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Transport Properties of Nonelectrolyte Liquid Mixtures—I. Viscosity Coefficients for n-Alkane Mixtures at Saturation Pressure from 283 to 378 K

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Viscosity coefficient measurements at saturation pressure are reported for n-hexane + n-hexadecane, n-hexane + n-octane + n-hexadecane, and n-hexane + n-octane + n-dodecane + n-hexadecane at temperatures from 283 to 378 K. The results show that the Congruence Principle applies to the molar excess Gibbs free energy of activation for flow, Δ^*G^E, at temperatures other than 298 K. However, curves of Δ^*G^E versus index number of the mixture are temperature dependent, and this must be taken into account for accurate prediction of mixture viscosity coefficients by this approach. The purely empirical equation of Grunberg and Nissan,

$$\ln\eta = x_1 \ln\eta_1 + x_2 \ln\eta_2 + x_1 x_2 G$$

which has the advantage of not involving molar volumes, satisfactorily reproduces the experimental results for the binary mixture, but G is definitely composition dependent.

KEY WORDS: viscosity; n-alkane mixtures; congruence principle; Grunberg and Nissan equation.

1. INTRODUCTION

The interpretation of transport properties of dense fluids on a molecular basis is proving extremely difficult due to the problem of accounting for many-body interactions. Beyond the pair interaction term, corresponding to a low density gas, it is only the triple collision contribution that has been solved theoretically [1], and then only for the simplest of molecular interactions, the hard-sphere model. For this reason, the computer simulation technique of

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molecular dynamics is becoming more widely used [2–8] for calculating transport properties of dense fluids. However, because of the long computing time involved, most studies relate to diffusion. Studies on the viscosity coefficient have been made for systems of pure liquids in which the pair interaction has a simple form [2, 9, 10], but no simulation results are available for mixtures of fluids with realistic interaction potential energy functions.

It is therefore appropriate to consider other methods for expressing mixture viscosity coefficient data. For this purpose, accurate data are required over as wide a range of experimental conditions as possible. In this paper, viscosity coefficient measurements are reported at saturation pressure for n-hexane + n-hexadecane, n-hexane + n-octane + n-hexadecane, and n-hexane + n-octane + n-dodecane + n-hexadecane from 283 to 378 K. These results are used to test the general applicability of the Principle of Congruence, which has been shown by Coursey and Heric [11] to apply to the molar excess Gibbs free energy of activation for flow in n-alkane mixtures at 298 K. Finally, the results are used as a critical test of the Grunberg and Nissan equation [12], which has recently been shown [13] to be the most effective expression for representing viscosity coefficient data for binary mixtures.

2. EXPERIMENTAL

2.1. The Viscometers

Suspended-level viscometers, specially designed to allow measurements to be made at temperatures above the normal boiling point, are as illustrated in Fig. 1. The side arm and filler section was sealed with an S13 Rotaflo tap R, capable of withstanding internal pressures up to five atmospheres before the seal gave way, and vapor escaped. A and B were reservoir bulbs of 15 cm^3 capacity, connected by the measuring section consisting of capillary C, made from precision bore Veridia tubing, and bulb D, whose volume depended on the other dimensions as shown in Table I. The glass tubing on either side of D had the same diameter to minimize surface tension effects. Marks X and Y etched around the glass above and below bulb D defined a precise volume for timing the passage of the liquid meniscus. The capillary tube opened smoothly in a bell shape into the tubing at each end.

Scrupulous cleanliness of the viscometers was essential for consistent results. The viscometers were cleaned periodically by filling with chromic acid, followed by rinsing with water and then acetone. Drying was ensured by evacuating the viscometers on a vacuum line. All liquids were filtered, before entering the viscometer, through a millipore syringe filter. It was essential

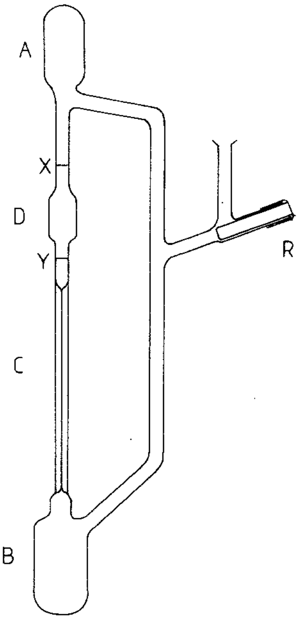

Fig. 1. The suspended-level viscometer.

that all liquids used were of the highest possible purity, and as a matter of routine, n-hexadecane was fractionally crystallized and the other liquids fractionally distilled before use. After filling, the viscometers were cooled significantly to reduce the liquid vapor pressure and the space above the liquid evacuated. In this way, flow time measurements were made under saturated vapor pressure.

Temperature control to better than 0.02 K was maintained by immersion of the viscometers in a Townson and Mercer Bridge Controlled Thermostat Bath E270 Series 3. One bath containing distilled water was used to cover the temperature range from 283 to 368 K, and a second bath containing Shell Risella 33 oil was used for temperatures in the range 333 to 393 K. Temperatures were measured using a Zeal mercury thermometer calibrated

Table I. Viscometer Dimensions

Volume, D (cm^3)	Capillary length (mm)	Capillary bore (mm)	Calibration range ($mm^2\,s^{-1}$)	Number of viscometers
2	115	0.30	0.46–1.82	4
6	55	0.61	1.15–3.97	2
9	55	0.50	0.46–3.97	2
9	75	0.50	0.46–3.97	2

by the National Physical Laboratory, and could be estimated to 0.02 K. Each bath could hold two viscometers, and it was usual to make measurements on a given mixture using two viscometers of different dimensions to obtain a more reliable estimate of the precision of the kinematic viscosity coefficients. The viscometers were mounted on stands that could be rotated freely. They were inverted to fill bulb A and then returned to the original position using a plumb line and two cathetometers at right angles for vertical alignment. The time for the liquid meniscus to pass between marks X and Y was measured to the nearest 0.1 s using a Junghans stopwatch or a Racal Universal Counter 9835.

2.2. Materials

n-Hexane, n-octane, n-dodecane, n-hexadecane, and cyclohexane were purchased from British Drug Houses Ltd., Poole. All had a stated purity of 99+ mol %. Ultra R benzene was purchased from Hopkin and Williams Ltd. Freezing-point determinations on the n-hexadecane indicated its purity was 99.6 mol %. Fractional freezing of the n-hexadecane and fractional distillations of the other hydrocarbons produced fractions whose measured densities and refractive indices are in close agreement with literature values as shown in Table II.

2.3. Calibration

The viscometers were calibrated using benzene and cyclohexane at 298.12 and 333.14 K, and n-dodecane and n-hexadecane at 298.12 and 313.14 K. Values for the kinematic viscosity coefficients are available in API44 [14], but recent measurements on n-hexane [20] cast some doubt on the accuracy of these values. The kinematic viscosity of the calibration liquids was therefore measured in a Master Viscometer, a Technico BS/IP/SL/MV number 2511, with bore size 0.38 mm and length 40 cm, which was calibrated

Table II. Densities and Refractive Indices

	ρ_{298} (kg m^{-3})		n (D, 298.15 K)	
Liquid	Measured	Literature [ref. no.]	Measured	Literature [ref. no.]
n-Hexane	655.0	654.8 [15]	1.37240	1.37229 [15]
n-Octane	698.9	698.67 [16]	1.39507	1.39505 [14]
n-Dodecane	745.5	745.27 [17]	1.41956	1.41949 [14]
n-Hexadecane	770.2	769.96 [18]	1.43244	1.43250 [14]
Cyclohexane	773.8	773.92 [17]	1.42355	1.42350 [19]
Benzene	873.6	873.6 [15]	1.49790	1.49793 [15]

Table III. Kinematic Viscosity Coefficients for the Calibration Liquids

		η/ρ (mm^2 s^{-1})	
Liquid	T (K)	Measured	Literature[a]
Benzene	298.12	0.6906	0.6878
	333.14	0.4662	0.466
Cyclohexane	298.12	1.160	1.157
	333.14	0.7149	0.712
n-Dodecane	298.12	1.824	1.843
	313.14	1.445	1.465
n-Hexadecane	298.11	3.974	4.008
	313.14	2.918	2.953

[a] From Ref. [14].

with freshly distilled water at 298.11 and 333.14 K, and for which the kinematic viscosity coefficients were taken to be 0.8928 and 0.4750 mm^2 s^{-1} [21]. The following general formula was used:

$$\eta/\rho = A(1 - \rho_v/\rho)(1 + \alpha\Delta T)t - B/t \tag{1}$$

where A and B are apparatus constants, t is the flow time, ρ and ρ_v are the densities of liquid and vapor, a is the coefficient of linear thermal expansion of glass, ΔT has been taken as (T − 313.2), and η is the dynamic viscosity coefficient.

This equation takes no account of surface tension effects, which are widely assumed [20–23] to be insignificant in suspended-level viscometers. It should be noted, however, that neglect of surface tension corrections may lead to errors of a few parts of a percent [24].

Values obtained using Eq. (1) for the kinematic viscosity coefficients of the calibration liquids are given in Table III. Agreement with the API44 values [14] is satisfactory for benzene and cyclohexane, but differences between 0.8 and 1.4% occur for n-dodecane and n-hexadecane. A similar discrepancy exists between the results of Eicher and Zwolinski [20] for n-hexane and the AP144 values at temperatures around 313 K. Further support for the present results is given by Heric and Brewer [25], who obtained a value of 3.9723 mm^2 s^{-1} for the kinematic viscosity of n-hexadecane at 298.15 K. The results given in Table III were used for the calibration of the viscometers over the kinematic viscosity ranges listed in Table I. Values of A and B giving the least squares fit to these kinematic viscosities were derived from the measured flow times using Eq. (1). A comparison of kinematic viscosity coefficients calculated using these A and B

values with the results in Table III gave agreement generally well within 0.3%, and the deviations were random. It is concluded that, provided the viscometers are used for liquids whose kinematic viscosity coefficients fall within the limits given, the results obtained should be accurate to within 0.5%. Support for this estimate of the accuracy is given by results obtained for n-hexane, for which careful measurements have recently been made [20]. A comparison of these values with kinematic viscosity coefficients derived from flow time measurements is given in Fig. 2. The agreement is within 0.5%, the combined experimental uncertainties, even at temperatures where the kinematic viscosity coefficient is somewhat lower than the lower limit given in Table I.

2.4. Density Measurement

Densities at 298.2 K were measured using a calibrated Lipkin pyknometer of approximately 5 cm^3 capacity fitted with end caps to prevent

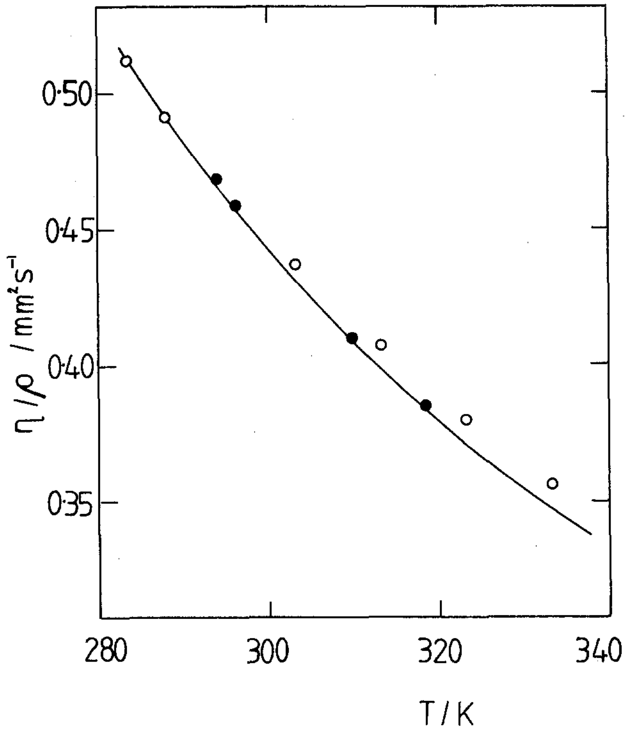

Fig. 2. Kinematic viscosity coefficients for n-hexane; solid line, present work; O, Ref. [14]; ●, Ref. [20].

evaporation losses. Standard corrections [26] were applied for the effects of buoyancy and vapor pressure of the liquid. For densities at other temperatures, a volume change apparatus was used in which the change in volume was measured for a given change in temperature. The apparatus consisted of a 60 cm long capillary tube of precision bore Veridia glass connected to a small reservoir of volume approximately 0.4 cm^3 and a Rotaflo tap at each end. The lower bulb was filled completely with liquid from a syringe and the apparatus mounted on a brass stand with an engraved scale. Meniscus heights in the capillary were recorded after thermal equilibrium was attained at different temperatures up to 363 K. Calibration of the apparatus was made using benzene, for which recent measurements [27] are available up to 490 K with an accuracy of ± 0.1 kg m^{-3}. Corrections were made for glass expansion and liquid evaporation. The results for benzene were reproduced to within 0.1 kg m^{-3} up to 333 K and to within 0.3 kg m^{-3} at higher temperatures.

2.5. Experimental Results

Values of the kinematic viscosity coefficient, density, and dynamic viscosity coefficient are given in Table IV for n-hexane, Table V for n-octane, Table VI for n-dodecane, Table VII for n-hexadecane, Table VIII for mixtures of n-hexane + n-hexadecane, Table IX for ternary mixtures of

Table IV. Viscosity Coefficients of n-Hexane

			η (mN s m^{-2})	
T (K)	ρ (kg m^{-3})	η/ρ (mm^2 s^{-1})	This work	Literature [ref. no.]
283.15	668.6^a	0.5141	0.3437	0.3426 [14]
288.15	664.1^a	0.4911	0.3261	0.3265 [14]
298.19	655.0	0.4504	0.2950	0.2976 [14]
313.23	641.1	0.3994	0.2561	0.2611 [14]
318.26	636.3	0.3843	0.2448	0.2504 [14]
333.36	621.8	0.3450	0.2146	0.2216 [14]
338.36	616.7	0.3348	0.2064	0.2128 [14]
353.29	601.6	0.3028	0.1821	
358.28	596.0	0.2971	0.1771	
373.28	580.4^a	0.2672	0.1550	
374.55				0.1562 [28]
378.29	574.9^a	0.2648	0.1522	
393.2	558.4^a	0.2403	0.1341	
395.25				0.1324 [28]

^{a}Extrapolated.

Table V. Viscosity Coefficients of n-Octane

T (K)	ρ (kg m^{-3})	η/ρ (mm^2 s^{-1})	η (mN s m^{-2})	
			This work	Ref. [14]
283.15	710.9[a]	0.8646	0.6146	0.6184
288.15	706.9[a]	0.8154	0.5764	0.5798
298.19	698.9	0.7255	0.5070	0.5136
313.23	686.7	0.6272	0.4307	0.4355
333.36	670.0	0.5231	0.3505	0.3576
353.29	653.1[a]	0.4461	0.2913	0.3001
373.28	635.7[a]	0.3869	0.2459	0.2547
393.2	617.8[a]	0.3420	0.2113	0.2188

[a] Extrapolated values.

n-hexane + n-octane + n-hexadecane, and in Table X for a mixture of n-hexane + n-octane + n-dodecane + n-hexadecane. The estimated accuracy is ±0.5%, except for n-hexane above 313 K, where it is estimated to be ±1.0%. From a comparison with the values given in API44 [14], it appears that these literature values are too high by up to 3% for these n-alkanes. For the mixtures, corrections were applied for composition changes due to different liquid-vapor equilibria at higher temperatures.

2.6. Principle of Congruence

Although there are reasons for considering that the Eyring activation theory of transport properties [29] does not have a sound physical basis [30],

Table VI. Viscosity Coefficients of n-Dodecane

T (K)	ρ (kg m^{-3})	η/ρ (mm^2 s^{-1})	η (mN s m^{-2})	
			This work	Ref. [14]
283.15	756.4[a]	2.3954	1.812	1.828
288.15	752.8[a]	2.1740	1.637	1.653
298.19	745.5	1.8231	1.359	1.374
313.23	734.6	1.4446	1.061	1.078
333.36	719.9	1.1128	0.8011	0.8123
353.29	705.3	0.8928	0.6297	0.6379
373.28	690.5[a]	0.7379	0.5095	0.5168
393.2	675.8[a]	0.6235	0.4214	0.4278

[a] Extrapolated.

Table VII. Viscosity Coefficients for n-Hexadecane

T (K)	ρ (kg m^{-3})	η/ρ (mm^2 s^{-1})	η (mN s m^{-2})	
			This work	Ref. [14]
298.19	770.2	3.9799	3.065	3.086
313.23	759.8	2.9210	2.219	2.243
318.26	756.3	2.6675	2.017	2.038
333.36	746.0	2.0763	1.549	1.568
338.36	742.5	1.9292	1.432	1.448
353.29	732.3	1.5721	1.151	1.162
358.28	728.9	1.4769	1.076	1.087
373.28	718.5	1.2396	0.8906	0.8992
378.29	715.1^a	1.1752	0.8404	0.8479
393.2	704.7^a	1.0159	0.7159	0.7181

^{a}Extrapolated.

it has proved useful for the correlation and prediction of viscosity coefficients for n-alkane mixtures [11]. If the ideal viscosity of a mixture is defined by

$$\Delta^* G^{\text{ideal}} = \sum_i x_i \Delta^* G_i \tag{2}$$

then a molar excess Gibbs free energy of activation for flow $\Delta^* G^E$ given by

$$\Delta^* G^E = \Delta^* G - \Delta^* G^{\text{ideal}} \tag{3}$$

can be derived from experimental viscosity coefficient data according to

$$\Delta^* G^E = RT\{\ln(\eta V) - \sum_i x_i \ln(\eta_i V_i)\} \tag{4}$$

Coursey and Heric [11] have shown that the Principle of Congruence, originally proposed by Bronsted and Koefoed [31] for the molar excess Gibbs free energy of mixing, applies also to the molar excess Gibbs free energy of activation for flow at 298.2 K. On the basis of this, they made accurate predictions of viscosity coefficients for n-alkane mixtures at this temperature, knowing the viscosity coefficients of the pure components.

The present accurate viscosity coefficient data allow the range of validity of this approach to be tested. Values of $\Delta^* G^E$ calculated from Eq. (4) for n-hexane + n-hexadecane mixtures at different temperatures are plotted in

Table VIII. Viscosity Coefficients for Mixtures of n-Hexane (1) + n-Hexadecane (2)

T (K)	x (1)	ρ (kg m^{-3})	η (mN s m^{-2})	x (1)	ρ (kg m^{-3})	η (mN s m^{-2})
283.15		Frozen		0.3784	756.8	2.072
288.15	0.1996	766.8	2.784	0.3784	754.5	1.948
298.19	0.1987	759.5	2.235	0.3772	747.1	1.607
318.26	0.1976	745.2	1.534	0.3758	732.3	1.155
338.36	0.1958	731.0	1.128	0.3734	717.6	0.8753
358.28	0.1929	716.8	0.8711	0.3695	702.9	0.6932
378.29	0.1885	702.4	0.6938	0.3637	688.1	0.5623
283.15	0.5993	738.0	1.237	0.7988	709.7	0.7014
288.15	0.5993	734.1	1.172	0.7988	707.2	0.6740
298.19	0.5983	726.5	1.001	0.7982	699.1	0.5933
318.26	0.5971	711.0	0.7591	0.7975	682.5	0.4712
338.36	0.5949	695.5	0.5993	0.7963	665.7	0.3845
358.28	0.5916	679.8	0.4880	0.7944	648.6	0.3203
378.29	0.5864	664.2	0.4058	0.7915	631.3	0.2710

Table IX. Viscosity Coefficients of Mixtures of n-Hexane (1) + n-Octane (2) + n-Hexadecane (3)

T (K)	x (1)	x (2)	ρ (kg m^{-3})	η (mN s m^{-2})
288.15	0.1097	0.1152	766.6	2.733
298.15	0.1093	0.1152	759.5	2.194
318.26	0.1086	0.1151	745.2	1.512
338.36	0.1076	0.1151	730.9	1.114
358.28	0.1058	0.1149	716.7	0.8598
378.29	0.1032	0.1146	702.5	0.6870
298.19	0.3286	0.3297	727.4	0.9767
318.26	0.3276	0.3300	712.0	0.7399
338.26	0.3259	0.3303	696.4	0.5836
358.28	0.3229	0.3317	680.4	0.4746

Table X. Viscosity Coefficients for Mixtures of n-Hexane (1) + n-Octane (2) + n-Dodecane (3) + n-Hexadecane (4)

T (K)	x (1)	x (2)	x (3)	ρ (kg m^{-3})	η (mN s m^{-2})
288.15	0.2596	0.2996	0.2506	734.1	1.084
298.19	0.2593	0.2997	0.2507	726.5	0.9261
318.26	0.2584	0.2997	0.2510	711.1	0.7041
338.36	0.2569	0.3004	0.2517	695.6	0.5556
358.28	0.2545	0.3009	0.2529	679.4	0.4523

Fig. 3 against the mixture average carbon chain length n given by

$$n = \sum_i x_i C_i \tag{5}$$

where C_i is the number of carbon atoms in the n-alkane chain of component i. For the three-component and four-component n-alkane mixtures, $\Delta^* G^E$ values from Eq. (4) are used to test the Principle of Congruence by plotting values of $h_{616:n}$ against n in Fig. 3, where

$$h_{616:n} = \Delta^* G^E + \sum_i x_i h_{616:i} \tag{6}$$

$h_{616:i}$ is the height of the curve above the x axis, where $n = C_i$, and x_i is the mole fraction of n-alkane with carbon chain C_i.

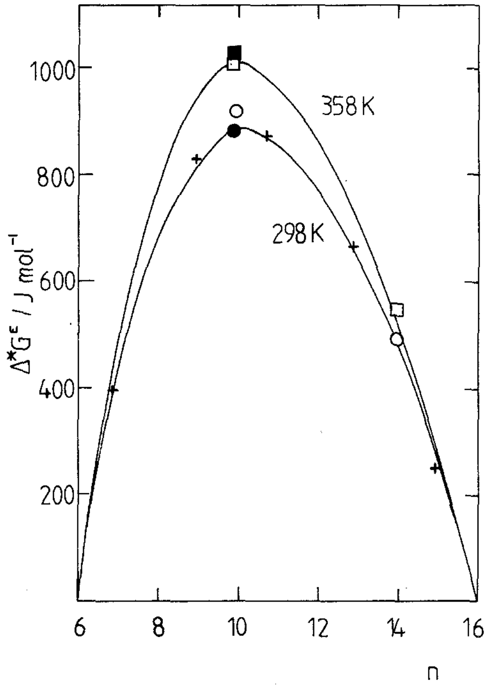

Fig. 3. Temperature dependence of $\Delta^* G^E$ versus index number of the mixture. Lines drawn through the present results for n-hexane + n-hexadecane; +, 298 K, Ref. [11]. Ternary mixture; ○, 298 K; □, 358 K. Quaternary mixture: ●, 298 K; ■, 358 K.

The curves show that the Principle of Congruence is obeyed not only at 298.2 K but also at the higher temperatures. However, Δ^*G^E is definitely temperature dependent, increasing as the temperature is raised, for a given mixture. This is important for the calculation of viscosity coefficients of n-alkane mixtures at saturation pressure at temperatures above 298 K. If the 298 K reference curve is used to calculate Δ^*G^E for the mixture, the predicted values will be low; for example, by 4% at 358 K for the three-component mixture with $x_1 = 0.3229$, $x_2 = 0.3317$.

2.7. The Grunberg and Nissan Equation

The Grunberg and Nissan equation [12] has been recommended by Irving [13] as being the most effective equation in representing viscosity coefficient data for binary mixtures. This empirical expression may be written as

$$\ln\eta = x_1 \ln\eta_1 + x_2 \ln\eta_2 + x_1 x_2 G \tag{7}$$

where η is the viscosity coefficient of the mixture, and η_i is the coefficient of component i. G is the Grunberg and Nissan constant, which Irving recommended should be treated as a single disposable parameter.

The present accurate data for n-hexane + n-hexadecane allow the composition and temperature dependence of G to be determined. The results are presented in Table XI. For this system, G is practically temperature independent, but varies with change in composition, decreasing with increasing mole fraction of n-hexadecane. Isdale [32] found that the equation

$$G_x = G_{0.5}\,(1.343 - 0.685x) \tag{8}$$

satisfactorily represented the mole fraction dependence of G for the data then

Table XI. G Values for n-Hexane (1) + n-Hexadecane (2)

	x_1			
T (K)	0.199	0.377	0.598	0.798
298.19	0.93	1.01	1.17	1.40
318.26	0.90	1.00	1.17	1.41
338.36	0.89	0.99	1.17	1.40
358.28	0.88	0.97	1.14	1.35
378.29	0.85	0.95	1.12	1.32

available for binary n-alkane mixtures. The present viscosity coefficient data from 283 to 378 K agree to within ±1.5% with values calculated from Eqs. (7) and (8) when $G_{0.5}$ is taken as 1.08.

3. CONCLUSIONS

Viscosity coefficient measurements at saturation pressure are reported for n-hexane + n-hexadecane, n-hexane + n-octane + n-hexadecane and n-hexane + n-octane + n-dodecane + n-hexadecane from 283 to 378 K with an estimated accuracy of 0.5%. The results show that the Principle of Congruence applies to the molar excess Gibbs free energy of activation for flow at temperatures other than 298 K. For accurate prediction of mixture viscosity coefficients by this method, the temperature variation of Δ^*G^E must be taken into account. For the binary mixture, the Grunberg and Nissan equation satisfactorily represents the data at all temperatures, with the composition dependence of G given by

$$G = 1.45 - 0.74x$$

where x is the mole fraction of n-hexadecane.

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