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Density, Refractive Index, Viscosity, and Surface Tension of Binary Mixtures of *exo*-Tetrahydrodicyclopentadiene with Some *n*-Alkanes from (293.15 to 313.15) K

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ABSTRACT: Densities and viscosities were determined over the whole composition range for the binary mixtures of *exo*-tetrahydrodicyclopentadiene with *n*-nonane, *n*-dodecane, or *n*-tridecane at different 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 from the experimental data. The refractive index and surface tension of the systems were also measured for temperatures of (293.15 and 303.15) K.

INTRODUCTION

exo-Tetrahydrodicyclopentadiene (*exo*-tricyclo[5.2.1.0$^{2.6}$]decane) is the principal component of the high-energy density hydrocarbon fuel commonly identified as JP-10. The compound's strained cyclic geometry allows for efficient energy storage and high thermal stability; it can be used as a missile fuel.[1−3] However, one problem with JP-10 is that its properties of ignition and combustion can be too poor in applications such as ramjets.[4] Alkanes are an important series of homologous, nonpolar, organic solvents and are widely used as an enhancement of octane rating and fuel additives to enhance combustion rates.[5]

Some experimental data about JP-10 and *n*-alkanes have been reported in the literature.[6−14] Our previous research has presented some physical properties of several binary systems, such as JP-10 + *n*-undecane and *n*-tetradecane.[15] In the present work, the physical properties (densities, viscosities, refractive index, and surface tension) of the binary mixtures of JP-10 + *n*-nonane, *n*-dodecane, and *n*-tridecane were measured at several temperatures and atmospheric pressure as fundamental data. Furthermore, the excess molar volumes (V_m^E) of these systems were calculated, which can be used to analyze and discuss the nature and strength of intermolecular interactions in binary mixtures.

EXPERIMENTAL SECTION

Materials and Characterization. *N*-Nonane is supplied by Aladdin Chemistry Co. Ltd., Shanghai, China, and *n*-dodecane and *n*-tridecane are supplied by J&K Chemical Reagent Company, Shanghai, China. Their mass fraction purities are better than 99 %. JP-10 is obtained from Liming Research Institute of Chemical Industry in China with a mass fraction purity better than 98 %. All reagents were used without further purification.

Apparatus and Procedure. The mixtures were prepared by mass using a Mettler Toledo AL204 balance with a stated precision of $\pm 1 \cdot 10^{-4}$ g. A special stoppered glass bottle was used to avoid evaporation in mass measurement, after which densities and viscosities were measured immediately. The uncertainty in the mole fractions was $\pm 1 \cdot 10^{-4}$.

The densities, ρ, of these pure liquids and their binary mixtures were measured by DMA 5000 M density meter, Anton Paar at several temperatures, (293.15, 298.15, 303.15, and 313.15) K, and the repeatability in the density and temperature measurements provided by the manufacturer is $\pm 1 \cdot 10^{-6}$ g·cm^{-3} and $\pm$ 0.001 K, respectively. The uncertainty of the thermometer and the density measurements is $\pm$ 0.01 K and $\pm 5 \cdot 10^{-6}$ g·cm^{-3}, corresponding to the uncertainty of $\pm 1 \cdot 10^{-3}$ cm^3·mol^{-1} in the excess volume, V_m^E.

Refractive index, n_D, was determined at temperatures (293.15 and 303.15) K using a WAY-2S refractometer with a precision of 0.0001 at the wavelength of the D line of sodium, 589.3 nm. The temperature of the liquid was maintained constant by circulating water from the HD-501S thermostatic water bath. The calibration of the instrument was carried out by deionized water. For rapid achievement of thermal equilibrium, samples were kept in an external thermostat at the constant temperature of the refractometer before measurement. Each refractive index value reported came from an average of three measurements, and the uncertainty of the measurement was $3 \cdot 10^{-4}$.

The surface tension, γ, was determined by employing a dropmeter a-100p tensiometer with temperature control within $\pm$ 0.01 K. In this technique, the primary quantity measured is the volume of a drop that detaches from a capillary of known diameter joined to a syringe containing the sample. As the syringe plunger is depressed mechanically, a drop forms at the tip of the capillary and grows until its weight reaches a critical value that cannot be counterbalanced by the surface tension, whereupon it falls. The critical volume V is related to γ by:

$$\gamma = \frac{V\rho g}{R}F \qquad (1)$$

where ρ is the density of mixture, g is the gravitational acceleration constant, R is the radius of the capillary, and F is a correction factor supplied by the manufacturers. The samples were thermostatted in a closed vessel before the surface tension measurements. An average of five measurements was taken for each sample mixture, after each measurement, the syringe and

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Table 1. Densities, ρ, Excess Molar Volumes, V_m^E, of JP-10 + n-Alkane Mixtures at (293.15, 298.15, 303.15, and 313.15) K

$x_{n\text{-alkane}}$	ρ / $g \cdot cm^{-3}$	V_m^E / $cm^3 \cdot mol^{-1}$	ρ / $g \cdot cm^{-3}$	V_m^E / $cm^3 \cdot mol^{-1}$
JP-10 + n-Nonane				
	T/ K = 293.15		T/ K = 298.15	
0.0000	0.935285	0.000	0.931374	0.000
0.1089	0.907615	−0.119	0.903689	−0.122
0.2212	0.880128	−0.193	0.876200	−0.200
0.3244	0.855834	−0.233	0.851910	−0.242
0.4159	0.835074	−0.247	0.831154	−0.258
0.5099	0.814465	−0.243	0.810552	−0.255
0.6161	0.792015	−0.221	0.788111	−0.233
0.7128	0.772324	−0.185	0.768429	−0.196
0.8078	0.753664	−0.139	0.749778	−0.149
0.9133	0.733691	−0.077	0.729814	−0.084
1.0000	0.717780	0.000	0.713899	0.000
	T/ K = 303.15		T/ K = 313.15	
0.0000	0.927462	0.000	0.919624	0.000
0.1089	0.899772	−0.127	0.891914	−0.138
0.2212	0.872280	−0.209	0.864411	−0.228
0.3244	0.847988	−0.254	0.840115	−0.278
0.4159	0.827234	−0.271	0.819360	−0.299
0.5099	0.806634	−0.269	0.798763	−0.299
0.6161	0.784195	−0.246	0.776330	−0.275
0.7128	0.764515	−0.208	0.756656	−0.235
0.8078	0.745866	−0.157	0.738014	−0.179
0.9133	0.725905	−0.088	0.718060	−0.101
1.0000	0.709996	0.000	0.702143	0.000
JP-10 + n-Dodecane				
	T/ K = 293.15		T/ K = 298.15	
0.0000	0.935285	0.000	0.931374	0.000
0.0803	0.912991	−0.026	0.909116	−0.028
0.1712	0.889888	−0.040	0.886051	−0.045
0.2563	0.870029	−0.030	0.866224	−0.036
0.3428	0.851430	−0.011	0.847656	−0.018
0.4443	0.831449	0.014	0.827708	0.006
0.5407	0.814086	0.044	0.810374	0.036
0.6527	0.795684	0.070	0.792001	0.063
0.7596	0.779776	0.058	0.776117	0.053
0.8855	0.762744	0.026	0.759110	0.023
1.0000	0.748595	0.000	0.744980	0.000
	T/ K = 303.15		T/ K = 313.15	
0.0000	0.927462	0.000	0.919624	0.000
0.0803	0.905235	−0.030	0.897449	−0.033
0.1712	0.882203	−0.049	0.874461	−0.051
0.2563	0.862406	−0.041	0.854719	−0.045
0.3428	0.843867	−0.024	0.836246	−0.030
0.4443	0.823952	−0.001	0.816406	−0.011
0.5407	0.806647	0.029	0.799185	0.012
0.6527	0.788301	0.057	0.780878	0.046
0.7596	0.772459	0.043	0.765081	0.035
0.8855	0.755457	0.021	0.748131	0.017
1.0000	0.741351	0.000	0.734070	0.000
JP-10 + n-Tridecane				
	T/ K = 293.15		T/ K = 298.15	
0.0000	0.935285	0.000	0.931374	0.000
0.0757	0.913895	−0.030	0.910022	−0.032
0.1546	0.893546	−0.037	0.889710	−0.040
0.2392	0.873654	−0.012	0.869856	−0.016
0.3284	0.854614	0.029	0.850853	0.024
0.4236	0.836311	0.067	0.832586	0.062
0.5258	0.818689	0.085	0.815000	0.079
0.6320	0.802316	0.069	0.798660	0.063
0.7477	0.786302	0.042	0.782677	0.036
0.8665	0.771478	0.020	0.767877	0.016
1.0000	0.756488	0.000	0.752909	0.000
	T/ K = 303.15		T/ K = 313.15	
0.0000	0.927462	0.000	0.919624	0.000
0.0757	0.906150	−0.034	0.898383	−0.036
0.1546	0.885874	−0.044	0.878182	−0.049
0.2392	0.866057	−0.021	0.858440	−0.028
0.3284	0.847091	0.019	0.839548	0.010
0.4236	0.828859	0.056	0.821391	0.045
0.5258	0.811307	0.072	0.803912	0.060
0.6320	0.794998	0.057	0.787671	0.044
0.7477	0.779043	0.030	0.771762	0.023
0.8665	0.764268	0.013	0.757041	0.009
1.0000	0.749324	0.000	0.742156	0.000

capillary were cleaned with ethanol. The uncertainty of the measurement was 0.1 $mN \cdot m^{-1}$, and the results of surface tension were accurate to 0.01 $mN \cdot m^{-1}$.

The dynamic viscosities, η, were determined using an AMVn viscometer (Anton Paar) at different temperatures and atmospheric pressure. The measurement cell temperature was controlled by a thermostat with a precision of ± 0.01 K, and an accuracy of the efflux time is ± 0.001 s; each viscosity value of the fluid was reported by averaging over two consecutive runs. The uncertainty of the viscosity measurements was within ± 0.001 mPa·s.

■ RESULTS AND DISCUSSION

Volumetric Properties. The experimental densities and excess molar volumes of the binary mixtures at different temperatures and atmospheric pressure are summarized in Table 1. The experimental values of densities are used to calculate the excess molar volumes, V_m^E, of the mixture as

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

In the equation, M_1 and M_2 are the molar masses; x_1 and x_2 are the mole fractions; and ρ_m, ρ_1, and ρ_2 are the densities of the mixture, where the component 1 is n-nonane, n-dodecane, or n-tridecane and the component 2 is JP-10.

The values of the excess molar volumes varying with the mole fraction of the *n*-alkanes are presented in Figure 1. It can be seen that values of V_m^E are negative for the binary mixture of JP-10 + *n*-nonane over the entire composition range, and the excess molar volume decreases with the increase in temperature. However, V_m^E values for *n*-dodecane or *n*-tridecane with JP-10 mixtures do not follow the trend, which is shown in Figure 1b,c. In general, excess volume mainly depends on two reasons: (1) variation of intermolecular forces when two components come into contact; (2) variation of molecular packing due to the differences in the shape and size of components.[16]

JP-10 is a cyclic alkane, and *n*-nonane, *n*-dodecane, and *n*-tridecane are chain alkanes. There is an opportunity for the chain molecule to enter into the interstitial space of JP-10, which can lead to a contraction in volume. On the other hand, compounds of the binary system are nonpolar molecules, and the weak dispersive interactions between molecules can also play a role in this aspect resulting in positive V_m^E value. For the system of JP-10 + *n*-nonane, the contribution of the dispersive interactions is weaker than that of structural characteristics to induce the negative excess molar volume. For the system of JP-10 + *n*-dodecane and *n*-tridecane, when the concentration of *n*-alkanes is low the dispersive interactions are weaker than structural characteristics resulting in the negative excess molar volume; nevertheless, the contribution of two effects varying

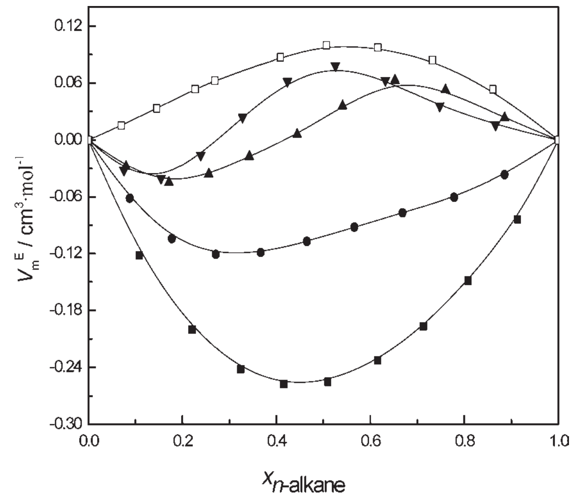

Figure 2. Plot of excess molar volume V_m^E vs the molar fraction x_1 for binary systems *n*-alkane/JP-10 at 298.15K, from this work: ■, *n*-nonane; ▲, *n*-dodecane; ▼, *n*-tridecane; and from the literature:[15] ●, *n*-undecane; □, *n*-tetradecane.

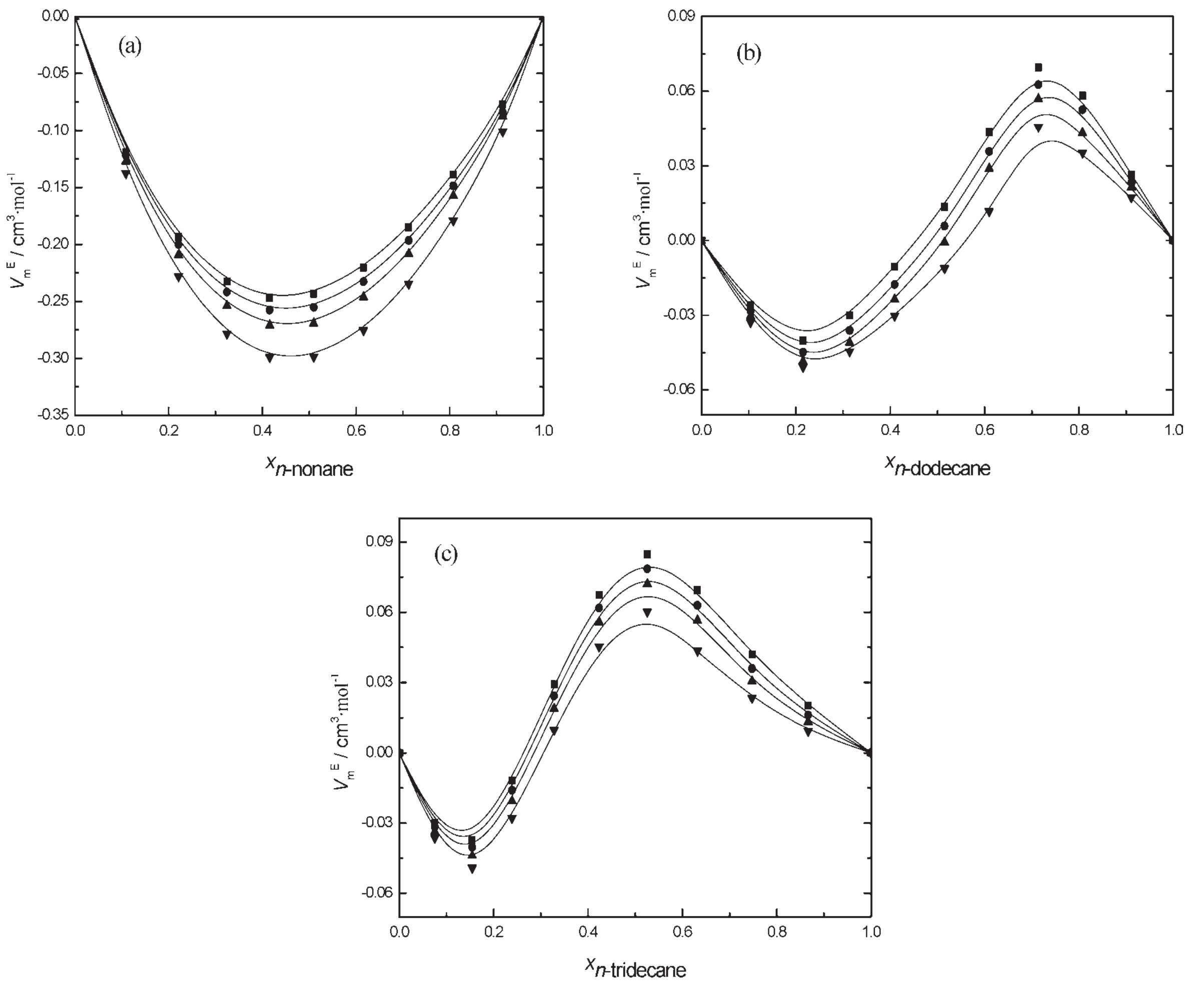

Figure 1. Excess molar volumes variation with mole fraction for the systems (a) JP-10 + *n*-nonane; (b) JP-10 + *n*-dodecane; (c) JP-10 + *n*-tridecane at different temperatures (T): ■, 293.15 K; ●, 298.15 K; ▲, 303.15 K; ▼, 313.15 K.

Table 2. Viscosities, η, of JP-10 + n-Alkane Mixtures at (293.15, 298.15, 303.15, and 313.15) K

$x_{n\text{-alkane}}$	η/ mPa·s			
	T/ K = 293.15	T/ K = 298.15	T/ K = 303.15	T/ K = 313.15
	JP-10 + n-Nonane			
0.0000	3.077	2.784	2.525	2.112
0.1089	2.455	2.229	2.040	1.728
0.2212	2.005	1.836	1.687	1.441
0.3244	1.683	1.545	1.426	1.227
0.4159	1.459	1.344	1.244	1.077
0.5099	1.277	1.176	1.091	0.951
0.6161	1.109	1.028	0.958	0.839
0.7128	0.985	0.917	0.856	0.754
0.8078	0.887	0.827	0.775	0.686
0.9133	0.795	0.744	0.699	0.622
1.0000	0.733	0.687	0.647	0.579
	JP-10 + n-Dodecane			
0.0000	3.077	2.784	2.525	2.112
0.0803	2.796	2.535	2.308	1.939
0.1712	2.540	2.307	2.104	1.775
0.2563	2.340	2.127	1.944	1.644
0.3428	2.169	1.974	1.807	1.532
0.4443	2.006	1.827	1.674	1.422
0.5407	1.881	1.715	1.572	1.337
0.6527	1.764	1.610	1.476	1.257
0.7596	1.672	1.526	1.400	1.193
0.8855	1.574	1.438	1.321	1.128
1.0000	1.507	1.377	1.266	1.084
	JP-10 + n-Tridecane			
0.0000	3.077	2.784	2.525	2.112
0.0757	2.880	2.609	2.370	1.975
0.1546	2.703	2.448	2.233	1.860
0.2392	2.552	2.311	2.112	1.759
0.3284	2.419	2.189	2.000	1.669
0.4236	2.302	2.082	1.904	1.587
0.5258	2.198	1.992	1.822	1.521
0.6320	2.108	1.913	1.748	1.466
0.7477	2.028	1.842	1.689	1.416
0.8665	1.963	1.789	1.631	1.369
1.0000	1.894	1.726	1.577	1.325

Table 3. Refractive Index, n_D, and Surface Tension, γ, of JP-10 + n-Alkane Mixtures at (293.15 and 303.15) K

$x_{n\text{-alkane}}$	n_D		γ/ mN·m^{-1}	
	T/ K = 293.15	T/ K = 303.15	T/ K = 293.15	T/ K = 303.15
	JP-10 + n-Nonane			
0.0000	1.4877	1.4832	31.86	31.09
0.1089	1.4777	1.4728	30.17	29.01
0.2212	1.4667	1.4627	27.92	27.45
0.3244	1.4570	1.4517	27.12	26.63
0.4159	1.4500	1.4450	26.34	25.74
0.5099	1.4422	1.4377	25.59	24.86
0.6161	1.4332	1.4282	24.35	23.36
0.7128	1.4260	1.4210	23.84	22.88
0.8078	1.4192	1.4139	23.14	22.39
0.9133	1.4114	1.4062	22.60	21.81
1.0000	1.4048	1.4001	22.20	21.41
	JP-10 + n-Dodecane			
0.0000	1.4877	1.4832	31.86	31.09
0.0803	1.4796	1.4733	30.13	29.37
0.1712	1.4711	1.4657	29.15	28.51
0.2563	1.4620	1.4571	28.63	27.82
0.3428	1.4573	1.4524	27.91	27.16
0.4443	1.4502	1.4455	27.32	26.51
0.5407	1.4448	1.4396	26.45	25.86
0.6527	1.4383	1.4333	25.98	25.28
0.7596	1.4325	1.4279	25.41	24.62
0.8855	1.4271	1.4214	24.51	23.82
1.0000	1.4219	1.4168	24.07	23.67
	JP-10 + n-Tridecane			
0.0000	1.4877	1.4832	31.86	31.09
0.0757	1.4795	1.4743	30.40	29.94
0.1546	1.4726	1.4683	29.63	28.98
0.2392	1.4662	1.4614	28.61	28.20
0.3284	1.4586	1.4541	28.03	27.21
0.4236	1.4528	1.4483	27.19	26.74
0.5258	1.4468	1.4420	26.68	25.72
0.6320	1.4407	1.4357	25.95	25.38
0.7477	1.4349	1.4305	25.54	24.83
0.8665	1.4306	1.4258	25.19	24.50
1.0000	1.4253	1.4213	24.79	24.08

with the concentration of n-alkanes is opposite leading to the positive V_m^E value.

The variation of V_m^E with the mole fraction, $x_{n\text{-alkane}}$, of all systems that include JP-10 + n-undecane or n-tetradecane (the previous work[15]) at 298.15 K are shown in Figure 2. It shows an interesting behavior: values of this parameter increase with the chain length of n-alkanes.

The effect of temperature on V_m^E is noteworthy. With the rise in temperature, the molecular motion becomes fiercer, so there is a systematic decrease in V_m^E values for the system of JP-10 + n-alkanes.

Viscosity Data Correlation. The study of viscosities has been found to be very useful in understanding the nature of the molecular interactions occurring within the binary mixtures. The viscosities of the binary mixtures at different temperatures and atmospheric pressure are summarized in Table 2. It can be noticed from Table 2 that the viscosity values decrease as temperature increases, and there is a decrease in the value of this physical property when the n-alkane concentration increases in the mixture (see Figure 3).

Other Properties. Refractive index and surface tension corresponding to the binary systems of JP-10 + n-alkanes were also determined for temperatures of (293.15 and 303.15) K, and the experimental data are presented in Table 3. Figure 4 shows the experimental behaviors obtained for these mixtures for refractive index and surface tension. An decrease in both refractive index and surface tension is observed when mixtures are enriched in n-alkane.

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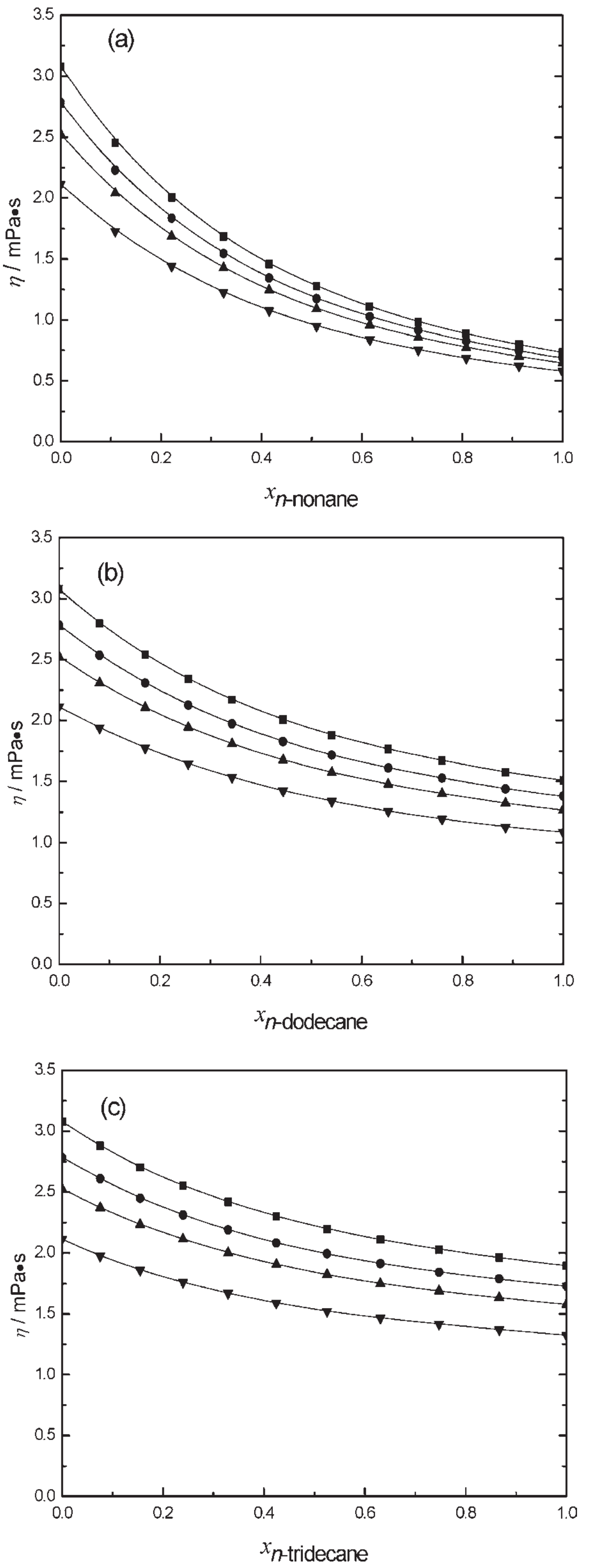

Figure 3. Viscosity variation with mole fraction for the systems (a) JP-10 + *n*-nonane; (b) JP-10 + *n*-dodecane; (c) JP-10 + *n*-tridecane at different temperatures (T): ■, 293.15 K; ●, 298.15 K; ▲, 303.15 K; ▼, 313.15 K.

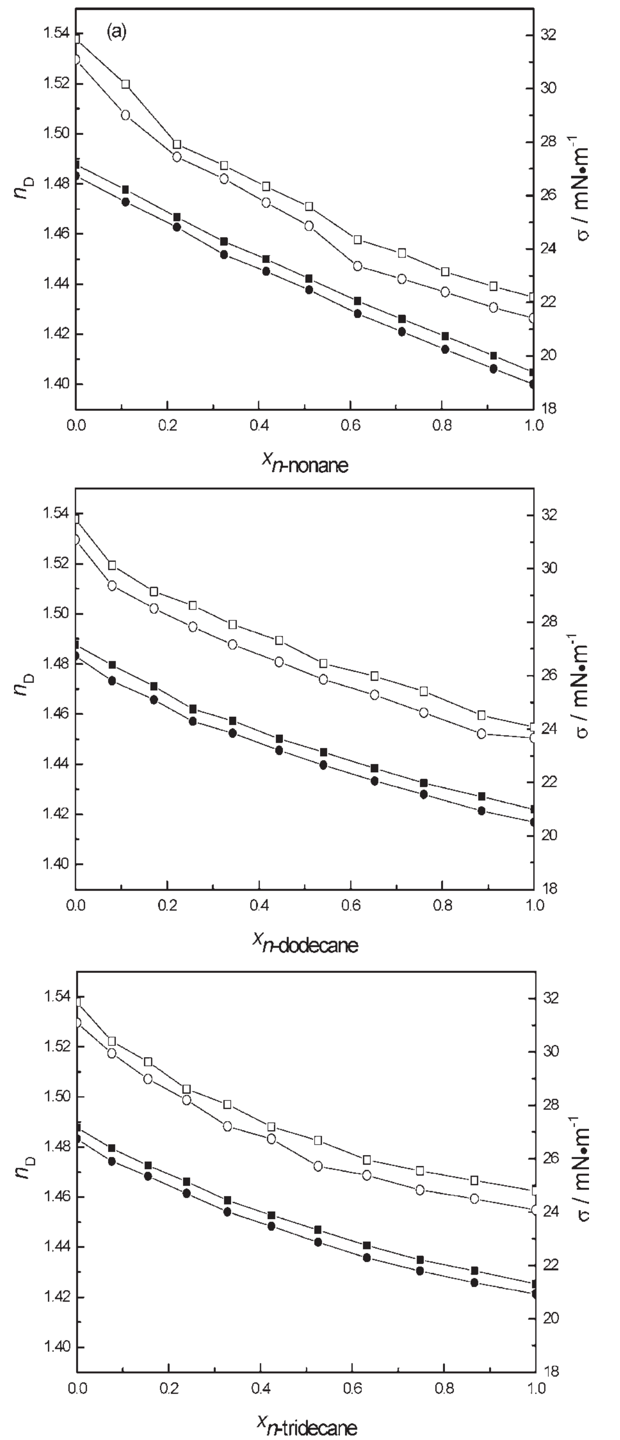

Figure 4. Influence of mixture composition and temperature on surface tension and refractive index for the systems (a) JP-10 + *n*-nonane; (b) JP-10 + *n*-dodecane; (c) JP-10 + *n*-tridecane. Refractive index: ■, 293.15 K; ●, 303.15 K; surface tension: □, 293.15 K; ○, 303.15 K.

An increase in temperature produces a decrease in both physical properties.

■ CONCLUSIONS

The V_m^E values are negative for the binary mixture of JP-10 with *n*-nonane, and the V_m^E values have minimum values at about a mole fraction of *n*-nonane $x \approx 0.4$, but for other two systems, the numerical V_m^E values of the excess molar volume change from negative to positive, there are minimum and maximum values. For the system of JP-10 + *n*-dodecane, the V_m^E values have minimum and maximum values at $x \approx 0.2$ and 0.7, respectively. For the system of JP-10 + *n*-tridecane, the extreme values are observed at $x \approx 0.15$ and $x \approx 0.5$, respectively. In relation with the value of viscosity, refractive index, and surface tension, a linear influence is caused by the composition, with a reduction in the value when the *n*-alkane concentration increases in systems. The temperature causes a slight decrease in the values.

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