

# Studies on Transport and Thermodynamic Properties of Binary Mixtures of Hexan-1-ol with Halogenated Compounds at 293.15 K

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Viscosities ( $\eta$ ) of binary mixtures of hexan-1-ol with 1,2-dichloroethane, 1,2-dibromoethane, and 1,1,2,2-tetrachloroethene have been determined at 293.15 K and at the atmospheric pressure for the whole composition range. Deviations in viscosity ( $\Delta\eta$ ) from the mole fraction average have been derived from the experimental data. The viscosity data have been correlated using Kendall–Monroe, Lobe, Grunberg–Nissan, Tamura–Kurata, Hind–McLaughlin–Ubbelohde, and Katti–Chaudhary viscosity models and McAllister's three-body interaction model.

## Introduction

The measurements of transport and thermodynamic properties of binary mixtures of alcohols and halogenated compounds have been employed in understanding the nature of molecular systems and physicochemical behavior of binary and multicomponent liquid mixtures.<sup>1–9</sup> These physicochemical properties are of great significance in the selection of appropriate solvents<sup>10</sup> for (i) gas and liquid chromatography, (ii) dissolution of materials, and (iii) their use in propellants and aerosols. The nonrectilinear behavior of the transport, thermodynamic, and acoustic parameters of the liquid mixtures with varying mole fractions has been, in fact, related to the differences in molecular size, shape, and the strength of intra- and intermolecular interactions.<sup>11,12</sup> In an attempt to explore the nature of the interactions occurring between the mixing components, we report here the viscosities ( $\eta$ ) of binary mixtures of hexan-1-ol with 1,2-dichloroethane, 1,2-dibromoethane, and 1,1,2,2-tetrachloroethene at 293.15 K and at the atmospheric pressure for the whole composition range. The deviations in viscosity ( $\Delta\eta$ ) from the mole fraction average have been derived from the experimental data<sup>3</sup> and have been fitted to the Redlich–Kister polynomial equation<sup>13</sup> to derive the coefficients ( $A_i$ ) and the standard errors ( $\sigma$ ) between the experimental and the calculated quantities, and the results have been analyzed in terms of molecular interactions and structural effects. The viscosity data have been correlated using Kendall–Monroe,<sup>14</sup> Vittal Prasad et al.,<sup>15</sup> Grunberg–Nissan,<sup>16</sup> Tamura–Kurata,<sup>17</sup> Hind–McLaughlin–Ubbelohde,<sup>18</sup> and Katti–Chaudhary<sup>19</sup> viscosity models and McAllister's three-body interaction model.<sup>20</sup>

The present communication helps to correlate the density, viscosity, and the other thermodynamic parameters of binary mixtures in terms of the properties of pure liquid components.

## Experimental Section

**Materials and Methods.** The mole fraction purity of the liquids from S.D. Fine Chem Ltd. were as follows: 1,2-dichloroethane (99.7 %), 1,2-dibromoethane (99.5 %), 1,1,2,2-tetrachloroethene (99.4 %), and hexan-1-ol (99.7 %). Prior to experimental measurements, all liquids were stored in dark

bottles over 0.4 nm molecular sieves to reduce water content and were partially degassed with a vacuum pump under nitrogen atmosphere. The purities of all of the samples determined by chromatographic analysis were better than 99.5 % on a mole fraction basis. Density of the pure liquids and their mixtures at 293.15 K were measured with an Anton Paar digital densimeter (model DSA 5000) provided with automatic viscosity correction and two integrated Pt 100 platinum thermometers operated in the static mode and automatically thermostatted within  $\pm 0.001$  K with a solid-state thermostat. Before each series of measurements, the instrument was calibrated with triple-distilled freshly degassed water and dry air at atmospheric pressure. Densities of both water and dry air at the various working temperatures were supplied by the manufacturer in the instruction manual. The calibration was accepted if the measurements were within  $2 \cdot 10^{-6} \text{ g} \cdot \text{cm}^{-3}$  of the published values. The uncertainties were estimated to be within  $\pm 2 \cdot 10^{-6} \text{ g} \cdot \text{cm}^{-3}$ . Air-tight stoppered bottles were used for the preparation of the mixtures. The mass of the dry bottle was first determined, and the less volatile component of the mixture was introduced in the bottle; the total mass was measured. Subsequently, the other liquid component was introduced, and the mass of bottle along with the two components was determined. All of the mixtures were weighed on an electronic balance accurate to 0.1 mg. The average uncertainty in the mole fraction of the mixtures was found to be less than  $\pm 1 \cdot 10^{-4}$ . The densities have a precision better than  $\pm 10^{-6} \text{ g} \cdot \text{cm}^{-3}$ . The densities ( $\rho$ ) of the pure liquids were in good agreement with the values found in the literature<sup>3,10</sup> (Table 1). The densities of the pure liquids and their mixtures are presented in Table 2.

Kinematic viscosities ( $\nu$ ) at 293.15 K and at atmospheric pressure were measured with a modified Ubbelohde suspended-level viscometer. The viscometer was suspended in a thermostatted water bath maintained to  $\pm 0.01$  K. An electronic digital stopwatch with an uncertainty of  $\pm 0.01$  s was used for flow time measurements. At least four flow time measurements were performed for each composition and temperature, and the results were averaged. Calibration of the viscometer was carried out with high-purity cyclohexane (99.5 %) and toluene (99.7 %). The compositions of the mixture are given in mole fraction and were determined by using an analytical balance with an accuracy of  $\pm 0.0001$  g. The possible uncertainty in the mole fractions

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**Table 1. Physical Properties of Pure Liquid Components**

pure liquid	<i>T</i>	$\rho/\text{g}\cdot\text{cm}^{-3}$		$\eta/\text{mPa}\cdot\text{s}$	
	K	expt	lit	expt	lit
hexan-1-ol	293.15	0.818856	0.81875 <sup>a</sup>	5.291	4.592 <sup>a</sup> (298.15 K)
1,2-dichloroethane	293.15	1.252766	1.25209 <sup>a</sup>	0.827	0.779 <sup>b</sup> (298.15 K)
1,2-dibromoethane	293.15	2.179591	2.1791 <sup>a</sup>	1.733	1.629 <sup>c</sup> (298.15 K)
1,1,2,2-tetrachloroethane	293.15	1.622745	1.62283 <sup>a</sup>	0.890	0.932 <sup>a</sup> (288.15 K)

<sup>a</sup> Ref 10. <sup>b</sup> Ref 3. <sup>c</sup> Ref 4.**Table 2. Density ( $\rho$ ), Viscosity ( $\eta$ ), and Viscosity Deviation ( $\Delta\eta$ ) for Halohydrocarbons (1) + Hexan-1-ol (2) at 293.15 K**

$x_1$	$\phi_1$	$\rho$ kg·m <sup>-3</sup>	$\eta$ mPa·s	$\Delta\eta$ mPa·s
1,2-Dichloroethane (1) + Hexan-1-ol (2) (293.15 K)				
0.0000	0.0000	818.86	5.291	0.000
0.0370	0.0237	828.77	5.008	-0.118
0.0769	0.0501	839.85	4.696	-0.252
0.1499	0.1004	860.58	4.191	-0.432
0.2147	0.1475	879.73	3.787	-0.546
0.2935	0.2082	904.95	3.292	-0.689
0.3952	0.2926	940.48	2.726	-0.802
0.4532	0.3441	962.11	2.405	-0.863
0.5596	0.4458	1005.05	1.894	-0.899
0.6113	0.4989	1027.55	1.677	-0.885
0.6971	0.5930	1067.98	1.372	-0.807
0.7791	0.6907	1111.06	1.137	-0.677
0.8563	0.7905	1155.16	0.977	-0.492
0.9335	0.8989	1205.43	0.873	-0.251
0.9790	0.9672	1236.99	0.833	-0.088
1.0000	1.0000	1252.77	0.827	0.000
1,2-Dibromoethane (1) + Hexan-1-ol (2) (293.15 K)				
0.0000	0.0000	818.86	5.291	0.000
0.0402	0.0281	856.61	4.925	-0.224
0.0682	0.0481	883.41	4.703	-0.346
0.1276	0.0918	941.92	4.275	-0.562
0.2056	0.1517	1022.36	3.781	-0.779
0.2686	0.2023	1090.47	3.412	-0.924
0.3892	0.3056	1228.97	2.862	-1.045
0.4277	0.3405	1276.43	2.711	-1.059
0.5444	0.4522	1427.13	2.325	-1.029
0.6405	0.5517	1562.00	2.070	-0.943
0.7326	0.6543	1701.64	1.877	-0.808
0.8519	0.7989	1899.69	1.726	-0.534
0.9273	0.8981	2036.52	1.690	-0.302
0.9587	0.9413	2096.51	1.692	-0.188
0.9782	0.9687	2135.29	1.710	-0.100
1.0000	1.0000	2179.59	1.733	0.000
1,1,2,2-Tetrachloroethane (1) + Hexan-1-ol (2) (293.15 K)				
0.0000	0.0000	818.86	5.291	0.000
0.0688	0.0571	864.64	4.756	-0.233
0.1230	0.1030	901.52	4.351	-0.399
0.2457	0.2106	987.76	3.493	-0.717
0.3618	0.3171	1072.99	2.772	-0.927
0.4502	0.4014	1140.46	2.308	-1.001
0.5583	0.5086	1226.30	1.853	-0.981
0.6518	0.6052	1303.62	1.534	-0.888
0.7068	0.6638	135.05	1.382	-0.799
0.7419	0.7019	138.12	1.297	-0.729
0.7797	0.7435	1414.65	1.213	-0.646
0.8315	0.8016	1461.44	1.116	-0.515
0.8718	0.8478	1498.80	1.051	-0.403
0.9179	0.9015	1542.25	0.982	-0.270
0.9504	0.9401	1573.61	0.942	-0.166
1.0000	1.0000	1622.75	0.890	0.000

is estimated to be less than  $\pm 10^{-4}$ . The equation for viscosity, according to Poiseuille's law, is

$$\eta = \rho\nu = \rho(kt - c/t) \quad (1)$$

where  $k$  and  $c$  are the viscometer constants and  $t$ ,  $\eta$ , and  $\nu$  are the efflux time, dynamic viscosity, and kinematic viscosity, respectively. The uncertainty in the viscosity measurements was of the order of  $\pm 2 \cdot 10^{-3}$  mPa·s.

## Results

The experimental values of viscosity ( $\eta$ ) of the binary mixtures at 293.15 K and at the atmospheric pressure are listed in Table 2. The deviations in viscosity ( $\Delta\eta$ ) from a linear dependence have been calculated from the mole fraction average<sup>3</sup> using the equation

$$\Delta\eta = \eta_{\text{mix}} - (x_1\eta_1 + x_2\eta_2) \quad (2)$$

where  $x_1$ ,  $x_2$ ,  $\eta_1$ , and  $\eta_2$  refer to the mole fraction and the viscosity of pure components and  $\eta_{\text{mix}}$  is the viscosity of the mixture.

The values of  $\Delta\eta$  have been fitted to the Redlich–Kister equation<sup>13</sup> of the type

$$\Delta\eta = x_1(1 - x_1) \sum_{k=1}^n A_k(1 - 2x_1)^{k-1} \quad (3)$$

where  $k$  is the number of estimated parameters and  $A_i$ , and the polynomial coefficients were obtained by fitting the equation to the experimental results by least-squares regression method. The standard deviations,  $\sigma$ , were calculated using the relation<sup>13</sup>

$$\sigma(\eta) = \left[ \frac{\sum (\eta_{\text{expt}} - \eta_{\text{calc}})^2}{N - n} \right]^{1/2} \quad (4)$$

where  $N$  is the number of data points and  $n$  is the number of coefficients. The calculated values of the polynomial coefficients  $A_1$ ,  $A_2$ ,  $A_3$ ,  $A_4$ ,  $A_5$ , and  $A_6$  along with their standard deviations,  $\sigma$ , are presented in Table 3. The curves presented in Figure 1 are the calculated values, and the points represent the experimental values.

## Discussion

The values of  $\Delta\eta$  obtained from eq 2 for the binary mixtures of hexan-1-ol with 1,2-dichloroethane, 1,2-dibromoethane, and 1,1,2,2-tetrachloroethane at 293.15 K are graphically represented as a function of mole fraction of halohydrocarbons in Figure 1. It has been observed that the  $\Delta\eta$  values are negative over the entire composition range for all of the studied systems. The negative values of  $\Delta\eta$  at the minimum of the curves vary in the order:  $\Delta\eta_{\text{DBE-Hex}} > \Delta\eta_{\text{TCE-Hex}} > \Delta\eta_{\text{DCE-Hex}}$ . Since the molar volumes of hexan-1-ol ( $124.78 \cdot 10^{-6}$  m<sup>3</sup>·mol<sup>-1</sup>), 1,2-dichloroethane ( $78.99 \cdot 10^{-6}$  m<sup>3</sup>·mol<sup>-1</sup>), 1,2-dibromoethane ( $86.19 \cdot 10^{-6}$  m<sup>3</sup>·mol<sup>-1</sup>), and 1,1,2,2-tetrachloroethane ( $102.19 \cdot 10^{-6}$  m<sup>3</sup>·mol<sup>-1</sup>) at 293.15 K differ considerably, it appears that the halohydrocarbon molecules intercalate between the polymeric entities of hexan-1-ol and disrupt their H-bonding.<sup>3</sup> The new possible H-bond interactions of the type Cl···H–O, Br···H–O, and  $\pi$ -bond interaction of the type OH··· $\pi$  and weak physical intermolecular interactions in the 1,1,2,2-tetrachloroethane + hexan-1-ol system are weaker than the dipolar interactions in pure halohydrocarbon and H-bonding in hexan-1-ol.<sup>21–24</sup>

The intermolecular interaction strength of hexan-1-ol and the halohydrocarbons may be related to the number of halogen atoms present in the halohydrocarbons.<sup>1–3</sup> Because of the

**Table 3. Coefficients of Redlich–Kister Equation and Standard Deviations for Viscosity Deviations ( $\Delta\eta$ ) for Halohydrocarbons (1) + Hexan-1-ol (2) at 293.15 K**

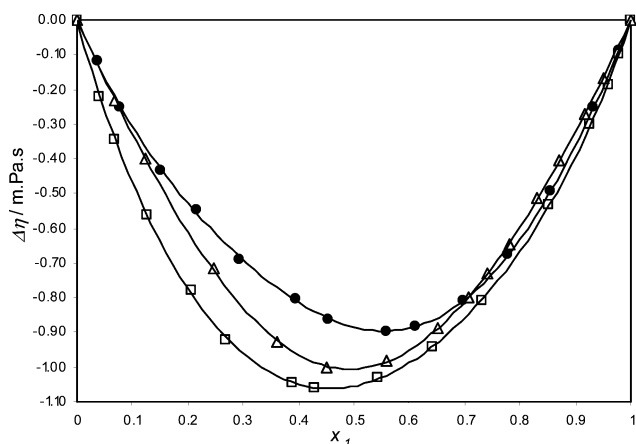
property	$A_1$	$A_2$	$A_3$	$A_4$	$A_5$	$A_6$	$\sigma$
$\Delta\eta/\text{mPa}\cdot\text{s}$	-3.534	-0.782	1,2-Dichloroethane (1) + Hexan-1-ol (2)				0.007
			-0.276	0.636			
$\Delta\eta/\text{mPa}\cdot\text{s}$	-4.221	0.714	1,2-Dibromoethane (1) + Hexan-1-ol (2)				0.008
			-0.590	-0.727	-0.551	0.653	
$\Delta\eta/\text{mPa}\cdot\text{s}$	-4.027	0.192	1,1,2,2-Tetrachloroethene (1) + Hexan-1-ol (2)				0.003
			0.836	-0.521	-0.325	0.465	

presence of four chlorine atoms in 1,1,2,2-tetrachloroethene as compared to two chlorine atoms in 1,2-dichloroethane and two bromine atoms in 1,2-dibromoethane, four H-bonds of the type  $\text{Cl}\cdots\text{H}-\text{O}$  will be formed between the OH group of hexan-1-ol polymeric entities and the 1,1,2,2-tetrachloroethene molecule as compared to two  $\text{Cl}\cdots\text{H}-\text{O}$  bonds in the 1,2-dichloroethane + hexan-1-ol and two  $\text{Br}\cdots\text{H}-\text{O}$  bonds in the 1,2-dibromoethane + hexan-1-ol system, respectively. Unsaturation present in the 1,1,2,2-tetrachloroethene will also contribute toward the interaction strength of 1,1,2,2-tetrachloroethene + hexan-1-ol and may be involved in the formation of a  $\text{OH}\cdots\pi$  electron H-bonded complex which will result in positive viscosity deviations,  $\Delta\eta$ , of the liquid mixtures. Because of the large size and small electronegativity of the Br atom as compared to the Cl atom, the interaction strength of H-bonds  $\text{Cl}\cdots\text{H}-\text{O}$  and  $\text{Br}\cdots\text{H}-\text{O}$  will vary in the order: 1,2-dibromoethane + hexan-1-ol < 1,2-dichloroethane + hexan-1-ol < 1,1,2,2-tetrachloroethene + hexan-1-ol.

The viscosity data have been correlated using Kendall–Monroe,<sup>14</sup> Lobe,<sup>15</sup> Grunberg–Nissan,<sup>16</sup> Tamura–Kurata,<sup>17</sup> Hind–McLaughlin–Ubbelohde,<sup>18</sup> and Katti–Chaudhary<sup>19</sup> viscosity models and the interaction parameters  $G_{12}$ ,  $T_{12}$ ,  $H_{12}$ , and  $W_{\text{vis}}/RT$ .  $W_{\text{vis}}/RT$  for single parameter equations<sup>16–19</sup> have been calculated. The values of the interaction parameters,  $G_{12}$ ,  $T_{12}$ ,  $H_{12}$ , and  $W_{\text{vis}}/RT$ , may have positive or negative values depending upon the nature of molecular interactions.<sup>3,25</sup> The correlating ability of these models was tested by calculating the root-mean-square deviations ( $\sigma^*$ ) using the relation

$$\sigma^* = [(1/N) \sum (\eta_{\text{exp}} - \eta_{\text{cal}})^2]^{1/2} \quad (5)$$

where  $N$  is the number of experimental data. The proposed models provide a reasonably accurate estimation of the viscosity of the binary liquid mixtures under consideration at 293.15 K



**Figure 1.** (1) Viscosity deviation ( $\Delta\eta$ ) versus mole fraction ( $x_1$ ) at 293.15 K for the binary mixtures of hexan-1-ol with: ●, 1,2-dichloroethane; □, 1,2-dibromoethane; △, 1,1,2,2-tetrachloroethene. Solids curves represent the correlated values obtained by the Redlich–Kister equation.

which is evident from the small root-mean-square deviations ( $\sigma^*$ ) of the viscosity values. The values of  $\sigma^*$  are in the range from 0.0960 to 0.5041 for the Kendall–Monroe<sup>14</sup> and Lobe equations<sup>15</sup> without any adjustable parameter and from 0.0443 to 0.4227 for the single-parameter equations at 293.15 K. Analysis of the results of single-parameter equations reveals that  $\sigma^*$  values obtained from the Hind Models are higher than the values obtained from Tamura–Kurata, Grunberg–Nissan, and Katti–Chaudhary models, and the average values of  $G_{12}$ ,  $T_{12}$ ,  $H_{12}$ , and  $W_{\text{vis}}/RT$  are positive for the binary mixtures of the 1,2-dichloroethane + hexan-1-ol systems. The average values of  $T_{12}$  and  $H_{12}$  are positive for the binary mixtures of the 1,2-dibromoethane + hexan-1-ol and 1,1,2,2-tetrachloroethene + hexan-1-ol system at 293.15 K, whereas the average values of  $G_{12}$  and  $W_{\text{vis}}/RT$  are negative for these two systems at 293.15 K. The study of root-mean-square deviations ( $\sigma^*$ ) reveals that the results of viscosity for all of the systems under consideration can be satisfactorily explained by Katti–Chaudhary model as is evident from the lowest  $\sigma^*$  values obtained using eq 5.

McAllister's three-body interaction model<sup>20</sup> has been used to correlate the kinematic viscosities of the binary mixtures with mole fraction. The correlating ability of the equation was tested by calculating the standard percentage deviations ( $\sigma$  %) using the relation

$$\sigma = \sum [(\eta_{\text{exp}} - \eta_{\text{cal}})/\eta_{\text{exp}}]^2 / (N - m)]^{1/2} \quad (6)$$

where  $\eta_{\text{exp}}$  and  $\eta_{\text{cal}}$  are the experimental and calculated values of viscosity, respectively,  $N$  represents the number of data points, and  $m$  represents the number of coefficients. It is observed that both of the parameters of McAllister's three-body interaction model,<sup>20</sup> namely,  $\nu_{12}$  and  $\nu_{21}$ , are positive, and the study of standard deviations calculated using eq 6 reveals that this model is adequate for correlating the kinematic viscosities of the binary mixtures under study.

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### Supporting Information Available:

Tables of Grunberg–Nissan ( $G_{12}$ ), Tamura–Kurata ( $T_{12}$ ), Hind ( $H_{12}$ ), and Katti–Chaudhary ( $W_{\text{vis}}/RT$ ) interaction parameters; average Grunberg–Nissan ( $G_{12}$ ), Tamura–Kurata ( $T_{12}$ ), Hind ( $H_{12}$ ), and Katti–Chaudhary ( $W_{\text{vis}}/RT$ ) interaction parameters, values of  $\alpha_1$  and  $\alpha_2$  and the root-mean-square deviations ( $\sigma^*$ ); McAllister interaction parameters ( $\nu_{12}$ ,  $\nu_{21}$ ) and standard percentage deviations ( $\sigma$ ). This material is available free of charge via the Internet at <http://pubs.acs.org>.

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