Viscosity and Density of Binary Liquid Mixtures of Hydrocarbons, Esters, Ketones, and Normal Chloroalkanes

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Kinematic viscosities and densities of 43 binary mixtures containing aromatic and cyclic hydrocarbons, esters, ketones, and normal chloroalkanes were measured at 298.15 K over the entire composition range. The experimental values of kinematic viscosities have been used to extend the interaction parameters of the UNIFAC-VISCO predictive model.

Introduction

A knowledge of the viscosity of liquid mixtures is required in many chemical engineering designs. In the present work, viscosities and densities of 43 aromatic and cyclic hydrocarbon, ester, ketone, and normal chloroalkane liquid mixtures have been measured at 298.15 K. Beyond the interest of knowing kinematic viscosities and densities of new binary systems, the aim of this work is to extend the table of interaction parameter values of the UNIFAC-VISCO model presented in a previous paper *(1)*.

Experimental Section

The components used are Fluka "puriss" (purity >99.5%) or "purum" (purity >98%) quality, Janssen Chimica (purity >98%), and Prolabo "pour HPLC" (purity >99.5%) products. In Table 1, we compared our experimental values of the pure components with those found in the literature.

All the mixtures composed of the original components are prepared by mass with a H-10 W Mettler balance to $\pm 0.5 \times 10^{-6}$ kg.

Kinematic viscosities are measured using a Lauda S automatic viscometer equipped with two KPG Ubbelhode capillary viscometers of 0.46×10^{-3} and 0.53×10^{-3} m diameter and two Ubbelhode microviscometers of 0.40×10^{-3} and 0.53×10^{-3} m diameter, all of them calibrated with doubly distilled water. Water has been used for calibration since it is the only product whose properties are known with good accuracy and which is easily available with a high level of purity.

The kinematic viscosity ν is determined from the following relationship:

$$\nu = k(t - v) \tag{1}$$

where t, k, and v are, respectively, the flow time, the viscometer constant, and the Hagenbach correction *(21)*.

Densities are measured with a DMA 02D Anton-Paar densimeter. The density-meter constant C_0 of eq 2 is determined from two reference liquids, octane and decane, the densities, ϱ_1 and ϱ_2, of which are known with absolute accuracy, better than ± 0.05 kg·m^{-3}.

The density ϱ is determined from the following relationship:

$$\varrho_1 - \varrho_2 = C_0(T_1^2 - T_2^2) \tag{2}$$

where ϱ and T are, respectively, the density and the period.

In all the cases, the temperature is maintained at 298.15 ± 0.02 K using a D20 Lauda ultrathermostat equipped with a R25 PID regulator.

The kinematic viscosities and densities of binary mixtures are determined with accuracies, respectively, better than ±0.1% and ±0.5 kg·m^{-3} *(22)*.

About the kinemtic viscosity accuracy, the indicated value of 0.1% only takes into account the experimental error and not the accuracy resulting from the presence of impurities in the original components. Moreover, the validity of the experimental values of pure component viscosities was discussed in a previous paper *(22)*.

Results and Discussion

The experimental viscosities and densities of the binary liquid mixtures are shown in Table 2.

From the experimental values of the kinematic viscosity, we have completed the group interaction parameter table published in a previous paper *(1)*. In that work the predictive UNIFAC-VISCO method was developed.

We recall the principal equations of the UNIFAC-VISCO model.

The theory of Eyring *(23)* applied at the nonideal mixture gives the following equation:

$$\ln(\nu M) = \sum_i x_i \ln(\nu_i M_i) + \Delta^* G^E/RT \tag{3}$$

with

$$M = \sum_i x_i M_i \tag{4}$$

where x_i, M_i, and ν_i are, respectively, the mole fraction, the molecular weight (kg·mol^{-1}), and the kinematic viscosity (m^2·s^{-1}) of the component i, M and ν are the molecular weight and the kinematic viscosity of the mixture, $\Delta^* G^E$ (J·mol^{-1}) is the excess molar free energy of activation for flow, R (J·mol^{-1}·K^{-1}) is the gas constant, and T (K) is the absolute temperature.

The excess molar free energy of activation for flow $\Delta^* G^E$ is represented by the UNIFAC group contribution method *(24)* adapted to viscosities. It is the sum of two contributions,

$$\Delta^* G^E = \Delta^* G^{EC} + \Delta^* G^{ER} \tag{5}$$

a combinatorial part, $\Delta^* G^{EC}$, which takes into account known UNIFAC group structural parameters and a residual part, $\Delta^* G^{ER}$, which takes into account unknown UNIFAC-VISCO group interaction parameters ψ^*_{nm}. n

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Table 1. Properties of Pure Components

component	$10^6\nu/(m^2 s^{-1})$ exptl	$10^6\nu/(m^2 s^{-1})$ lit.		$10^{-3}\rho/(kg m^{-3})$ exptl	$10^{-3}\rho/(kg m^{-3})$ lit.	
cyclohexane	1.148	1.1445–1.1644	(*8, 10, 16*)	0.7738	0.7736–0.7742	(*3, 8, 10*)
methylcyclohexane	0.8807			0.7648	0.76505	
cis-1,2-dimethylcyclohexane	1.278			0.7929		
1,2-dimethylcyclohexane (mixture of cis and trans)	1.154			0.7861		
1,2,4-trimethylcyclohexane (mixture of cis and trans)	0.9976			0.7816		
benzene	0.6844	0.6863–0.7017	(*6–8, 11, 13, 15, 16*)	0.8734	0.87355–0.8736	(*6–8*)
toluene	0.6345	0.6413–0.6507	(*7, 11, 13, 16*)	0.8616	0.8619–0.8623	(*2, 5, 7*)
o-xylene	0.8542	0.8658	(*16*)	0.8752	0.87596	(*4*)
p-xylene	0.6975	0.7085	(*16*)	0.8565	0.85669	(*4*)
1-chlorohexane	0.7856	0.783–0.796	(*9, 14*)	0.8732	0.87338–0.8745	(*9, 14*)
1-chlorohexadecane	6.305	6.299–6.40	(*9, 14*)	0.8607	0.86065–0.8616	(*9, 14*)
ethyl acetate	0.4714	0.476–0.490	(*12, 19*)	0.8948	0.8945	(*5, 19*)
propyl propionate	0.7111	0.720	(*12*)	0.8755		
acetone	0.3831	0.3857–0.398	(*10, 12*)	0.7847	0.7844–0.78499	(*7, 10, 20*)
methyl ethyl ketone	0.4676	0.478–0.481	(*12, 19*)	0.7996	0.79950–0.79970	(*4, 17, 18*)

Table 2. Kinematic Viscosities and Densities of Binary Mixtures at 298.15 K

x_1	$10^6\nu/(m^2 s^{-1})$	$10^{-3}\rho/(kg m^{-3})$	x_1	$10^6\nu/(m^2 s^{-1})$	$10^{-3}\rho/(kg m^{-3})$	x_1	$10^6\nu/(m^2 s^{-1})$	$10^{-3}\rho/(kg m^{-3})$
Acetone (1) + Cyclohexane (2)								
0.0996	0.9505	0.7711	0.3927	0.6580	0.7683	0.6877	0.4835	0.7715
0.2056	0.8149	0.7692	0.5042	0.5730	0.7689	0.8015	0.4362	0.7747
0.3401	0.6915	0.7683	0.5889	0.5336	0.7697	0.8872	0.4081	0.7784
Methyl Ethyl Ketone (1) + Methylcyclohexane (2)								
0.1137	0.7931	0.7655	0.4138	0.6399	0.7715	0.6889	0.5420	0.7815
0.2032	0.7378	0.7668	0.4901	0.6103	0.7738	0.7914	0.5122	0.7865
0.3235	0.6787	0.7692	0.5853	0.5759	0.7774	0.8938	0.4869	0.7925
Acetone (1) + *cis*-1,2-Dimethylcyclohexane (2)								
0.1145	1.017	0.7859	0.3957	0.7486	0.7822	0.6899	0.5360	0.7804
0.1946	0.9287	0.7846	0.4919	0.6753	0.7813	0.7896	0.4795	0.7806
0.2939	0.8338	0.7833	0.5808	0.6083	0.7807	0.8976	0.4230	0.7819
1-Chlorohexane (1) + Cyclohexane (2)								
0.1194	1.016	0.7865	0.4135	0.8676	0.8178	0.6693	0.8135	0.8431
0.1987	0.9599	0.7951	0.4969	0.8454	0.8262	0.7855	0.7991	0.8539
0.3014	0.9077	0.8068	0.6042	0.8206	0.8365	0.8816	0.7903	0.8627
1-Chlorohexadecane (1) + Cyclohexane (2)								
0.1016	1.574	0.7928	0.3807	2.973	0.8261	0.6648	4.523	0.8398
0.1959	2.018	0.8066	0.4850	3.541	0.8241	0.7605	5.051	0.8509
0.2955	2.518	0.8183	0.5811	4.066	0.8411	0.8672	5.625	0.8556
1-Chlorohexane (1) + Methylcyclohexane (2)								
0.1074	0.8511	0.7768	0.5004	0.7961	0.8200	0.8928	0.7821	0.8619
0.3018	0.8165	0.7983	0.6947	0.7864	0.8388			
1-Chlorohexane (1) + 1,2-Dimethylcyclohexane (Mixture of Cis and Trans) (2)								
0.1142	1.084	0.7975	0.5043	0.9061	0.8302	0.8970	0.8026	0.8640
0.3054	0.9827	0.8134	0.7010	0.8481	0.8470			
1-Chlorohexadecane (1) + Methylcyclohexane (2)								
0.1173	1.294	0.7878	0.5170	3.214	0.8336	0.8789	5.485	0.8532
0.3139	2.131	0.8145	0.6675	4.109	0.8440			
1-Chlorohexadecane (1) + 1,2,4-Trimethylcyclohexane (Mixture of Cis and Trans) (2)								
0.1133	1.378	0.7976	0.4960	3.105	0.8337	0.8979	5.596	0.8563
0.3163	2.207	0.8193	0.7029	4.312	0.8467			
Ethyl Acetate (1) + Cyclohexane (2)								
0.1282	0.8990	0.7831	0.4015	0.6491	0.8099	0.6986	0.5246	0.8472
0.2261	0.7835	0.7918	0.5092	0.5923	0.8225	0.7933	0.5010	0.8609
0.3215	0.7021	0.8013	0.5942	0.5584	0.8331	0.8821	0.4845	0.8746
Ethyl Acetate (1) + 1,2-Dimethylcyclohexane (Mixture of Cis and Trans) (2)								
0.0855	1.046	0.7930	0.3929	0.7510	0.8160	0.6802	0.5840	0.8460
0.2101	0.9044	0.8013	0.4945	0.6838	0.8255	0.7970	0.5342	0.8613
0.3261	0.8015	0.8103	0.5983	0.6248	0.8364	0.8950	0.4996	0.8760
Propyl Propionate (1) + Methylcyclohexane (2)								
0.1204	0.8233	0.7770	0.4008	0.7491	0.8070	0.6926	0.7150	0.8396
0.2062	0.7942	0.7860	0.4990	0.7339	0.8179	0.8025	0.7096	0.8523
0.2942	0.7710	0.7954	0.5994	0.7223	0.8291	0.8963	0.7081	0.8693
Propyl Propionate (1) + 1,2,4-Trimethylcyclohexane (Mixture of Cis and Trans) (2)								
0.1107	0.9353	0.7893	0.4066	0.8227	0.8128	0.7006	0.7520	0.8409
0.2027	0.8938	0.7961	0.5104	0.7940	0.8222	0.7925	0.7359	0.8508
0.3112	0.8531	0.8047	0.6105	0.7705	0.8318	0.8862	0.7238	0.8615
Acetone (1) + Benzene (2)								
0.1311	0.6195	0.8640	0.5194	0.4834	0.8329	0.8941	0.4036	0.7970
0.3321	0.5419	0.8486	0.6875	0.4407	0.8175			

Table 2. (Continued)

x_1	$10^6\nu/(m^2 \cdot s^{-1})$	$10^{-3}\rho/(kg \cdot m^{-3})$	x_1	$10^6\nu/(m^2 \cdot s^{-1})$	$10^{-3}\rho/(kg \cdot m^{-3})$	x_1	$10^6\nu/(m^2 \cdot s^{-1})$	$10^{-3}\rho/(kg \cdot m^{-3})$
				Acetone (1) + Toluene (2)				
0.0096	0.6042	0.8567	0.5026	0.4979	0.8321	0.8769	0.4082	0.7994
0.3257	0.5441	0.8442	0.7140	0.4453	0.8151			
				Acetone (1) + *o*-Xylene (2)				
0.1236	0.8050	0.8709	0.4988	0.5923	0.8433	0.8868	0.4242	0.8023
0.3303	0.6757	0.8562	0.7001	0.5003	0.8244			
				Methyl Ethyl Ketone (1) + Benzene (2)				
0.1292	0.6299	0.8643	0.5033	0.5335	0.8377	0.8609	0.4801	0.8111
0.3124	0.5754	0.8513	0.6883	0.5022	0.8240			
				Methyl Ethyl Ketone (1) + Toluene (2)				
0.1398	0.6028	0.8551	0.5166	0.5357	0.8347	0.8877	0.4809	0.8096
0.3207	0.5691	0.8459	0.7163	0.5045	0.8218			
				Methyl Ethyl Ketone (1) + *p*-Xylene (2)				
0.1382	0.6567	0.8514	0.5881	0.5643	0.8334	0.8821	0.4859	0.8096
0.3399	0.6070	0.8427	0.6956	0.5262	0.8231			
				1-Chlorohexane (1) + Benzene (2)				
0.1032	0.6601	0.8721	0.4810	0.6877	0.8717	0.8575	0.7491	0.8727
0.3077	0.6589	0.8715	0.6529	0.7078	0.8721			
				1-Chlorohexane (1) + Toluene (2)				
0.1318	0.6377	0.8638	0.5008	0.6860	0.8688	0.8708	0.7543	0.8722
0.3182	0.6581	0.8665	0.6948	0.7204	0.8707			
				1-Chlorohexane (1) + *o*-Xylene (2)				
0.0913	0.8377	0.8754	0.5003	0.8007	0.8753	0.8632	0.7838	0.8738
0.3050	0.8146	0.8750	0.6921	0.7904	0.8746			
				1-Chlorohexadecane (1) + Benzene (2)				
0.1022	0.9979	0.8665	0.4559	2.625	0.8612	0.8301	5.041	0.8608
0.2905	1.767	0.8625	0.6835	4.031	0.8609			
				1-Chlorohexadecane (1) + Toluene (2)				
0.1044	0.9375	0.8606	0.4686	2.525	0.8605	0.8197	4.841	0.8607
0.2756	1.570	0.8603	0.6490	3.628	0.8606			
				1-Chlorohexadecane (1) + *p*-Xylene (2)				
0.0953	0.9496	0.8577	0.4616	2.464	0.8599	0.8941	5.391	0.8607
0.2714	1.563	0.8590	0.6971	3.914	0.8604			
				Ethyl Acetate (1) + Benzene (2)				
0.1285	0.6281	0.8756	0.5062	0.5320	0.8833	0.8599	0.4821	0.8911
0.3257	0.5702	0.8795	0.6875	0.5032	0.8873			
				Ethyl Acetate (1) + Toluene (2)				
0.1426	0.6041	0.8660	0.4865	0.5433	0.8770	0.8764	0.4846	0.8900
0.3095	0.5736	0.8713	0.6635	0.5150	0.8828			
				Ethyl Acetate (1) + *o*-Xylene (2)				
0.1374	0.7866	0.8774	0.4938	0.6376	0.8837	0.8611	0.5101	0.8911
0.3296	0.7030	0.8807	0.6954	0.5644	0.8877			
				Propyl Propionate (1) + Benzene (2)				
0.1219	0.6701	0.8738	0.4856	0.6741	0.8749	0.8633	0.6969	0.8754
0.3185	0.6683	0.8744	0.6618	0.6833	0.8752			
				Propyl Propionate (1) + Toluene (2)				
0.1334	0.6423	0.8645	0.5087	0.6713	0.8707	0.8609	0.6970	0.8744
0.2991	0.6555	0.8676	0.6649	0.6827	0.8725			
				Propyl Propionate (1) + *p*-Xylene (2)				
0.1170	0.6948	0.8594	0.5116	0.6970	0.8675	0.8581	0.7029	0.8733
0.2997	0.6949	0.8634	0.6660	0.6991	0.8702			
				Acetone (1) + 1-Chlorohexane (2)				
0.1087	0.7380	0.8669	0.4289	0.6037	0.8453	0.7064	0.4919	0.8204
0.2728	0.6693	0.8562	0.4916	0.5778	0.8398	0.7838	0.4617	0.8117
0.3560	0.6340	0.8504	0.5810	0.5428	0.8323	0.8977	0.4185	0.7985
				Methyl Ethyl Ketone (1) + Chlorohexane (2)				
0.1364	0.7394	0.8658	0.4146	0.6445	0.8482	0.7386	0.5443	0.8239
0.2372	0.7030	0.8595	0.5605	0.6114	0.8410	0.7811	0.5296	0.8200
0.3443	0.6674	0.8527	0.6134	0.5814	0.8338	0.8758	0.5015	0.8116
				Acetone (1) + 1-Chlorohexadecane (2)				
0.1256	5.310		0.4495	3.028		0.6846	1.684	
0.2149	4.648		0.5309	2.536		0.8076	1.086	
0.2643	4.279		0.6166	2.044		0.8940	0.7343	
				Methyl Ethyl Ketone (1) + 1-Chlorohexadecane (2)				
0.1799	4.775	0.8563	0.4537	2.879	0.8467	0.7082	1.523	0.8326
0.2625	4.175	0.8539	0.5145	2.520	0.8440	0.8029	1.119	0.8246
0.3411	3.599	0.8512	0.6206	1.950	0.8385	0.8965	0.7873	0.8146

Table 2. (Continued)

x_1	$10^6\nu/(m^2 s^{-1})$	$10^{-3}\rho/(kg m^{-3})$	x_1	$10^6\nu/(m^2 s^{-1})$	$10^{-3}\rho/(kg m^{-3})$	x_1	$10^6\nu/(m^2 s^{-1})$	$10^{-3}\rho/(kg m^{-3})$
				Acetone (1) + Ethyl Acetate (2)				
0.1220	0.4617	0.8843	0.4359	0.4333		0.7285	0.4070	0.8213
0.2353	0.4551	0.8738	0.5082	0.4267	0.8466	0.8026	0.4006	0.8122
0.3453	0.4419	0.8634	0.6074	0.4197	0.8356	0.9001	0.3932	0.7991
				Acetone (1) + Propyl Propionate (2)				
0.1139	0.6758	0.8695	0.4140	0.5777	0.8504	0.7171	0.4769	0.8242
0.2335	0.6362	0.8623	0.5275	0.5399	0.8406	0.8056	0.4475	0.8123
0.3062	0.6124	0.8575	0.5916	0.5188	0.8355	0.9271	0.4075	0.7961
				Methyl Ethyl Ketone (1) + Propyl Propionate (2)				
0.1074	0.6833	0.8697	0.3934	0.6130	0.8521	0.6832	0.5432	0.8303
0.2725	0.6426	0.8600	0.4885	0.5902	0.8455	0.7824	0.5192	0.8225
0.3326	0.6283	0.8562	0.5907	0.5656	0.8378	0.8867	0.4979	0.8116
				Methyl Ethyl Ketone (1) + Ethyl Acetate (2)				
0.1389	0.4703	0.8818	0.3977	0.4667	0.8582	0.6875	0.4654	0.8310
0.2272	0.4696	0.8741	0.4882	0.4662	0.8499	0.7766	0.4654	0.8220
0.3167	0.4673	0.8656	0.5812	0.4657	0.8409	0.8850	0.4654	0.8121
				Ethyl Acetate (1) + 1-Chlorohexane (2)				
0.1012	0.7447	0.8736	0.4022	0.6363	0.8765	0.7090	0.5432	0.8828
0.2179	0.7005	0.8742	0.4898	0.6080	0.8778	0.7947	0.5209	0.8855
0.3239	0.6626	0.8754	0.5960	0.5756	0.8799	0.8896	0.4964	0.8890
				Ethyl Acetate (1) + 1-Chlorohexadecane (2)				
0.1189	0.5212	0.8610	0.4035	3.067	0.8638	0.7005	1.482	0.8706
0.2139	4.420	0.8619	0.5154	2.402	0.8656	0.8046	1.066	0.8753
0.3510	3.419	0.8632	0.6163	0.1872	0.8680	0.8902	0.7743	0.8812
				Propyl Propionate (1) + 1-Chlorohexane (2)				
0.1046	0.7730	0.8726	0.4154	0.7475	0.8727	0.7033	0.7252	0.8736
0.2146	0.7629	0.8727	0.5138	0.7379	0.8729	0.7966	0.7197	0.8740
0.3112	0.7543	0.8727	0.6186	0.7310	0.8732	0.8925	0.7165	0.8746
				Propyl Propionate (1) + 1-Chlorohexadecane (2)				
0.1119	5.323	0.8602	0.4290	3.065	0.8625	0.6978	1.716	0.8657
0.2107	4.534	0.8612	0.5030	2.650	0.8631	0.8019	1.333	0.8678
0.3075	4.042	0.8617	0.6009	2.156	0.8642	0.8902	1.025	0.8705

Table 3. UNIFAC-VISCO Group Interaction Parameters α_{nm}

	CH_2	CH_3	CH_{2cy}	CH_{ar}	Cl	CO	COO
CH_2	0	66.53	224.9	406.7	60.30	859.5	1172
CH_3	−709.5	0	−130.7	−119.5	82.41	11.86	−172.4
CH_{2cy}	−538.1	187.3	0	8.958	251.4	−125.4	−165.7
CH_{ar}	−623.7	237.2	50.89	0	177.2	128.4	−49.85
Cl	−710.3	375.3	−163.3	−139.8	0	−404.3	−525.4
CO	586.2	−21.56	740.6	−117.9	−4.145	0	29.20
COO	541.6	−44.25	416.2	−36.17	240.5	22.92	0

Table 4. Comparison of the Experimental (ν_{exptl}) and Calculated (ν_{calcd}) Values of the Kinematic Viscosity for All Systems (N_p Measurements) Using the UNIFAC-VISCO Model AAD (av abs dev) = $1/N_p\Sigma|\nu_{exptl} - \nu_{calcd}|/\nu_{exptl} \times 100$

class	number of measurements	AAD
ketone/cyclalkane	27	2.17
1-chloroalkane/cyclalkane	38	2.82
ester/cyclalkane	36	2.58
ketone/aromatic	30	0.96
1-chloroalkane/aromatic	30	1.71
ester/aromatic	30	1.20
ketone/1-chloroalkane	36	3.41
ketone/ester	36	0.32
ester/1-chloroalkane	36	2.68

and m are the different functional groups engaged in the molecules.

$$\psi^*_{nm} = \exp(-\alpha_{nm}/T) \quad (6)$$

The new group interaction parameters $\alpha_{nm}(T)$ are given in Table 3.

The comparison of the experimental and calculated values of the kinematic viscosity for all systems using the UNIFAC-VISCO model is shown in Table 4.

The deviation between experimental and calculated values can seem large for some mixtures, in particular for ketone/1-chloroalkane. But one must take into account the fact that the UNIFAC-VISCO model is a predictive one which does not require the determination of adjustable parameters that would reduce this deviation.

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