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Received for review May 3, 1982. Revised manuscript received June 24, 1983. Accepted October 12, 1983. Acknowledgment is made to the donors of the Petroleum Research Fund, administered by the American Chemical Society, for support of this investigation.

Viscosities and Activation Energies of Viscous Flow of the Binary Mixtures of *n*-Hexane with Toluene, Chlorobenzene, and 1-Hexanol

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Viscosities of three binaries, viz., *n*-hexane–toluene, *n*-hexane–chlorobenzene, and *n*-hexane–1-hexanol, have been determined at 30, 40, 50, and 60 °C over the complete composition range. Experimental viscosities are compared with values calculated by using equations based on the concept of significant liquid structures as well as McAllister type three-body interactions. Energies of activation for viscous flow have been obtained and their variations with composition have been discussed.

Introduction

As part of a program (*1–3*) to collect and correlate viscosity data of binary liquid mixtures with varying nonideality, we have determined the viscosities and activation energies of the binary mixtures *n*-hexane (1)–toluene (2), *n*-hexane (1)–chlorobenzene (2), and *n*-hexane (1)–1-hexanol (2) in the temperature range 30–60 °C and tested the predictive abilities of viscosity equations based on the concept of significant liquid structures (SLS) (*4*) as well as McAllister type three-body interactions (*5*).

Experimental Section

All the organic chemicals, viz, *n*-hexane, toluene, chlorobenzene, and 1-hexanol, used in the present study were of BDH origin. These were purified (*6*) by fractional distillation and drying and the experimental values of their densities, refractive indices, and viscosities checked with the corresponding literature values (*7*), within allowed limits. The binary liquid mixtures were prepared by weighing the individual components and the viscosities were measured with the help of a thoroughly cleaned, dried, and calibrated Ostwald viscometer following standard experimental procedure described elsewhere (*8a*). Due care was taken to ascertain that limbs of the viscometer coincided with the vertical within 0.5°. The time of flow was determined cautiously several times and the standard deviations were found well within 0.1%. Further, the densities required were measured by the standard method (*8b*) using a specific gravity bottle and the same were used along with the measured absolute viscosity data to obtain the corresponding kinematic viscosities. In each case, the viscosity and density measurements were carried out in a Toshniwal GL-15 precision thermostat limiting the temperature fluctuations to ±0.1 °C.

The uncertainty in viscosity measurements entered in the fourth decimal place for the binaries rich in *n*-hexane, toluene, and chlorobenzene and in the third decimal place for the binaries rich in 1-hexanol. As regards densities, the repeat measurements differed by a few units only in the fourth decimal place in each case.

Results and Discussion

The experimental viscosity–composition–temperature data for the title binaries are recorded in Tables I–III. The mixture kinematic viscosities ν_m were used to test the following equation (*5*) based on McAllister type three-body interactions:

$$\ln \nu_m = X_1^3 \ln \nu_1 + X_2^3 \ln \nu_2 + 3X_1^2X_2 \ln \nu_{12} + 3X_1X_2^2 \ln \nu_{21} - \ln [X_1 + X_2M_2/M_1] + 3X_1^2X_2 \ln \left[\frac{2 + M_2/M_1}{3}\right] + 3X_1X_2^2 \ln \left[\frac{1 + 2M_2/M_1}{3}\right] + X_2^3 \ln M_2/M_1 \quad (1)$$

where ν is the kinematic viscosity in centistokes, M is the molecular weight, X is the mole fraction, and the subscripts m, 1, and 2 represent the mixture and components 1 and 2, respectively, while ν_{12} and ν_{21} are mixture interaction parameters to be determined from experimental data and are given in Table IV.

Again the absolute viscosity data were used to test the following equation (*4*) based on the SLS concept:

$$\eta_m = \frac{Nh}{r_m}\,\frac{6}{2^{1/2}}\,\frac{1}{(1 - e^{-\Theta_1/T})^{X_1}}\,\frac{1}{(1 - e^{-\Theta_2/T})^{X_2}}\,\frac{1}{(V_m - V_{sm})} \times \exp\frac{a_mE_{sm}V_{sm}}{RT(V_m - V_{sm})} + \frac{V_m - V_{sm}}{V_m}\left[\frac{2}{3d_1^2}\left(\frac{m_1kT}{\pi^3}\right)^{1/2} X_1 + \frac{2}{3d_2^2}\left(\frac{m_2kT}{\pi^3}\right)^{1/2} X_2\right] \quad (2)$$

where η is the absolute viscosity, V is the molar volume, V_s is the solidlike volume in V, Θ is the Einstein characteristic temperature, E_s is the energy of sublimation, m is the mass of a single molecule with diameter d in the gaseous state, and a is determined by another parameter n whereas r is equal to the product of the number of nearest neighbors Z and the transmission coefficient κ. Since it is difficult to determine the value of Z a priori and evaluate κ theoretically, r is treated as a dispossable parameter and is usually fixed by forcing agreement between the experimental and calculated viscosities at each temperature. The values of V, V_s, and M for the binary components were taken from an earlier work (*7*) and those of E_s, Θ, a, n, d, and r were evaluated by following the procedure of Singh and Sinha (*1*) and listed in Table V. The mixture

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0021-9568/84/1729-0132$01.50/0

Table I. Viscosities of the Binary Mixtures of *n*-Hexane (1)-Toluene (2) at Different Temperatures

	η_m, cP				ν_m, cS			
X_1	30 °C	40 °C	50 °C	60 °C	30 °C	40 °C	50 °C	60 °C
0.0000	0.5372	0.4851	0.4272	0.3905	0.6196	0.5675	0.5028	0.4608
0.0826	0.4983	0.4542	0.4006	0.3637	0.5959	0.5449	0.4836	0.4419
0.1685	0.4637	0.4265	0.3747	0.3396	0.5679	0.5249	0.4643	0.4255
0.2578	0.4362	0.4033	0.3545	0.3205	0.5463	0.5074	0.4499	0.4114
0.3509	0.4117	0.3782	0.3354	0.3042	0.5285	0.4899	0.4367	0.4008
0.4477	0.3884	0.3592	0.3184	0.2892	0.5120	0.4748	0.4259	0.3914
0.5487	0.3671	0.3375	0.3022	0.2756	0.4968	0.4610	0.4162	0.3844
0.6541	0.3492	0.3202	0.2867	0.2635	0.4853	0.4510	0.4078	0.3796
0.7643	0.3299	0.3043	0.2736	0.2535	0.4726	0.4409	0.4018	0.3761
0.8795	0.3136	0.2896	0.2620	0.2433	0.4625	0.4322	0.3958	0.3726
1.0000	0.2977	0.2768	0.2524	0.2347	0.4523	0.4247	0.3922	0.3691

Table II. Viscosities of the Binary Mixtures of *n*-Hexane (1)-Chlorobenzene (2) at Different Temperatures

	η_m, cP				ν_m, cS			
X_1	30 °C	40 °C	50 °C	60 °C	30 °C	40 °C	50 °C	60 °C
0.0000	0.7184	0.6469	0.5724	0.5198	0.6530	0.5913	0.5257	0.4796
0.0793	0.6623	0.6011	0.5314	0.4849	0.6251	0.5713	0.5076	0.4654
0.1624	0.6095	0.5513	0.4893	0.4498	0.6022	0.5487	0.4896	0.4513
0.2494	0.5578	0.5124	0.4543	0.4146	0.5730	0.5299	0.4739	0.4337
0.3407	0.5101	0.4699	0.4164	0.3912	0.5514	0.5117	0.4571	0.4337
0.4367	0.4658	0.4288	0.3835	0.3529	0.5273	0.4886	0.4403	0.4067
0.5377	0.4254	0.3946	0.3551	0.3258	0.5082	0.4748	0.4307	0.3973
0.6440	0.3927	0.3647	0.3268	0.3002	0.4955	0.4623	0.4119	0.3879
0.7562	0.3549	0.3305	0.2997	0.2784	0.4739	0.4435	0.4078	0.3808
0.8747	0.3227	0.3021	0.2745	0.2559	0.4574	0.4322	0.3969	0.3726
1.0000	0.2977	0.2768	0.2524	0.2347	0.4523	0.4247	0.3922	0.3691

Table III. Viscosities of the Binary Mixtures of *n*-Hexane (1)-1-Hexanol (2) at Different Temperatures

	η_m, cP				ν_m, cS			
X_1	30 °C	40 °C	50 °C	60 °C	30 °C	40 °C	50 °C	60 °C
0.0000	3.769	2.934	2.169	1.655	4.628	3.619	2.689	2.059
0.0954	2.883	2.219	1.682	1.321	3.602	2.788	2.125	1.676
0.1918	2.180	1.710	1.320	1.030	2.780	2.195	1.705	1.338
0.2892	1.600	1.295	1.020	0.8313	2.076	1.690	1.341	1.101
0.3876	1.190	0.9896	0.8098	0.6497	1.578	1.323	1.090	0.8804
0.4870	0.8951	0.7581	0.6263	0.5261	1.210	1.032	0.8601	0.7299
0.5874	0.6793	0.5894	0.5103	0.4298	0.9402	0.8231	0.7182	0.6100
0.6890	0.5301	0.4749	0.4040	0.3577	0.7496	0.6765	0.5822	0.5195
0.7915	0.4188	0.3791	0.3396	0.2995	0.6060	0.5549	0.5016	0.4466
0.8952	0.3557	0.3285	0.2844	0.2607	0.5273	0.4924	0.4307	0.3996
1.0000	0.2977	0.2768	0.2524	0.2347	0.4523	0.4247	0.3922	0.3691

Table IV. Values of Interaction Parameters ν_{12} and ν_{21} for the Binary Systems at Different Temperatures

	ν_{12}				ν_{21}			
binary system	30 °C	40 °C	50 °C	60 °C	30 °C	40 °C	50 °C	60 °C
n-hexane (1)-toluene (2)	0.5057	0.4433	0.4027	0.3785	0.4981	0.4792	0.4230	0.3821
n-hexane (1)-chlorobenzene (2)	0.4736	0.4453	0.4091	0.3750	0.5471	0.5089	0.4517	0.4252
n-hexane (1)-1-hexanol (2)	0.4836	0.4898	0.4268	0.3836	2.470	1.827	1.498	1.199

Table V. Values of Physical Properties and Parameters Used in Eq 2 for the Binary Components

parameters		*n*-hexane	toluene	chlorobenzene	1-hexanol	ref
E_s, cal mol^{-1}		10022.00	9517.00	11019.00	15480.00	*1*
Θ		62.10	103.62	105.41	74.67	*1*
n		10.9982	11.3269	11.3531	11.2653	*1*
$a \times 10^3$		3.168	1.434	1.322	1.708	*1*
$d \times 10^8$, cm		5.179	4.915	4.868	4.988	*1*
r,	30 °C	1.426	0.7359	0.7425	0.1864	
	40 °C	1.485	0.7598	0.8016	0.2342	
	50 °C	1.556	0.8502	0.8888	0.3076	
	60 °C	1.612	0.9355	0.9632	0.3952	

parameters V_m, V_{sm}, E_{sm}, a_m, and r_m were obtained from respective pure components (Table V) by applying mixture rules (*4*).

From the values of percent average deviations as listed in Table VI, it is obvious that both the viscosity models tested gave comparatively inferior results for the *n*-hexane–1-hexanol system. The McAllister model involving three-body interactions with two adjustable mixture parameters was found to be better, giving an overall mean percent average deviation of 1.75 for the three systems at four different temperatures and a percent average deviation of 4.24 for *n*-hexane–1-hexanol at the same temperatures. As such the McAllister equation (eq 1) can profitably be used to predict the missing mixture viscosity values requiring limited mixture viscosity–composition data only. The

Table VI. Percent Average Deviations for Different Binary Systems Using SLS equation (Eq 2) and Three-Body Interaction Equation (Eq 1) at Different Temperatures

	± % av deviation[a]							
	eq 2				eq 1			
binary system	30 °C	40 °C	50 °C	60 °C	30 °C	40 °C	50 °C	60 °C
n-hexane (1)-toluene (2)	1.312	1.699	0.6167	0.7444	1.439	0.1002	0.0999	0.0918
n-hexane (1)-chlorobenzene (2)	4.610	5.032	4.556	4.837	0.4598	0.3433	0.6083	0.8254
n-hexane (1)-1-hexanol (2)	21.63	16.53	12.42	7.914	5.185	4.237	3.951	3.604

[a] % av deviation = $[\sum_{i=1}^{n} d_i^2/n]^{1/2}$ where n is the number of observations and d = 100[(exptl − calcd)/exptl].

Table VII. Values of $\Delta G^{\ddagger}_m$ at Different Temperatures along with the Values of $\Delta H^{\ddagger}_m$ and $\Delta S^{\ddagger}_m$ for the Binary Mixtures of *n*-Hexane (1)-Toluene (2)

X_1	$\Delta G^{\ddagger}_m$, cal mol^{-1}				$\Delta H^{\ddagger}_m$, cal mol^{-1}	$\Delta S^{\ddagger}_m$, cal mol^{-1}
	30 °C	40 °C	50 °C	60 °C		
0.0000	2990.00	3024.91	3059.82	3094.73	1932.22	−3.49
0.0826	2963.17	2991.09	3035.03	3070.95	1874.59	−3.59
0.1685	2931.44	2968.07	3004.69	3041.33	1821.59	−3.66
0.2578	2902.23	2939.87	2977.49	3015.13	1761.98	−3.76
0.3509	2877.75	2916.32	2954.89	2993.47	1708.99	−3.86
0.4477	2852.75	2892.47	2932.19	2971.90	1649.38	−3.97
0.5487	2833.78	2874.84	2915.89	2956.95	1589.76	−4.11
0.6541	2815.41	2857.83	2900.25	2942.67	1530.14	−4.24
0.7643	2793.33	2836.77	2880.21	2923.65	1477.15	−4.34
0.8795	2775.15	2820.39	2865.64	2910.88	1404.29	−4.52
1.0000	2758.99	2805.59	2852.19	2898.79	1346.66	−4.65

Table VIII. Values of $\Delta G^{\ddagger}_m$ at Different Temperatures along with the Values of $\Delta H^{\ddagger}_m$ and $\Delta S^{\ddagger}_m$ for the Binary Mixtures of *n*-Hexane (1)-Chlorobenzene (2)

X_1	$\Delta G^{\ddagger}_m$, cal mol^{-1}				$\Delta H^{\ddagger}_m$, cal mol^{-1}	$\Delta S^{\ddagger}_m$, cal mol^{-1}
	30 °C	40 °C	50 °C	60 °C		
0.0000	3141.01	3177.18	3213.36	3249.53	2046.82	−3.61
0.0793	3104.08	3142.03	3179.98	3217.94	1954.08	−3.79
0.1624	3069.15	3108.57	3147.99	3187.42	1874.59	−3.94
0.2494	3028.19	3068.89	3109.59	3150.28	1795.10	−4.07
0.3407	2990.05	3031.67	3073.29	3114.92	1728.86	−4.16
0.4367	2945.48	2988.26	3031.03	3073.81	1649.38	−4.28
0.5377	2911.06	2955.10	2999.15	3043.19	1576.51	−4.40
0.6440	2871.89	2916.61	2961.33	3006.05	1516.89	−4.47
0.7562	2830.36	2875.68	2920.99	2966.31	1457.28	−4.53
0.8747	2787.39	2833.48	2879.57	2925.65	1391.04	−4.61
1.0000	2758.99	2805.59	2852.19	2898.79	1346.66	−4.65

Table IX. Values of $\Delta G^{\ddagger}_m$ at Different Temperatures along with the Values of $\Delta H^{\ddagger}_m$ and $\Delta S^{\ddagger}_m$ for the Binary Mixtures of *n*-Hexane (1)-1-Hexanol (2)

X_1	$\Delta G^{\ddagger}_m$, cal mol^{-1}				$\Delta H^{\ddagger}_m$, cal mol^{-1}	$\Delta S^{\ddagger}_m$, cal mol^{-1}
	30 °C	40 °C	50 °C	60 °C		
0.0000	4268.03	4229.62	4191.22	4152.82	5431.68	3.84
0.0954	4100.68	4072.71	4044.74	4016.77	4948.13	2.79
0.1918	3920.30	3900.15	3880.01	3859.86	4530.82	2.01
0.2892	3752.57	3741.97	3731.37	3720.77	4073.76	1.06
0.3876	3583.32	3582.88	3582.43	3581.99	3596.83	0.04
0.4870	3399.74	3406.57	3413.39	3420.23	3192.77	−0.68
0.5874	3248.16	3263.54	3278.93	3294.31	2782.08	−1.54
0.6890	3102.13	3125.16	3148.18	3171.20	2404.51	−2.30
0.7915	2956.87	2986.69	3016.50	3046.32	2053.44	−2.98
0.8952	2866.71	2904.48	2942.26	2980.03	1722.24	−3.78
1.0000	2758.99	2805.59	2852.19	2898.79	1346.66	−4.65

SLS model involving no adjustable mixture parameter gave under similar experimental conditions an overall mean percent average deviation of 6.53 for the three systems and a percent average deviation of 14.6 for *n*-hexane–1-hexanol, indicating that it can be employed for tolerable prediction of mixture viscosity of the nonassociated mixtures studied even in the absence of any mixture data.

The activation energies of the viscous flow are related to the mixture viscosities by the Eyring equation (*9*) in the following form:

$$\nu_m M_m = hNe^{-\Delta S^{\ddagger}_m/R} e^{\Delta H^{\ddagger}_m/RT} \qquad (3)$$

where $\Delta S^{\ddagger}$ and $\Delta H^{\ddagger}$ are the entropy and enthalpy of activation of viscous flow, h is Planck's constant, N is Avogadro's number, R is the gas constant, and T is the temperature in Kelvin, while M_m for a liquid is given by $M_m = \sum X_i M_i$. Since the plots of ln ($\nu_m M_m$) vs. $1/T$ for the binary systems studied were found to be satisfactorily linear, $\Delta S^{\ddagger}$ and $\Delta H^{\ddagger}_m$ were obtained by the usual procedure (*8c*) and used to calculate (*9*) corresponding values of free energies of activation $\Delta G^{\ddagger}_m$. The results are given in Tables VII–IX.

In the temperature range investigated, the values of $\Delta H^{\ddagger}$ are positive and vary in the order 1-hexanol > chlorobenzene > toluene > *n*-hexane with $\Delta H^{\ddagger}$ for 1-hexanol nearly 4 times larger than that for *n*-hexane whereas $\Delta H^{\ddagger}$ for toluene and chlorobenzene are close to each other and nearly $1^1/_2$ larger than that of *n*-hexane, indicating that association and dipole-dipole interactions increase the value of $\Delta H^{\ddagger}$. The negative values of $\Delta S^{\ddagger}$ for nonassociated liquids viz., *n*-hexane, toluene, and chlorobenzene indicate that the formation of activated complexes introduces increased molecular order while positive values of $\Delta S^{\ddagger}$ for 1-hexanol show that the formation of activated complexes in this case involves less order probably due to destruction of hydrogen bonds. The values of $\Delta G^{\ddagger}$ depend on the sign and the magnitude of the corresponding $\Delta H^{\ddagger}$ and $\Delta S^{\ddagger}$ and are positive in each case. The variation of $\Delta G^{\ddagger}$ at each temperature studied follows the same order as that of $\Delta H^{\ddagger}$ because the corresponding $\Delta S^{\ddagger}$ is comparatively smaller in each case.

The values of $\Delta H^{\ddagger}{}_{m}$ and $\Delta G^{\ddagger}{}_{m}$ are positive for each of the binary systems studied. But $\Delta S^{\ddagger}{}_{m}$ values are negative for *n*-hexane–toluene and *n*-hexane–chlorobenzene systems as well as *n*-hexane-rich binaries of the *n*-hexane–1-hexanol system. however, the $\Delta S^{\ddagger}{}_{m}$ values for *n*-hexane-poor binaries of the *n*-hexane–1-hexanol system are positive. This indicates that, for *n*-hexane-rich binaries of this system, the chemical effects of H-bond breaking and nonspecific interactions between the real species in the mixture, e.g., 1-hexanol monomers and multimers and also *n*-hexane molecules, lead to less molecular order in the unactivated state than that in the activated state. On the other hand, in the *n*-hexane-poor binaries, *n*-hexane molecules leading to increased molecular order in the unactivated state than that in the activated state. It is interesting to note that $\Delta S^{\ddagger}{}_{m} = 0$ for a critical composition $X_1 = 0.42$, obtained from $\Delta S^{\ddagger}{}_{m}$ vs. X_1 plots and consequently the *n*-hexane–1-hexanol binary with $X_1 = 0.42$ involves the activated and unactivated states of similar molecular order.

Acknowledgment

We are grateful to the authorities of Bhagalpur College of Engineering, Bhagalpur, for providing laboratory facilities.

Glossary

a	adjustable parameter
d	diameter in gaseous state
d	percent deviation
E_s	energy of sublimation
$\Delta G^{\ddagger}$	free energy of activation of viscous flow
$\Delta H^{\ddagger}$	enthalpy of activation of viscous flow
h	planck's constant
M	molecular weight
m	mass of a single molecule
N	Avogadro's number
$\Delta S^{\ddagger}$	entropy of activation of viscous flow
V	molar volume, cm^3 mol^{-1}
V_s	solidlike volume in V, cm^3 mol^{-1}
X	mole fraction
ν	kinematic viscosity, cS
η	absolute viscosity, cP
θ	Einstein characteristic temperatures
κ	transmission coefficient

Subscripts

i	component in a mixture
m	mixture
1, 2	component number in a mixture

Registry No. Hexane, 110-54-3; toluene, 108-88-3; chlorobenzene, 108-90-7; 1-hexanol, 111-27-3.

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Received for review July 26, 1982. Revised manuscript received May 9, 1983. Accepted June 14, 1983. We are grateful to the UGC, New Delhi, for the award of a teacher followship to C.P.S.

Vapor Pressures of *o*- and *m*-Nitrochlorobenzene

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Vapor pressures of *o*- and *m*-nitrochlorobenzene were measured over the pressure range 50–700 mmHg by using a standard Swietoslawski type ebulliometer. The data were fitted by the Antoine equation of the form $\ln P = A - B/(t + C)$. The calculated molal heat of vaporization at the normal boiling point is 12.19 kcal/mol for *o*-nitrochlorobenzene and 12.07 kcal/mol for *m*-nitrochlorobenzene.

Introduction

The nitrochlorobenzenes are important starting materials for the manufacture of azo and sulfur dyes and they also find application in the synthesis of fungicides, preservatives, and pharmaceuticals. While measurements of vapor pressure data were reported for *p*-nitrochlorobenzene (*1*), few data are available on the vapor pressures of ortho and meta isomers. In this paper we present measurements on the vapor pressures of *o*- and *m*-nitrochlorobenzene and their suitable correlations.

Experimental Details

o-Nitrochlorobenzene of laboratory reagent grade supplied by M/s Sisco-Chem Industries, Bombay, India, and *m*-nitrochlorobenzene prepared at the Regional Research Laboratory, Hyderabad, India, were distilled and the middle cuts of the distillates were used in the experimental measurements. The physical constant density, refractive index, and boiling point of the chemicals used are presented in Table I, along with the literature data.

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0021-9568/84/1729-0135$01.50/0