Densities and Viscosities of *exo*-Tetrahydrodicyclopentadiene + *n*-Butanol and *exo*-Tetrahydrodicyclopentadiene + *n*-Pentanol at Temperatures of (293.15 to 313.15) K

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Densities and viscosities of *exo*-tetrahydrodicyclopentadiene + *n*-butanol and *exo*-tetrahydrodicyclopentadiene + *n*-pentanol were investigated over the whole composition range at several temperatures, (293.15, 298.15, 303.15, and 313.15) K, and atmospheric pressure. The excess molar volumes V_m^E for the mixtures were calculated based on these experimental data. The calculated quantities of V_m^E were fitted to the Redlich−Kister type polynomial equation, and the regression coefficients and the standard deviations of the fits were given. Several semiempirical equations were used to correlate the viscosity data, and the double-parameter McAllister equation gives satisfactory results.

Introduction

exo-Tetrahydrodicyclopentadiene ($C_{10}H_{16}$, tricyclo[5.2.1.$0^{2,6}$]-decane) is a synthetic hydrocarbon fuel, also called JP-10. Due to its strained cyclic structure, it has a high volumetric energy density and is used in volume limited applications. This characteristic as well as the suitable flashpoint and low freezing point[1,2] have made it the only air-breathing missile fuel used by the Navy and the Air Force at the present time. One problem with *exo*-tetrahydrodicyclopentadiene is that its properties of ignition and combustion are poor for efficient combustion in applications such as ramjets.

n-Alkanols are widely used in the chemical industry as a solvent, petrol formulation as enhancement of octane rating, and fuel additives.[3] *n*-Butanol is a kind of bioenergy alternative to petroleum fuels.[4,5] Due to its good miscibility and octane improving power, this alternative fuel also has a chance of playing an important role in promoting the combustion process of hydrocarbon fuel as additives. Accurate property data for *exo*-tetrahydrodicyclopentadiene + *n*-alkanol can guide research of high density fuel.

In the present work, two *n*-alkanols (*n*-butanol and *n*-pentanol) were added into *exo*-tetrahydrodicyclopentadiene, respectively, and the densities and viscosities of the binary mixtures were measured over the entire concentration range at four temperatures. From the experimental results, the excess molar volumes were calculated and fitted to a Redlich−Kister type equation. The excess properties derived from these physical property data can provide important information about the molecular structure of the liquid and intermolecular interactions in mixtures.[3]

Experimental Section

Materials and Characterization. *exo*-Tetrahydrodicyclopentadiene is supplied by Liming Research Institute of Chemical Industry with mass fraction purity better than 0.98 analyzed by a Hewlett-Packard 6890/5973 GC/MS. The chemical composition is listed in Table 1. The *n*-alkanols (*n*-butanol and *n*-pentanol) with mass fraction purity better than 0.99 are obtained from TOKYO Chemical Industry Company. All reagents were used without further purification. The densities and viscosities of the pure compounds were measured at 298.15 K and atmospheric pressure and compared with their corresponding literature values shown in Table 2.

Table 1. Chemical Composition of the *exo*-Tetrahydrodicyclopentadiene (JP-10) Sample

component	mass fraction
tricyclo[5.2.1.$0^{2,6}$] decane	0.9806
bicyclo[4.4.0]decane	0.0113
tricyclo[3.3.1.$1^{3,7}$]decane	0.0081

Table 2. Densities, ρ, and Viscosities, η, of Pure Compounds at 298.15 K

	ρ/g·cm^{-3}		η/mPa·s	
compound	exptl	lit.	exptl	lit.
exo-tetrahydrodicyclopentadiene	0.9312	0.9318[a]	2.7696	2.7673[a]
n-butanol	0.8056	0.8056[b]	2.5612	2.5609[c]
n-pentanol	0.8115	0.8116[d]	3.5344	3.5128[e]

[a] Ref 6. [b] Ref 7. [c] Ref 8. [d] Ref 9. [e] Ref 10.

Apparatus and Procedure. The binary mixtures were prepared by mass using a Mettler Toledo AL204 balance with a stated precision of ± 1·10^{-4} g, and a special stoppered glass bottle was used to avoid evaporation and reduce possible errors in mole fraction calculations. Hence, the uncertainty in the mole fractions was within ± 2.2·10^{-4}. Then, densities and viscosities were measured immediately.

The densities of the pure liquids and their binary mixtures were measured using a vibrating-tube digital densimeter (DMA55 Anton Paar), and the measurement cell temperature was controlled to better than ± 0.01 K by a thermostat. In a typical run, the densimeter was calibrated with double distilled water and dried air. The uncertainty of density measurements was ± 5·10^{-5} g·cm^{-3}, corresponding to an uncertainty of ± 3·10^{-3} cm^3·mol^{-1} in the excess volumes, V_m^E.

The kinematic viscosities were determined at atmospheric pressure and different temperature, using an Ubbelohde viscometer which was calibrated with double distilled water.

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Table 3. Densities, ρ, Viscosities, η, and Excess Molar Volumes, V_m^E, of *exo*-Tetrahydrodicyclopentadiene + *n*-Alkanol Mixtures at (293.15, 298.15, 303.15, and 313.15) K for Mole Fraction *x*

$x_{n\text{-alkanol}}$	ρ/g·cm^{-3}	η/mPa·s	V_m^E/cm^3·mol^{-1}	$x_{n\text{-alkanol}}$	ρ/g·cm^{-3}	η/mPa·s	V_m^E/cm^3·mol^{-1}
exo-Tetrahydrodicyclopentadiene + *n*-Butanol				*exo*-Tetrahydrodicyclopentadiene + *n*-Pentanol			
T = 293.15 K				T = 293.15 K			
0.0000	0.9356	3.076	0.0000	0.0000	0.9356	3.076	0.0000
0.1697	0.9201	2.796	0.1607	0.1465	0.9206	2.919	0.1843
0.3148	0.9053	2.675	0.2714	0.2782	0.9064	2.901	0.3169
0.4378	0.8915	2.646	0.3425	0.3987	0.8927	2.954	0.4028
0.5481	0.8779	2.660	0.3835	0.5049	0.8799	3.058	0.4674
0.6472	0.8647	2.694	0.3975	0.6067	0.8672	3.190	0.4928
0.7336	0.8523	2.743	0.3810	0.6985	0.8553	3.372	0.4761
0.8102	0.8407	2.807	0.3289	0.7827	0.8440	3.549	0.4292
0.8799	0.8295	2.855	0.2513	0.8623	0.8333	3.718	0.3071
0.9439	0.8191	2.906	0.1154	0.9326	0.8236	3.916	0.1722
1.0000	0.8091	2.940	0.0000	1.0000	0.8141	4.060	0.0000
T = 298.15 K				T = 298.15 K			
0.0000	0.9312	2.770	0.0000	0.0000	0.9312	2.770	0.0000
0.1697	0.9161	2.513	0.1178	0.1465	0.9168	2.621	0.1405
0.3148	0.9014	2.397	0.2279	0.2782	0.9027	2.595	0.2753
0.4378	0.8877	2.355	0.2949	0.3987	0.8892	2.622	0.3663
0.5481	0.8743	2.355	0.3315	0.5049	0.8767	2.700	0.4216
0.6472	0.8612	2.372	0.3370	0.6067	0.8642	2.807	0.4405
0.7336	0.8490	2.416	0.3112	0.6985	0.8525	2.953	0.4211
0.8102	0.8374	2.465	0.2655	0.7827	0.8413	3.095	0.3800
0.8799	0.8264	2.505	0.1844	0.8623	0.8308	3.270	0.2570
0.9439	0.8156	2.543	0.0886	0.9326	0.8212	3.411	0.1206
1.0000	0.8056	2.561	0.0000	1.0000	0.8115	3.534	0.0000
T = 303.15 K				T = 303.15 K			
0.0000	0.9276	2.519	0.0000	0.0000	0.9276	2.519	0.0000
0.1697	0.9126	2.282	0.0911	0.1465	0.9131	2.379	0.1248
0.3148	0.8981	2.177	0.1803	0.2782	0.8992	2.346	0.2380
0.4378	0.8845	2.128	0.2417	0.3987	0.8856	2.354	0.3322
0.5481	0.8710	2.118	0.2910	0.5049	0.8730	2.410	0.3928
0.6472	0.8579	2.135	0.3017	0.6067	0.8605	2.487	0.4068
0.7336	0.8457	2.165	0.2801	0.6985	0.8487	2.607	0.3871
0.8102	0.8341	2.211	0.2410	0.7827	0.8376	2.726	0.3321
0.8799	0.8231	2.242	0.1555	0.8623	0.8270	2.852	0.2214
0.9439	0.8123	2.272	0.0608	0.9326	0.8173	2.971	0.0932
1.0000	0.8020	2.287	0.0000	1.0000	0.8073	3.058	0.0000
T = 313.15 K				T = 313.15 K			
0.0000	0.9193	2.094	0.0000	0.0000	0.9193	2.094	0.0000
0.1697	0.9045	1.882	0.0709	0.1465	0.9050	1.972	0.1154
0.3148	0.8900	1.773	0.1461	0.2782	0.8911	1.920	0.2134
0.4378	0.8764	1.708	0.1980	0.3987	0.8776	1.914	0.3023
0.5481	0.8629	1.685	0.2488	0.5049	0.8651	1.922	0.3517
0.6472	0.8498	1.682	0.2671	0.6067	0.8525	1.957	0.3692
0.7336	0.8374	1.700	0.2559	0.6985	0.8408	2.032	0.3483
0.8102	0.8258	1.719	0.2195	0.7827	0.8297	2.107	0.2903
0.8799	0.8147	1.743	0.1396	0.8623	0.8191	2.180	0.1773
0.9439	0.8039	1.766	0.0456	0.9326	0.8093	2.253	0.0675
1.0000	0.7934	1.767	0.0000	1.0000	0.7992	2.300	0.0000

Table 4. Parameters of the Redlich−Kister Equation and Standard Deviations of the Fits σ of *exo*-Tetrahydrodicyclopentadiene + *n*-Alkanol Mixtures

T/K	A_0	A_1	A_2	A_3	σ
exo-Tetrahydrodicyclopentadiene + *n*-Butanol					
293.15	1.48698	0.98523	0.39299	−0.55249	0.01046
298.15	1.29151	0.77166	−0.04866	−0.32175	0.00619
303.15	1.11048	1.04945	−0.09713	−1.05227	0.01145
313.15	0.94722	1.17814	−0.05567	−1.39435	0.01502
exo-Tetrahydrodicyclopentadiene + *n*-Pentanol					
293.15	1.86755	0.94962	0.35164	−0.32806	0.00576
298.15	1.71735	1.03895	−0.17067	−0.75711	0.01435
303.15	1.57970	1.06068	−0.34413	−1.04726	0.01133
313.15	1.43590	0.99992	−0.45668	−1.27447	0.01091

The viscometer was kept in a well-stirred water bath controlled automatically within ± 0.1 K of the set value. Flow times were always > 200 s by selecting an appropriate viscometer, which was obtained by more than three times parallel determination, using an accurate stopwatch to an accuracy of ± 0.01 s. The dynamic viscosity η of the liquid was then calculated according to the following equation

$$\eta = \upsilon\rho \tag{1}$$

where ν is the kinematic viscosity; η is the dynamic viscosity; and ρ is the density. The uncertainty of the viscosity measurements was within ± 0.5 %.

Results and Discussion

Density and Viscosity Measurement. Experimental values of density ρ and viscosity η of all binary mixtures at different temperatures and atmospheric pressure are given in Table 3. The excess molar volumes V_m^E can be calculated from the experimental results using the equation

$$V_m^E = \frac{M_1x_1 + M_2x_2}{\rho_m} - \left(\frac{M_1x_1}{\rho_1} + \frac{M_2x_2}{\rho_2}\right) \tag{2}$$

In the equation, x_1 and x_2 are the mole fractions; M_1 and M_2 are the molar masses; and ρ_m, ρ_1, and ρ_2 are the densities of the mixture and of the pure components 1 (*n*-butanol or

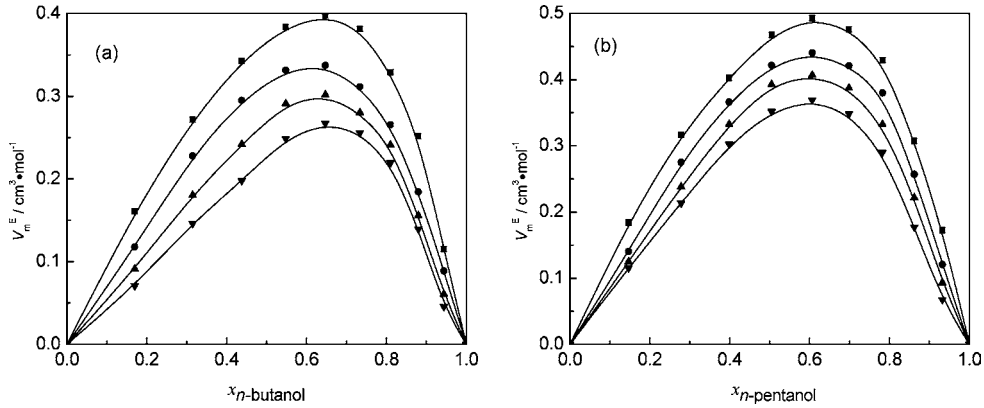

Figure 1. Excess molar volumes V_m^E as a function of mole fraction x for (a) (*exo*-tetrahydrodicyclopentadiene + *n*-butanol) and (b) (*exo*-tetrahydrodicyclopentadiene + *n*-pentanol) at temperatures T. ■, 293.15 K; ●, 298.15 K; ▲, 303.15 K; ▼, 313.15 K.

Table 5. Correlation Deviations and Parameters of Semi-Empirical Viscosity Equations for *exo*-Tetrahydrodicyclopentadiene + *n*-Alkanol Systems at (293.15, 298.15, 303.15, and 313.15) K

		T/K							
		293.15	298.15	303.15	313.15	293.15	298.15	303.15	313.15
		exo-Tetrahydrodicyclopentadiene + *n*-Butanol				*exo*-Tetrahydrodicyclopentadiene + *n*-Pentanol			
Kendall−Monroe	Dev/%	7.22	7.01	6.93	7.17	8.92	8.94	8.41	7.57
	σ/mPa•s	0.2384	0.2384	0.2384	0.2384	0.3344	0.2969	0.2501	0.1804
Grunberg−Nissan	A_{12}	−0.5772	−0.5830	−0.5777	−0.5993	−0.6599	−0.6788	−0.6607	−0.6187
	Dev/%	1.59	1.77	1.75	1.88	1.73	1.80	1.91	0.02
	σ/mPa•s	0.0582	0.0566	0.0498	0.0396	0.0794	0.0729	0.0670	0.0502
Grunberg−Nissan ($A_{12} = 0$)	Dev/%	7.21	6.99	6.91	7.10	8.71	8.77	8.30	7.55
	σ/mPa•s	0.2382	0.2061	0.1836	0.1504	0.3266	0.2916	0.2471	0.1799
Hind	A_{12}	2.1979	1.9356	1.7486	1.3768	2.4403	2.1392	1.9221	1.5639
	Dev/%	1.61	1.81	1.82	1.92	1.20	1.38	1.59	1.72
	σ/mPa•s	0.0574	0.0574	0.0512	0.0424	0.0536	0.0528	0.0535	0.0444
Hind ($A_{12} = 0$)	Dev/%	24.67	24.87	24.94	24.91	23.62	23.70	24.13	24.81
	σ/mPa•s	0.7855	0.7001	0.6325	0.5016	0.8774	0.7765	0.7035	0.5742
Katti−Chaudry	W_{vis}/RT	−0.4672	−0.4732	−0.4687	−0.4922	−0.6058	−0.6240	−0.6064	−0.5651
	Dev/%	1.79	1.94	1.91	2.00	1.82	1.86	1.95	1.92
	σ/mPa•s	0.0657	0.0621	0.0543	0.0428	0.0839	0.0757	0.0688	0.0510
Katti−Chaudry ($x_1x_2W_{vis} = 0$)	Dev/%	5.28	5.11	5.05	5.27	7.73	7.82	7.38	6.67
	σ/mPa•s	0.1793	0.1549	0.1381	0.1144	0.2911	0.2609	0.2208	0.1598
McAllister	A_{12}	1.1173	0.9837	0.8720	0.6077	1.3001	1.1598	1.0301	0.7590
	A_{21}	0.7958	0.6876	0.5904	0.3856	0.9193	0.8087	0.7106	0.5263
	Dev/%	0.49	0.34	0.33	0.18	0.31	0.24	0.23	0.27
	σ/mPa•s	0.0189	0.0118	0.0099	0.0044	0.0155	0.0104	0.0089	0.0092

n-pentanol) and 2 (*exo*-tetrahydrodicyclopentadiene), respectively. The calculated values of V_m^E are reported in Table 3.

The values of excess molar volumes V_m^E for each mixture, at each investigated temperature, were fitted to the Redlich−Kister type polynomial equation

$$Y = x_1x_2\sum_{i=0}^{K} A_i(x_1 - x_2)^i \qquad (3)$$

where Y is V_m^E; x_i is the mole fraction of component i in the mixture; and A_i are the fitting coefficients obtained by the least-squares method. Table 4 shows the best fitting parameters for each of the binary mixtures at all four temperatures, together with the standard deviation of the fit σ, which was defined as

$$\sigma = \left[\frac{\sum (Y_{exptl} - Y_{calcd})^2}{n - k}\right]^{1/2} \qquad (4)$$

where n is the total number of measurements and k is the number of estimated parameters.

Figure 1 shows the variations of V_m^E with the mole fraction of the *n*-alkanols at different temperatures.

As noted from Figure 1, for both systems studied, the excess molar volume V_m^E values are found to be positive over the entire composition range at the four temperatures. The variations can be discussed in terms of physical, chemical, and molecular structural factors. The physical interactions include electrostatic forces, induction forces, and dispersion forces, which make a weak contribution to V_m^E. The chemical factors are specific forces between molecules, such as charge transfer and hydrogen bonds, breaking of hydrogen bonds, and complexes giving positive excess molar volumes. The structural characteristics arise from the geometrical fitting of one component into another's structure, due to the differences in shape and size of components and free volume.[11,12]

exo-Tetrahydrodicyclopentadiene is a cyclic alkane, and there is a possibility that small chain molecules may enter into the interstitial accommodation of *exo*-tetrahydrodicyclopentadiene leading to contraction in volume. Nevertheless, for the systems of *exo*-tetrahydrodicyclopentadiene + *n*-butanol and *exo*-tetrahydrodicyclopentadiene + *n*-pentanol, the −OH group of *n*-alkanols can weaken this negative contribution. Hydrogen bonds are the main factors which affect the excess volumes. The molecules of *n*-alkanols are self-associated through hydrogen bonding in a pure state, and breaking up of intermolecular hydrogen bonds by *exo*-tetrahydrodicyclopentadiene gives positive deviation.[13] Therefore, the positive values of the excess molar volume can be attributed to the predominance of the rupture of hydrogen bonding. The effect of temperature on V_m^E

is noteworthy. There is a systematic decrease in V_m^E values with rise in temperature for both systems. A decrease in V_m^E values with increasing temperature suggests that the hydrogen bonds are weakened, and more declustering appears in pure *n*-alkanols at higher temperature.

Viscosity Data Correlation. Many semitheoretical and empirical equations for the viscosity of pure components and mixtures have been used to correlate the experimental data.[14,15] In the present work, five typical semiempirical relations are selected. The Kendall−Monroe equation, which has no adjustable parameter, was given by

$$\eta = (x_1\eta_1^{1/3} + x_2\eta_2^{1/3})^3 \quad (5)$$

The single-parameter correlations of Grunberg−Nissan, Hind, and Katti−Chaudry[16] are eqs 6, 7, and 8, respectively.

$$\ln \eta = x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_1x_2A_{12} \quad (6)$$

$$\eta = x_1^2\eta_1 + x_2^2\eta_2 + 2x_1x_2A_{12} \quad (7)$$

$$\ln \eta V = x_1 \ln \eta_1V_1 + x_2 \ln \eta_2V_2 + x_1x_2W_{vis}/RT \quad (8)$$

where V_i is the molar volume of pure component *i* and W_{vis} is an adjustable parameter.

The double-parameter McAllister equation was defined as

$$\ln \eta = x_1^3 \ln \eta_1 + x_2^3 \ln \eta_2 + 3x_1^2x_2 \ln A_{12} + 3x_1x_2^2 \ln A_{21} - \ln[x_1 + x_2M_2/M_1] + 3x_1^2x_2 \ln[2 + (M_2/M_1)/3] + 3x_1x_2^2 \ln[1 + (2M_2/M_1)/3] + x_2^3 \ln[M_2/M_1] \quad (9)$$

where A_{12} and A_{21} are interaction parameters.

The correlation parameters and the standard deviation for binary mixtures are listed in Table 5, together with the average percentage deviation that was given by

$$\mathrm{Dev} = \frac{1}{N}\sum_{i=1}^{N}\frac{|\eta_{calcd} - \eta_{exptl}|}{\eta_{exptl}} \cdot 100\ \% \quad (10)$$

where *N* is the number of experimental points.

Grunberg−Nissan, Hind, and Katti−Chaudry equations give comparatively similar deviations. The Kendall−Monroe equation gives large prediction deviations, which is not appropriate for the investigated binary mixtures. The double-parameter McAllister equation gives satisfactory results with deviations from (0.1 to 0.5) %.

Conclusions

Densities and viscosities of *exo*-tetrahydrodicyclopentadiene + *n*-butanol and *exo*-tetrahydrodicyclopentadiene + *n*-pentanol were measured over the whole composition range at several temperatures and atmospheric pressure. The excess molar volumes were calculated and correlated by the Redlich−Kister type polynomial equation. Positive values of excess molar volumes at all temperatures and over the entire range of compositions were obtained for two systems, which decrease with rising temperature from (293.15 to 313.15) K. The viscosity data were correlated with five semiempirical equations, and the McAllister equation gives satisfactory results.

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