

Viscosity and Density of Liquid Mixtures of *n*-Alkanes with Squalane¹

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Viscosity and density measurements are reported for binary liquid mixtures of *n*-butane and *n*-hexane with squalane in the temperature range from 273 to 333 K. The viscosity measurements have been carried out by using a capillary viscometer calibrated with standard liquids, that is, JS5, JS10, JS20, and water. The uncertainty in the viscosity data was estimated to be $\pm 1.7\%$. The density needed for the calculation of the viscosity has been measured by using a glass pycnometer within an accuracy of $\pm 0.04\%$. In the prediction of the viscosity, the scheme of Assael et al. fails for mixtures of this type differing greatly in size.

KEY WORDS: capillary viscometer; density; *n*-butane; *n*-hexane; squalane; viscosity.

1. INTRODUCTION

Precise information on thermophysical properties of the mixtures of new refrigerants with refrigeration oil is useful for developing prediction methods. Low molecular weight hydrocarbons such as propane and butane are environmentally acceptable refrigerants of recent interest in Europe. However, there are only a few studies which present the thermophysical properties of mixtures of hydrocarbons with oils.

In the present work, the viscosity and the density of mixtures of *n*-butane and *n*-hexane with squalane, which were intended to model mixtures of refrigerants with the refrigeration oil, were measured in the temperature range from 273 to 333 K.

¹ Paper presented at the Twelfth Symposium on Thermophysical Properties, June 19–24, 1994, Boulder, Colorado, U.S.A.

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2. EXPERIMENTS

Samples were provided by Aldrich Chemical Co. for *n*-hexane and by Tokyo Kasei Kogyo Co. for *n*-butane and squalane with purities of better than 99%. Mixtures were prepared by mixing weighed amounts of the pure liquids. The compositions of the liquid mixtures were calculated by considering vapor pressures which were mole-fraction averages of pure component values estimated by the Antoine equation [1].

The viscosity was measured using the capillary viscometer described earlier [2]. The viscometer was located in a water bath whose temperature was regulated to within 30 mK. The procedure was described in detail previously [3]. The viscosity η (mPa·s) was calculated from the measured efflux time t (s) with a reproducibility of 0.5% by using the following equation:

$$\eta = C_1 \rho t - C_2 \rho / t \quad (1)$$

where C_1 and C_2 are the apparatus constants, and ρ ($\text{kg} \cdot \text{m}^{-3}$) was the density of the sample liquid obtained by using the glass pycnometer, described earlier [4], with an accuracy of $\pm 0.04\%$. C_1 and C_2 were determined by using the capillary against standard liquids, that is, JS5, JS10, and JS20 (figures refer to viscosities in mPa·s at 293 K) calibrated by NRLM (Japan) and water, with an uncertainty of 0.4%. The uncertainties in the measured viscosities are estimated to be smaller than 1.7%. The measurements were performed at Reynolds numbers less than 670, which was below the critical value of 2300.

3. RESULTS

Density values of *n*-butane, *n*-hexane, and squalane obtained in the present work agree with the literature data [5–8] within 0.11% as shown in Table I. Most of the present viscosity values agree with the literature data [8–17] within 3% in spite of different experimental techniques, except for those of *n*-butane and *n*-hexane obtained by Kiran and Sen and for *n*-hexane by Oswal and Patel as shown in Figs. 1 and 2.

In Tables II and III, the present experimental data are listed for the density and viscosity of binary mixtures of *n*-butane and *n*-hexane with squalane. The viscosity isotherms of the mixtures are convex upward and decrease with the decrease in the mole fraction of squalane, x_s (Fig. 3).

One of the successful representations of the liquid viscosity is the hard-sphere theory proposed by Dymond [18]. Recently, Assael et al. [19] obtained the following expressions for correlating the viscosity of hydrocarbon mixture. The mixture behaves as a single-component liquid with

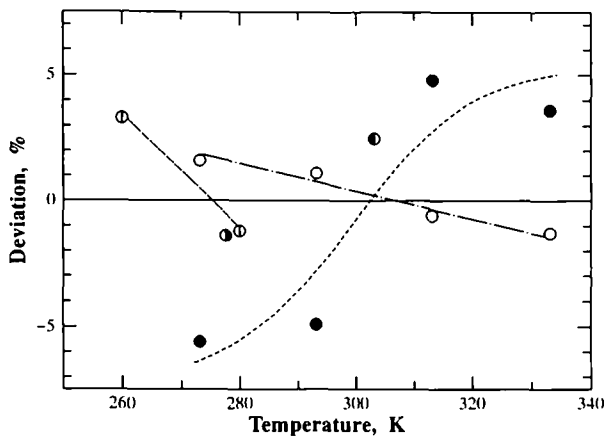


Fig. 1. Deviation of literature viscosity data of *n*-butane from the present work: (●) Ref. 9, rotating cylinder; (◐) Ref. 10, rolling ball; (○) Ref. 11, recommended values; (⊕) Ref. 12, torsional vibrating crystal; (●) Ref. 13, falling cylinder. Dev. = 100[($\eta_{\text{present}} - \eta_{\text{lit}}$)/ η_{present}].

Table I. Comparison of Saturated Liquid Densities, ρ of *n*-Butane, *n*-Hexane, and Squalane Obtained in the Present Measurement with Literature Values

T (K)	ρ ($\text{kg} \cdot \text{m}^{-3}$)		Dev. (%) ^a
	Present work	Literature	
<i>n</i> -Butane			
273.15	600.4	600.5 ^b	-0.02
293.15	578.2	578.4 ^b	-0.03
313.15	555.0	554.6 ^b	0.05
333.15	529.5	528.9 ^b	0.11
<i>n</i> -Hexane			
273.15	676.9	676.9 ^c	0
293.15	659.2	659.5 ^c	-0.05
298.15	654.7	654.8 ^d	-0.02
313.15	641.0	641.2 ^c	-0.03
333.15	622.2	622.1 ^c	0.02
Squalane			
298.15	805.0	805.1 ^e	-0.01

^a Dev. = 100[($\rho_{\text{present}} - \rho_{\text{lit}}$)/ ρ_{present}].

^b Ref. 5.

^c Ref. 6.

^d Ref. 7.

^e Ref. 8.

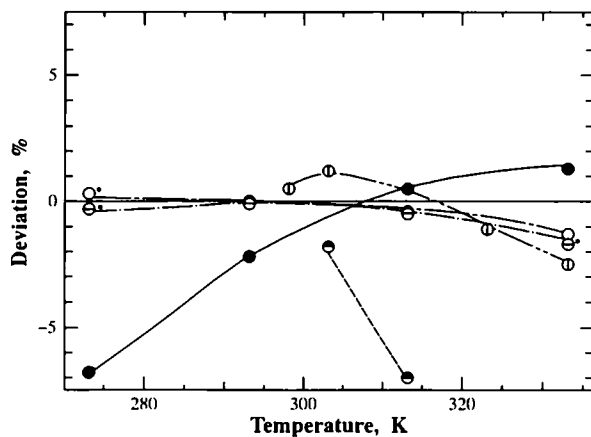


Fig. 2. Deviation of literature viscosity data of *n*-hexane from the present work: (\ominus) Ref. 14, oscillating cylinder; (\circ) Ref. 15, capillary; (\oplus) Ref. 16, torsional vibrating crystal; (\bullet) Ref. 17, capillary; (\bullet) Ref. 13, falling cylinder; (*) extrapolated value. Dev. = $100[(\eta_{\text{present}} - \eta_{\text{lit.}}) / \eta_{\text{present}}]$.

Table II. Density ($\text{kg} \cdot \text{m}^{-3}$) of *n*-Butane and *n*-Hexane + Squalane Mixtures

x_s	T (K)			
	273.15	293.15	313.15	333.15
<i>n</i> -Butane + squalane				
0	600.4	578.2	555.0	529.5
0.118	702.4	685.4	667.7	649.7
0.272	756.5	740.6	725.5	
0.273				710.2
0.517	793.7	779.3		
0.518			765.3	751.5
1	814.4	808.1	795.6	782.8
<i>n</i> -Hexane + squalane				
0	676.9	659.2	641.0	622.2
0.137	735.7	720.1	704.4	690.1
0.321	774.7	760.4	745.9	731.6
0.661	809.9	796.1	782.5	769.1

Table III. Viscosity (mPa · s) of *n*-Butane and *n*-Hexane + Squalane Mixtures

x_s	T (K)			
	273.15	293.15	313.15	333.15
<i>n</i> -Butane + squalane				
0	0.203	0.170	0.143	0.118
0.159	1.46			
0.161		1.06		
0.162			0.797	
0.164				0.643
0.314	5.07			
0.316		3.00		
0.319			2.04	
0.324				1.49
0.551	21.1			
0.555		9.55		
0.559			5.26	
0.569				3.19
0.714	42.5			
0.721		16.5		
0.724			8.16	
0.736				4.84
1	115.4	36.0	15.1	7.85
<i>n</i> -Hexane + squalane				
0	0.382	0.310	0.257	0.218
0.129	1.35	0.972	0.739	0.588
0.268	3.96	2.45	1.63	1.20
0.469	12.5	6.30	3.62	
0.762	49.1	18.3	8.84	4.98

molecular parameters, V_0 and R_η given by the mole-fraction average of the values for each component:

$$\log(\eta_{\text{exp}}^*/R_\eta) = \sum a_i (1/V_r)^i, \quad i = 0-7 \quad (2)$$

$$V_r = V_m/V_0 \quad (3)$$

$$\eta_{\text{exp}}^* = 6.035 \times 10^8 [1/\text{MRT}]^{1/2} \eta V_m^{2/3} \quad (4)$$

where a_i is the coefficient and V_m the molar volume of the liquid mixture. The applicability of Eq. (2) was examined by using the present viscosity

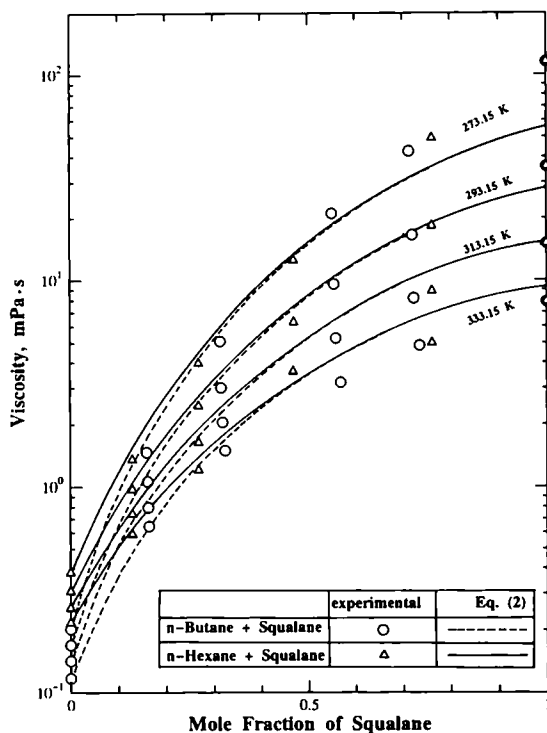


Fig. 3. Comparison of experimental viscosity data with the Dymond equation for mixtures of *n*-butane and *n*-hexane with squalane.

data. The results are shown in Fig. 3, where dashed and solid lines represent the viscosities calculated from Eq. (2) for mixtures of *n*-butane and *n*-hexane with squalane, respectively. Equation (2) deviates considerably from the experimental data in squalane-rich regions, especially at 273 K, with a maximum deviation of 110%. The scheme of Assael et al. fails for mixtures of this type differing greatly in size.

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