parameters like molar volume (V_m), and acoustic impedance (Z) using well established relations.

The molar volume (V_m) of binary liquid mixtures were calculated by using a following equation:

$$V_m = \frac{(X_1M_1 + X_2M_2)}{\rho} \dots\dots\dots(1)$$

The acoustic impedance is the parameter related to elastic properties of the medium and calculated by using the expression

$$Z = \rho \cdot U \dots\dots\dots (2)$$

where ρ is the density and u is the sound velocity.

The excess value of ultrasonic related parameters has been calculated by using the following relation

$$A^E = A_{exp.} - (X_1 A_1 + X_2 A_2) \dots\dots\dots (3)$$

Where A represents the parameter such as molar volume, and acoustic impedance and X_1 and X_2 is the mole fractions of components whose parameters

RESULT AND DISCUSSION

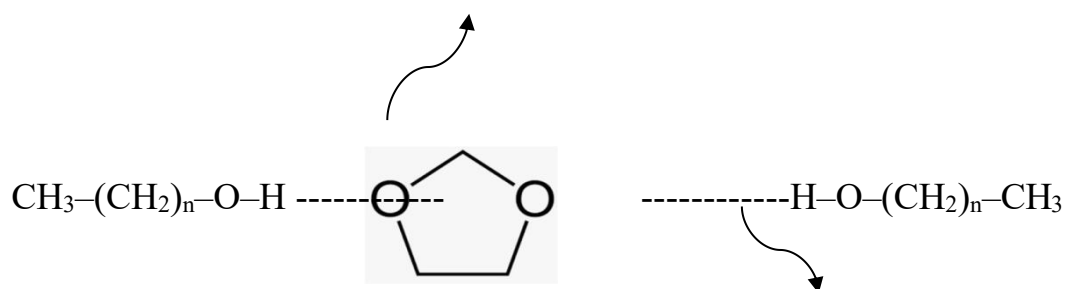
The experimentally determined values of density (ρ) and sound velocity (u) and derived parameters molar volume (V_m) and acoustic impedance (Z) at 298.15K for the binary liquid system 1,3-Dioxolane with Pentanol, Hexanol, Heptanol, Octanol, Nonanol and Decanol are listed in table-3. The same excess values for the binary liquid mixtures 1,3-Dioxolane with Pentanol, Hexanol, Heptanol, Octanol, Nonanol and Decanol are presented in table-3. The data related to excess molar volume and excess acoustic impedance for the binary liquid system 1,3-Dioxolane with Pentanol, Hexanol, Heptanol, Octanol, Nonanol and Decanol were graphically represented in figures 1 to 2 at 298.15K respectively. From the table-3, it was observed that the density and ultrasonic velocity increase with increasing mole fraction of 1,3-Dioxolane. This may be due to association of a very strong dipole- induced dipole interaction between the component molecules.

Excess Molar Volume (V_m^E)

The excess molar volume (V_m^E) data of all the binary mixtures of 1,3-Dioxolane with Pentanol, Hexanol, Heptanol, Octanol, Nonanol and Decanol are graphically presented Figures 1 at 298.15 K. A perusal of table-3 indicates that the values of excess molar volume (V_m^E) data for the binary mixtures of 1,3-Dioxolane with Pentanol, Hexanol, Heptanol, Octanol, Nonanol and Decanol are positive. On other hand, an inversion in sign for the binary mixtures containing 1,3-Dioxolane with Pentanol, Hexanol, Heptanol, Octanol, Nonanol and Decanol was observed. Further, it is observed the magnitude of positive excess molar volume (V_m^E) values decrease with increase in composition of 1,3-Dioxolane. According to Marcus

[14], the molecules of alkanols are associated through hydrogen bonding in pure state. Mixing these alcohol molecules with polar molecule like 1,3-Dioxolane would induce mutual dissociation of the hydrogen-bonded structure present in pure alcohols with subsequent formation of inter molecular hydrogen bonds (O-----OH) between the oxygen atom of ether group of 1,3-Dioxolane molecule and hydrogen atom of hydroxyl group of alcohols.

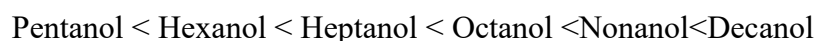
Hydrogen bonding



Hydrogen bonding

Figure: 1,3-dioxolane – 1-alkanols interactions are due to hydrogen bonding between the oxygen atom of the dicyclic- ether and the proton of hydrogen group of n-alkanols.

The positive excess molar volume (V_m^E) values suggest that the higher alcohols less proton donating ability than the lower alcohols. Hence hetero association affects decreases in the binary liquid mixtures with an increase of chain length of linear alcohols. The algebraic values of excess molar volume (V_m^E) for the mixtures of 1,3-Dioxolane with Pentanol, Hexanol, Heptanol, Octanol, Nonanol and Decanol fall in the order,



The order suggests that dipole-dipole interaction between unlike molecules decrease with increasing in chain length of alcohols as consequences of these decreases the degree of polymerization in the pure state. These experimental results suggest that the positive excess molar volume (V_m^E) indicate that, the breaking up off self-associated structure of the components of the mixtures is dominant over the effect of H-bonding and dipole-dipole interaction between unlike molecules.

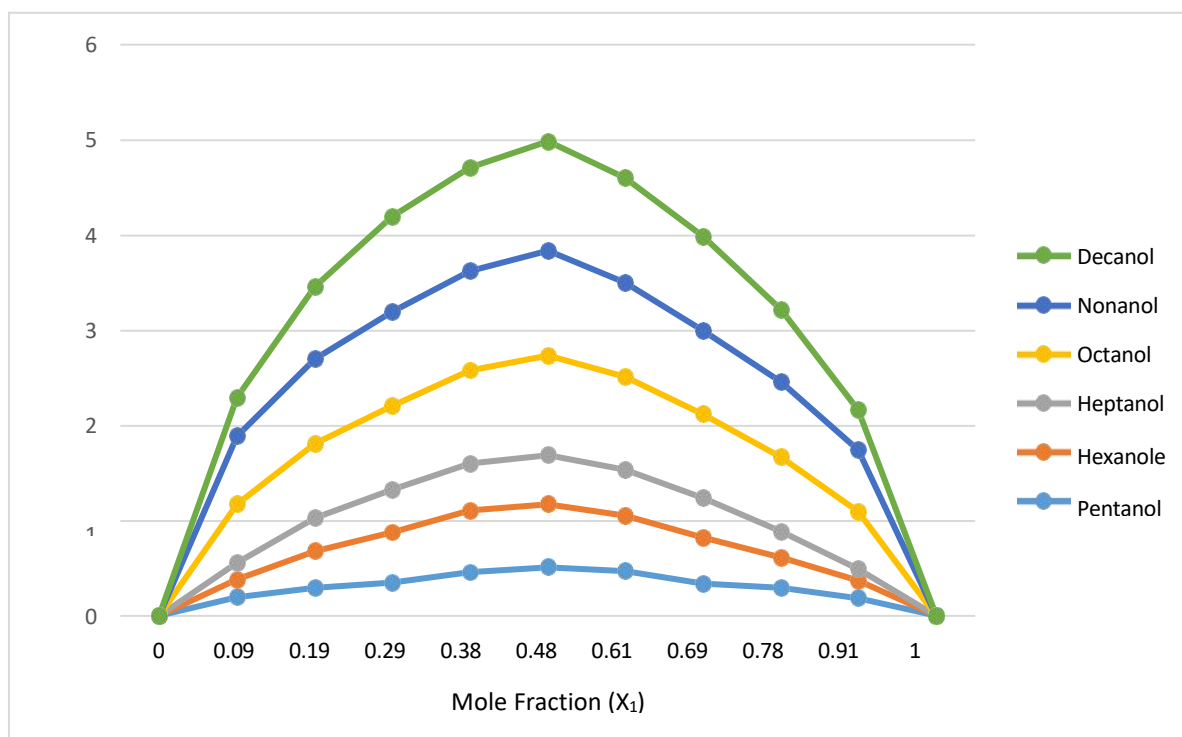


Figure 1. Curves of excess molar volume (V_m^E) against the mole fraction of 1,3-dioxolane x_1 , for the binary mixture (1,3-dioxolane (1) + Alkanols(2)) at 298.15K. The solid lines represent the values calculated from the Redlich–Kister equation

Acoustic impedance (Z)

The excess acoustic impedance (Z^E) data of all the binary mixtures of 1,3-Dioxolane with Pentanol, Hexanol, Heptanol, Octanol, Nonanol and Decanol are graphically presented Figures-2 at 298.15 K. An examination of curves in Figure- 2 shows that the values of excess acoustic impedance (Z^E) data for 1,3-Dioxolane with Pentanol, Hexanol, Heptanol, Octanol, Nonanol and Decanol are negative over the entire composition range at 298.15 K. The variation of excess acoustic impedance (Z^E) with mole fraction of 1,3-Dioxolane at 298.15K are shown in figure-2. It is observed the excess acoustic impedance (Z^E) data for 1,3-Dioxolane with Pentanol, Hexanol, Heptanol, Octanol, Nonanol and Decanol are negative over the entire composition range at 298.15 K. This is in agreement with requirement as both ultrasonic velocity and density increase with increase in concentration of the solute and also effective due to solute – solvent interaction.

The excess acoustic impedance Z^E values in all the binary mixtures are throughout negative tables3, figure 2. Trends in Z^E values figure 2 reinforce our earlier view that there is the possibility of specific intermolecular reactions in all the mixtures. Negative values of Z^E in all

mole fraction concentration further hints to the possibility of the presence of strong attractive forces in between the reacting components of the mixture. At this point when considering all the liquid mixtures, it is found that the maximum intermolecular association in all the binary mixtures takes place in between ~ 0.4 to ~ 0.6 mole fraction of alkanol concentration range. From figure 2 it is quite evident that as the mole fraction concentration of 1-alkanol increases in the mix, the magnitude of negative value starts decreasing. This can be attributed to the fact that with the increasing concentration of 1-alkanol, the number of similar (like–like) molecules of alkanol increases in the mixture leading to the origination of dispersive kind of forces in the system.

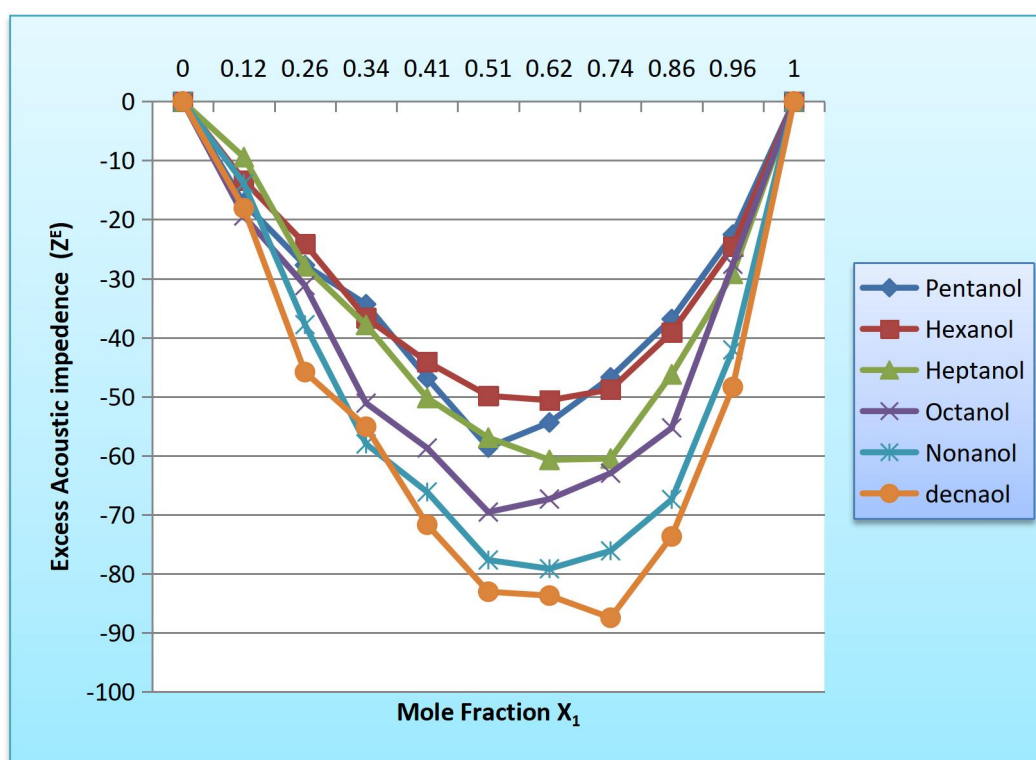


Figure 2. Curves of excess acoustic impedance (Z^E) against the mole fraction of 1,3-dioxolane (x_1), for the binary mixture (1,3-dioxolane (1) + Alkanols(2)) at 298.15K. The solid lines represent the values calculated from the Redlich–Kister equation.

CONCLUSIONS

From experimental results, negative excess molar volume (V_m^E) and excess acoustic impedance (Z^E) values can be attributed to the dipole–dipole interactions between unlike molecules through hydrogen bonding and positive values indicate that the effect due to breaking up of

self-associated structures of the components of the mixtures is dominant over the effect of H-bonding and dipole-dipole interaction between unlike molecules. The positive values of Excess molar volume (V_m^E) and excess acoustic impedance (Z^E) may be attributed to the formation of hydrogen bonding (O-H...O) resulting in the formation of complexes between the component molecules and negative values suggesting breaking of the self-associated alcohols and weak interactions between unlike molecules. From these data, several thermodynamic excess functions have been calculated and correlated using the Redlich – Kister type polynomial equation. The sign and magnitude of these quantities have been discussed in terms of hydrogen bond, electron-transfer complexes and dipole-dipole interactions between the component molecules. Thus based on the above interpretations, 1,3-dioxolane-1-alkanols seemingly appears to be a better binary mixture (based on thermodynamic predictions), in which maximum dispersion of the two components may be predicted.

Acknowledgement:

The authors thank to Uttar Pradesh Council of Science and Technology, Lucknow (No. CST/CHEM/D-648) for financial support (Project ID:3409).

Table 3. Experimental Values of density (ρ) and sound velocity (u) derived parameters molar volume (V_m), acoustic impedance (Z), excess molar volume (V_m^E) and excess acoustic impedance (Z^E) for the binary mixtures of 1,3-Dioxolane (1) + Alkanols (2) at 298.15K.

Mole fraction 1,3-Dioxolane (x_1)	Density (ρ) g.cm ⁻³	Sound velocity (u) ms ⁻¹	Molar volume (V_m) $\times 10^{-3}$ cm ³ .mole ⁻¹	Acoustic impedance (Z) $\times 10^{-4}$ g.cm..s ⁻¹	Excess molar volume (V_m^E) $\times 10^{-3}$ cm ³ .mole ⁻¹	Excess acoustic impedance (Z^E) $\times 10^{-4}$ g.cm..s ⁻¹
1,3-Dioxolane + Pentanol						
0	0.8124	1198	0.1085	0.0973	-	-
0.0939	0.8276	1284	0.1049	0.1062	0.0469	47.19498
0.1942	0.8436	1290	0.1012	0.1088	0.2682	27.73692
0.2941	0.8640	1296	0.0972	0.1119	0.1192	14.35296
0.3942	0.8836	1300	0.0934	0.1148	0.2446	-1.68484
0.4787	0.9068	1304	0.0897	0.1182	0.1860	-5.86255
0.5999	0.9316	1310	0.0855	0.1220	0.2867	-22.3876

0.6972	0.9596	1318	0.0816	0.1264	0.1314	-21.7466
0.7928	0.9876	1324	0.0779	0.1307	0.1568	-21.869
0.9035	1.0260	1332	0.0735	0.1366	0.0076	-12.5556
1.0000	1.0616	1340	0.0697	0.1422	-	-
1,3-Dioxolane + Hexanol						
0	0.8176	1306	0.1249	0.1067	-	-
0.0912	0.8252	1317	0.1207	0.1086	0.7779	-13.3654
0.1955	0.8432	1320	0.1146	0.1113	0.4810	-24.1169
0.2923	0.8584	1322	0.1094	0.1134	0.6245	-36.6767
0.3982	0.8792	1325	0.1034	0.1164	0.4952	-44.1104
0.4942	0.8992	1327	0.0981	0.1193	0.4919	-49.8688
0.6059	0.9264	1330	0.0919	0.1232	0.3860	-50.6217
0.6976	0.9508	1332	0.0868	0.1266	0.3783	-48.7995
0.8018	0.9836	1335	0.0809	0.1313	0.2564	-39.1249
0.8914	1.0168	1337	0.0758	0.1359	0.0816	-24.5556
1.0000	1.0616	1340	0.0697	0.1422	-	-
1,3-Dioxolane + Heptanol						
0	0.8196	1325	0.1417	0.1085	-	-
0.0928	0.8304	1334	0.1352	0.1107	0.1293	-9.44037
0.1905	0.8412	1334	0.1286	0.1122	0.5357	-27.9265
0.2939	0.8592	1335	0.1208	0.1147	0.2168	-37.8571
0.3894	0.8740	1335	0.1141	0.1166	0.4437	-50.2419
0.4818	0.8916	1336	0.1075	0.1191	0.4770	-56.9538
0.6021	0.9184	1337	0.0989	0.1227	0.4818	-60.7204
0.6952	0.9420	1337	0.0922	0.1259	0.5437	-60.5022
0.7892	0.9756	1338	0.0850	0.1305	0.0752	-46.2414
0.9006	1.0156	1339	0.0770	0.1359	0.1264	-29.2001
1.0000	1.0616	1340	0.0697	0.1422	-	-
1,3-Dioxolane + Octanol						
0	0.8296	1350	0.1581	0.1111	-	-
0.0885	0.8296	1350	0.1509	0.1119	0.6846	-19.3955

0.1967	0.8464	1349	0.1408	0.1141	0.0682	-31.1779
0.2998	0.8560	1348	0.1324	0.1153	0.8340	-51.1151
0.3902	0.8712	1348	0.1243	0.1174	0.6824	-58.7113
0.4963	0.8876	1348	0.1153	0.1196	1.0462	-69.5677
0.6008	0.9140	1347	0.1055	0.1231	0.5270	-67.3609
0.6925	0.9340	1348	0.0978	0.1259	0.8545	-67.9767
0.7975	0.9676	1348	0.0883	0.1304	0.6410	-55.3057
0.8940	1.0104	1348	0.0792	0.1362	0.0624	-27.5923
1.0000	1.0616	1340	0.0697	0.1422	-	-
1,3-Dioxolane + Nonanol						
0	0.8248	1366	0.1749	0.1126	-	-
0.0876	0.8336	1366	0.1656	0.1138	-0.0127	-13.8972
0.1913	0.8404	1363	0.1556	0.1145	0.8880	-37.811
0.2942	0.8504	1359	0.1453	0.1155	1.3824	-58.0273
0.3963	0.8692	1355	0.1339	0.1177	0.7277	-66.163
0.4959	0.8844	1352	0.1237	0.1195	0.9917	-77.6885
0.6050	0.9092	1349	0.1119	0.1226	0.6632	-79.1657
0.6947	0.9332	1346	0.1023	0.1256	0.4673	-76.1285
0.7993	0.9648	1343	0.0913	0.1295	0.5023	-67.4371
0.9013	1.0084	1340	0.0803	0.1351	0.1749	-42.0859
1	1.0616	1340	0.0697	0.1422	-	-
1,3-Dioxolane + Decanol						
0	0.8292	1378	0.1908	0.1142	-	-
0.0881	0.8364	1374	0.1803	0.1149	0.2115	-18.0838
0.191	0.8396	1370	0.1693	0.1150	1.6598	-45.848
0.2921	0.8560	1366	0.1561	0.1169	0.7076	-55.1023
0.3937	0.8672	1362	0.1442	0.1181	1.1236	-71.7103
0.4956	0.8824	1358	0.1320	0.1198	1.2487	-83.06
0.604	0.9076	1353	0.1183	0.1227	0.6457	-83.7183
0.7129	0.9308	1348	0.1055	0.1254	1.0260	-87.4645
0.7983	0.9616	1344	0.0946	0.1292	0.5040	-73.6965

0.8971	1.0040	1340	0.0824	0.1345	0.1778	-48.3816
1	1.0616	1340	0.0697	0.1422	-	-

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