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Viscosities of Nonelectrolyte Liquid Mixtures. II. Binary and Quaternary Systems of Some *n*-Alkanes

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This paper is the second in a series of viscosity and density studies on multicomponent mixtures of *n*-alkanes from 303 to 338 K. Reported here are the results of binary mixtures of *n*-tetracosane + *n*-octane as well as quaternary mixtures of *n*-tetracosane + *n*-octane + *n*-decane + *n*-hexane at 318.16, 328.16, and 338.16 K. Viscosities were determined using a standard U-tube Ostwald viscometer, and densities were determined using a flask-type pycnometer. Empirical relations tested include the Grunberg and Nissan equation and the method of corresponding states. In addition, comparisons were made regarding the behavior of this quaternary system and homologous binary mixtures of *n*-hexadecane + *n*-octane and *n*-tetracosane + *n*-octane at the same temperatures.

KEY WORDS: alkane; corresponding states; density; Grunberg and Nissan equation; liquid; mixtures; viscosity.

1. INTRODUCTION

This paper is the second in a series on the density and viscosity of alkane mixtures. The primary goal of these studies was to gain a clearer understanding of the effect of temperature and composition on the viscosities of liquid mixtures. A series of *n*-alkanes ranging from *n*-hexane to *n*-tetracosane was selected and the temperature range 303.16 to 338.16 K was selected based on the lack of experimental data for pure and mixture viscosities and densities for *n*-alkanes at these temperatures.

The first study in this series focused on the binary mixture *n*-hexadecane + *n*-octane at 318.16, 328.16, and 338.16 K [1]. To understand better the composition effects on mixture viscosity, the present studies

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focused on binary systems of *n*-tetracosane + *n*-octane prepared as a "pseudoternary" system using equimolar [*n*-tetracosane + *n*-octane] and *n*-octane to simulate essentially the composition of the previous binary mixture *n*-hexadecane + *n*-octane [1]. Also, viscosities and densities of quaternary mixtures composed of *n*-tetracosane + *n*-octane + *n*-decane + *n*-hexane were measured at 318.16, 328.16, and 338.16 K. The quaternary mixtures were prepared to simulate the same average carbon chain length as the previous binary mixtures. As the successful extension of binary mixture viscosity equations to multicomponent systems has not been made, it was considered useful to test the relations most frequently used for binary systems with the quaternary viscosity data.

2. EXPERIMENTAL

Samples of *n*-octane and *n*-tetracosane were obtained from Alfa Chemical Company, Danvers, Mass. These *n*-alkanes ranged from 99 to 99.9+ mol% purity. Gas–liquid chromatography using a flame ionization detector with capillary column indicated purities considerably better than label values, and hence they were used without further purification. The methods and techniques used to determine viscosities and densities have been described in detail previously [1]. Viscosities of the pure liquids were measured and compared with accepted literature values where available [2].

Treating an equimolar $C_{24} + C_8$ combination as a single component, new mixtures with *n*-octane as the "second" component were prepared in the same mole fractions as the previous binary *n*-hexadecane + *n*-octane mixtures [$x_{16} = 0.15$, 0.35, 0.50, 0.65, and 0.80, approximately]. As with the systems using equimolar amounts of *n*-tetracosane + *n*-octane to simulate *n*-hexadecane behavior, in this study *n*-octane behavior was simulated using equimolar amounts of *n*-decane + *n*-hexane. Thus, a profile of composition effects of the heavier *n*-alkanes on viscosity behavior could be more clearly developed.

3. RESULTS

Table I lists the experimental shear viscosities and densities for pure *n*-tetracosane, pure *n*-octane, and the mixtures of *n*-tetracosane + *n*-octane at 318.16, 328.16, and 338.16 K. Table II contains the experimental viscosities and densities for pure *n*-tetracosane, *n*-octane, *n*-decane, and *n*-hexane as well as for the quaternary mixtures of these alkanes. The accuracy of the viscosities is estimated to be between ±0.3 and 0.5%; densities are estimated as accurate to within $\pm 0.0001\ \mathrm{g \cdot cm^{-3}}$.

3.1. Arithmetic Average of Viscosities

A commonly used empirical relation relating pure component viscosities and mole fractions is the arithmetic average, which is frequently expressed as a deviation function,

$$\Delta\eta = \eta_M - \sum_{i=1}^{c} x_i \eta_i \tag{1}$$

where η_M is the mixture viscosity, x_i is the mole fraction of component i, η_i is the viscosity of pure component i, and c is the total number of components in the mixture. Deviations of mixture viscosity from the arithmetic average of pure components are given in Table I. The plot of $\Delta\eta$ vs $\bar{n}$ for $C_{24} + C_8$ is seen in Fig. 1, where $\bar{n}$ is the average carbon chain length, defined as

$$\bar{n} = \sum_{i=1}^{c} x_i n_i \tag{2}$$

where n_i is the number of carbon atoms in component i, and all other terms have been defined previously.

At lower $\bar{n}$ values, where *n*-octane is the predominant component in the system, somewhat regular smooth deviations are noted at all temperatures. As the mixture reaches an equimolar composition ($\bar{n} = 14$), the deviations fall above the smooth curves indicated by the previous data points at 318.16, 328.16, and 338.16 K. Compared to the very smooth viscosity–composition relations seen in the *n*-hexadecane + *n*-octane binary mixtures [1], the behavior is essentially the same for low concentrations of C_{24}, although the numerical values of the deviations are approximately four times larger. Therefore, until an equimolar concentration of *n*-tetracosane is reached, it can be concluded that the arithmetic deviations of *n*-hexadecane + *n*-octane [1] and the simulated *n*-hexadecane [$0.5\,C_{24} + 0.5\,C_8$] + *n*-octane are essentially the same.

The arithmetic deviation in the quaternary system of *n*-tetracosane + *n*-octane + *n*-decane + *n*-hexane exhibited major deviations from the arithmetic average at 328.16 K and showed a maximum and indicated a minimum. In contrast, the 318.16 and 338.16 K values yielded fairly reasonable smooth curves through the mixture data. Values for arithmetic deviations with temperature and composition are shown in Table II. Again, no measurements were taken for *n*-tetracosane at 318.16 K because it was in its solid state; thus, a value for the viscosity of the pure *n*-tetracosane was approximated at this temperature using an equation which approximates viscosities of pure *n*-alkanes and is based on group contribution for carbon and hydrogen number.

Table I. Results of Testing Empirical

	Mole fraction				
	n-Tetracosane, x_{24}	*n*-Octane, x_8	Shear viscosity, $\eta_M/(10^{-3}\ \mathrm{Pa \cdot s})$	Mixture density, $\rho_M/(\mathrm{g \cdot cm^{-3}})$	Average carbon chain length, $\bar{n}$
$T = 318.16$ K	0.0000	1.0000	0.4138	0.68277	8.0000
	0.0781	0.9219	0.63238	0.70280	9.24960
	0.1587	0.8413	0.97438	0.72253	10.53920
	0.2389	0.7611	1.28134	0.73510	11.82240
	0.3125	0.6875	1.62965	0.74408	13.00000
	0.3991	0.6009	1.92078	0.74740	14.38560
	1.0000	0.0000	5.729*	0.782*	16.00000
$T = 328.16$ K	0.0000	1.0000	0.3706	0.67448	8.0000
	0.0781	0.9219	0.56221	0.69586	9.24960
	0.1587	0.8413	0.85127	0.71503	10.53920
	0.2389	0.7611	1.11235	0.72796	11.82240
	0.3125	0.6875	1.39914	0.73794	13.00000
	0.3991	0.6009	1.62734	0.74034	14.38560
	1.0000	0.0000	4.78746	0.776*	16.00000
$T = 338.16$ K	0.0000	1.0000	0.3415*	0.66607	8.0000
	0.0781	0.9219	0.50534	0.68707	9.24960
	0.1587	0.8413	0.74978	0.70767	10.53920
	0.2389	0.7611	0.97735	0.72051	11.82240
	0.3125	0.6875	1.21824	0.73068	13.00000
	0.3991	0.6009	1.39716	0.73370	14.38560
	1.0000	0.0000	3.84926	0.770*	16.00000

* Denotes estimated value.

Equations for Systems Containing $C_{24} + C_8$

Arithmetic deviation, $(-)\Delta\eta/(10^{-3}\,\mathrm{Pa\cdot s})$	Grunberg & Nissan parameter, G	Gibbs energy of activation, $\Delta^{\ddagger}G^{E}/(\mathrm{J\cdot mol^{-1}})$	Trapp predicted viscosity, $\eta_M/(10^{-3}\,\mathrm{Pa\cdot s})$	Percentage deviation viscosity
0		0	0.4297	−3.8424
0.19654	3.03981	683.582	0.6221	+1.6256
0.28294	3.29080	1331.135	0.8551	12.2416
0.40226	2.76346	1550.712	1.1267	12.0686
0.44515	2.55774	1706.163	1.4174	13.0243
0.61432	2.02779	1574.427	1.8213	5.1791
0		0		
0		0	0.3873	−4.5062
0.15335	3.01280	694.470	0.5568	+0.9623
0.22029	3.18731	1332.368	0.7588	10.8626
0.31344	2.68305	1555.805	0.9901	10.9902
0.35173	2.46187	1696.719	1.2333	11.8530
0.50603	1.91156	1545.341	1.5651	3.8246
0		0	4.9994	−4.4270
0		0	0.3506	−2.2665
0.11012	2.81530	677.531	0.5011	+0.8390
0.14840	2.21098	1303.484	0.6779	9.5868
0.20215	2.60035	1560.699	0.8769	10.2778
0.21944	2.39639	1707.050	1.0828	11.1177
0.34429	1.84353	1543.842	1.3588	2.7456
0		0	3.9220	−1.8897

Table II. Mixture Viscosity Results

	Mole fraction				Shear viscosity, η_M (10^{-3} Pa · s)
	n-Hexane, x_6	*n*-Octane, x_8	*n*-Decane, x_{10}	*n*-Tetracosane, x_{24}	
$T = 318.16$ K	0.0000	0.0000	0.0000	1.0000	5.729*
	0.1000	0.4001	0.1000	0.3999	Solid
	0.1752	0.3250	0.1750	0.3248	2.11359*
	0.2507	0.2468	0.2508	0.2517	1.31481
	0.2960	0.2019	0.2961	0.2060	1.01673
	0.3997	0.1005	0.3997	0.1001	0.72837
	1.0000	0.0000	0.0000	0.0000	0.2504*
	0.0000	1.0000	0.0000	0.0000	0.4138
	0.0000	0.0000	1.0000	0.0000	0.6573
$T = 328.16$ K	0.0000	0.0000	0.0000	1.0000	4.78746
	0.1000	0.4001	0.1000	0.3999	2.89889
	0.1752	0.3250	0.1750	0.3248	1.91494
	0.2507	0.2468	0.2508	0.2517	1.14425
	0.2960	0.2019	0.2961	0.2060	0.90407
	0.3997	0.1005	0.3997	0.1001	0.65520
	1.0000	0.0000	0.0000	0.0000	0.2307*
	0.0000	1.0000	0.0000	0.0000	0.3706
	0.0000	0.0000	1.0000	0.0000	0.5823
$T = 338.16$ K	0.0000	0.0000	0.0000	1.0000	3.84926
	0.1000	0.4001	0.1000	0.3999	2.43824
	0.1752	0.3250	0.1750	0.3248	1.66483
	0.2507	0.2468	0.2508	0.2517	0.97919
	0.2960	0.2019	0.2961	0.2060	0.81305
	0.3997	0.1005	0.3997	0.1001	0.58740
	1.0000	0.0000	0.0000	0.0000	0.2128*
	0.0000	1.0000	0.0000	0.0000	0.3415*
	0.0000	0.0000	1.0000	0.0000	0.5220

* Denotes estimated value.

for the Quaternary Systems $C_{24} + C_8 + C_{10} + C_6$

Mixture density, $\rho_M/(\mathrm{g \cdot cm^{-3}})$	Average carbon chain length, $\bar{n}$	Deviations from arithmetic averages, $\Delta\eta/(10^{-3}\ \mathrm{Pa \cdot s})$	Gibbs energy of activation, $\Delta^{\ddagger}G^{E}/(\mathrm{J \cdot mol^{-1}})$	TRAPP predicted shear viscosity $\eta_M/(10^{-3}\ \mathrm{Pa \cdot s})$	Percentage deviation TRAPP vs shear (actual)
0.782*	24.00000		5489.300	1.4753	30.1993
	14.39840	−1.03708	1591.817	1.1812	10.1619
0.70783*	13.19640	−0.80607	1219.020	1.0182	−0.1446
0.73466	12.02740	−0.26769	994.859	0.6921	4.9796
0.72640	11.29620			0.2520	−0.6390
0.71018	9.60160			0.4297	−3.8424
0.6362*	6.00000			0.6695	−1.8561
0.6828	8.00000				
0.7112*	10.00000				
0.776*	24.00000	+0.75481	3047.122	4.9994	−4.4270
0.76315	14.39840	+0.09721	2460.213	1.5712	45.7999
0.74921	13.19640	−0.35610	1594.220	1.2817	33.0684
0.72766	12.02740	−0.39768	1258.822	1.0366	9.4079
0.71896	11.29620	−0.18623	1018.875	0.8988	0.5829
0.70644	9.60160			0.6184	5.6166
0.6268*	6.00000			0.2297	0.4335
0.6745	8.00000			0.3873	−4.5062
0.7035*	10.00000			0.5949	−2.1638
0.770*	24.00000	−0.35227	3029.438	3.9220	−1.8897
0.75659	14.39840	−0.27162	2496.966	—	—
0.74219	13.19640	−0.26006	1420.995	1.1238	32.4926
0.72060	12.02740	−0.32967	1321.469	0.9170	6.3512
0.71139	11.29620	−0.20799	1052.547	0.7992	1.7035
0.69467	9.60160			0.5558	5.3796
0.6172*	6.00000			0.2099	1.3628
0.66607	8.00000			0.3506	−2.6647
0.6957*	10.00000			0.5321	−1.9349

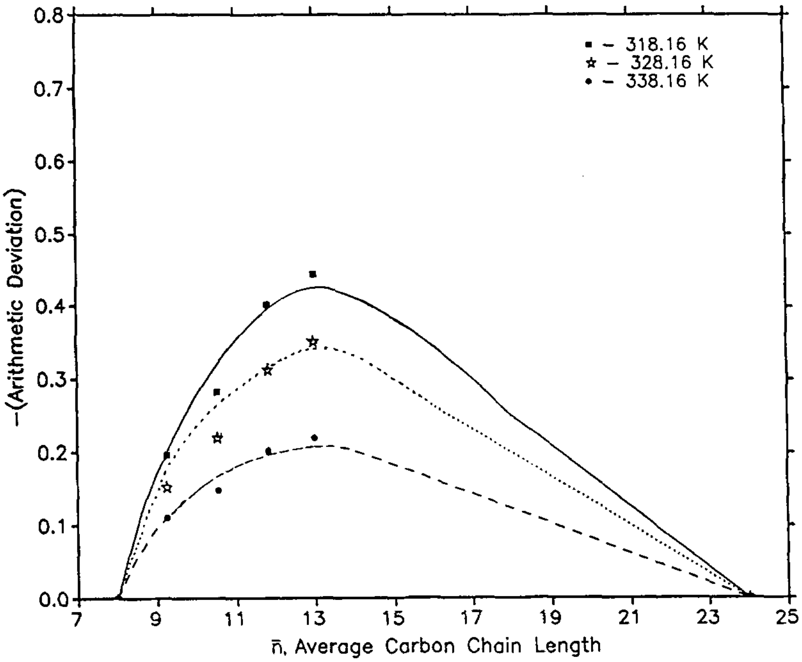

Fig. 1. Arithmetic deviation: *n*-tetracosane + *n*-octane.

In comparison to the deviations from the arithmetic average in the binary mixture $C_{16} + C_8$ [1] and the binary mixture $C_{24} + C_8$, the presence of *n*-tetracosane vs *n*-hexadecane in the simple binary mixtures resulted in negative deviations almost four times as large for the C_{24} mixture as the C_{16} system. In contrast, the quaternary systems of this study fluctuate between positive and negative deviations at all temperatures. Also, at 338.16 K, the deviations are not as smooth as those at 318.16 K, as the data points fall above and below those at 328.16 K. It is evident that the temperature dependence of mixture viscosity is less significant at all concentrations for the 318.16 K measurements. Temperature is slightly more significant at lower carbon chain length for the 338.16 K systems, possibly due to some evaporation of *n*-hexane, which could produce unstable behavior. However, all precautions were taken to reduce any possibilities of evaporation. Temperature dependence appears to predominate over the composition dependence of mixture viscosities at 328.16 K.

3.2. Grunberg and Nissan Equation

Dymond and Young [3] have tested the Grunberg and Nissan equation

$$G=\left[\ln \eta_M-\sum_{i=1}^{k} x_1 \ln \eta_i\right] \Big/ (x_1 x_2) \tag{3}$$

extensively on binary systems of *n*-alkanes. In Eq. (3), η_M is the mixture viscosity, x_i is the mole fraction of component i, η_i is the pure component viscosity, and G is defined as the Grunberg and Nissan parameter for the binary ($k=2$) mixture.

Table I lists the calculated values of the Grunberg and Nissan parameter, G, for the binary mixture *n*-tetracosane + *n*-octane at 318.16, 328.16, and 338.16 K. Variation of this parameter with average mixture carbon chain length of the mixture, $\bar{n}$, is presented in Fig. 2. Results of previous testing of binary mixtures by Dymond and Young [3] of *n*-hexane + *n*-hexadecane over the temperature range 298.19 to 378.29 K showed a straight-line plot for G against x.

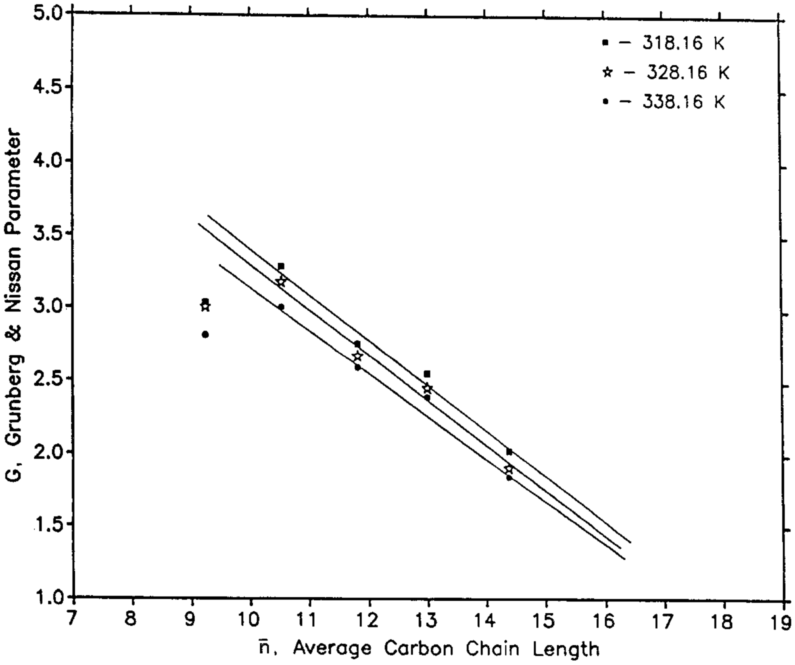

Fig. 2. Grunberg and Nissan equation: *n*-tetracosane + *n*-octane.

In comparison, for the binary *n*-tetracosane + *n*-octane mixtures in this study, it can be concluded that reasonable straight-line agreement is exhibited by the mixtures at all experimental temperatures, 318.16, 328.16, and 338.16 K. At the lowest concentrations of *n*-tetracosane, i.e., lower $\bar{n}$, average carbon chain length, some deviations from the straight-line behavior were noted.

The most significant deviation occurs for the mixture in which $x_{24} = 0.0781$ [$\bar{n} = 9.25$]; possible discrepancies could be due to general evaporation of some of the *n*-octane in the system, although the data points at all temperatures are characteristically about the same interval apart as those which follow straight-line behavior. The second point for which deviation from straight-line behavior occurs is for $x_{24} = 0.1587$ and $x_8 = 0.8413$ [$\bar{n} = 10.54$]. Here, the 318.16 and 328.16 K values appear to follow the Grunberg and Nissan equation, although major deviation is noted at the highest temperature, 338.16 K. It is possible that the deviation was partially caused by evaporation of *n*-octane, although precautions were taken during experiments to minimize any evaporation effects.

Similar deviation for lower average carbon chain length, $\bar{n}$, was noted for the system *n*-hexadecane + *n*-octane [1] over the same temperature range. In this mixture [$\bar{n} = 9.20$; $x_{16} = 0.1585$] the lowest hexadecane-containing mixture showed the 318.16 and 328.16 K Grunberg and Nissan parameters to fall above the straight line exhibited by the other points, while the 338.16 K point fell below the line. Although evaporation of *n*-octane could be partially responsible for some of the deviation, it is not clear to what extent composition/temperature competing effects also affected the viscosity behavior.

3.3. Principle of Congruence

The principle of congruence has been used successfully to predict mixture viscosities for many simple alkane systems [4–8]. Results for mixtures including *n*-tetracosane and *n*-octane supported congruence to a limited extent; however, when extended to quaternary systems of *n*-tetracosane + *n*-octane + *n*-decane + hexane, congruence was not followed to any reasonable extent.

3.4. Method of Corresponding States

Ely and Hanley [9] developed an empirical model for transport properties of hydrocarbons using as input data, critical pressure, critical temperature, molecular weight, fluid normal boiling point, and the acentric factor. As discussed by Baltatu [10], the premise for the model is the assumption that "the configurational properties of a mixture can be equated to those of a hypothetical pure fluid, which in turn are evaluated via the corresponding states principle with respect to a given reference fluid."

At present, the reference fluid used in this model is methane. Originally, the model was intended for use only for fluids of low hydrocarbon number, i.e., C_6 or C_7. Corrections and improvements have been made in the last 3 years to the original model to correct for the "noncorresponding behavior" and size difference effects of the higher-number hydrocarbon components.

While some methods used to correct the model have not been successful in accounting for the true nature of the intermolecular interactions between component molecules, as determined by the size, shape, and force field exerted by the individual component molecules, the technique has seen some success in predicting mixture viscosities. To date, the composition effect on viscosity has not been rigorously accounted for by the Hanley model or any other model.

Table I shows the percentage deviations between experimental data for the binary system of *n*-tetracosane + *n*-octane and those viscosities calculated by the TRAPP program at Fluor Engineering Company, Irvine, California, by Baltatu.

Agreement between 1 and 13% is noted for the corresponding-states method at 318.16 K. At 328.16 K agreement ranged from 0.96 to 11%; and at 338.16 K agreement ranged from 0.84 to 11%. Errors in determining the pure liquid and mixture viscosities with the method of corresponding states were determined as

$$\text{percentage error} = [\text{expt.} - \text{calc.}]/[\text{expt.}] \times 100\% \qquad (4)$$

The predicted values showed random deviation above and below the experimental data; positive deviations were noted for mixture viscosities, while predicted pure component viscosities were consistently higher than those observed experimentally. In general, for systems lower in tetracosane composition, the closest agreement between experimental and predicted viscosities was noted.

For purposes of comparison, Table II shows the deviations in pure and mixture viscosities for the quaternary mixtures. In the table, the composition dependence on viscosity can be seen in terms of the empirical corresponding-states predictive technique. The largest deviations from experimental values, in general, were for mixtures where the heaviest *n*-alkane was slightly predominant, although less significant deviations occurred in predicting pure-component viscosities of *n*-hexadecane + *n*-tetracosane. Deviations were smaller for $x_{24} = 0.3991$ than x_{24} in the

$C_{24}+C_8$ series, and in the $C_{24}+C_8+C_{10}+C_6$ series deviations were smaller for $x_{24}=0.3248$ than $x_{24}=0.3999$.

Thus, the TRAPP program based on corresponding-states principles and accepted thermodynamic data as input parameters as tested for the binary system of *n*-tetracosane + *n*-octane predicted mixture viscosities at 318.16, 328.16, and 338.16 K within 0.8–13% of the experimental values. While this range is not extremely accurate, it is commendable that a technique based on methane as a reference fluid and pure-fluid critical parameters can predict viscosities for C_{24} to within 5%. This model has shown particularly good success for hydrocarbon mixtures, and as significant corrections are anticipated in the future for the corresponding-states model, it is expected that greater accuracy in predictions will be achieved.

4. CONCLUSIONS

Viscosities and densities were measured for the binary system *n*-tetracosane + *n*-octane at 318.16, 328.16, and 338.16 K. Viscosities were strongly dependent on the mixture component *n*-tetracosane for all temperatures.

The arithmetic average, commonly used in some form to predict mixture viscosities, did not show smooth deviations at higher *n*-tetracosane mixture compositions. Although the binary system was prepared as equimolar $[C_{24}+C_8]+C_8$, the mixture did not show the same viscosity behavior as the previously measured *n*-hexadecane + *n*-octane binary mixtures from 318.16 to 338.16 K. In the quaternay *n*-tetracosane + *n*-octane + *n*-decane + *n*-hexane systems, a fairly smooth curve was seen in plots of $-\Delta\eta_i$ vs $\bar{n}$ at 318.16 and 338.16 K, while the 328.16 K curve showed maxima and indicated a minimum.

The Grunberg and Nissan equation, which had been shown to be successful by other researchers for similar binary mixtures of lower *n*-alkanes at various temperatures, was shown to be semiuseful for the *n*-tetracosane + *n*-octane mixtures in this study where *n*-tetracosane compositions were dominant. Some error is possibly due to the evaporation in higher compositions of *n*-octane. It is expected that there would be more intermolecular interactions in mixtures of *n*-tetracosane, causing the Grunberg and Nissan equation to be considered useful for binary systems of lower molecular weight *n*-alkanes over limited temperature ranges.

The principle of congruence, which has been used successfully for other systems of *n*-alkanes, was not seen to hold for the binary and quaternary mixtures including *n*-tetracosane as a component.

Program TRAPP, based on the method of corresponding states, predicted pure and mixture viscosities within 0.8 to 13% of experimental values from 318.16 to 338.16 K. Deviations for the quaternary system ranged from 1 to 45% throughout all temperatures; however, the failure of this empirical technique to predict mixture viscosities where *n*-tetracosane was a major component is expected, as the methane reference fluid of the model is far different in intermolecular size, shape, and force field from *n*-tetracosane. Although accuracies to within 2% are desirable for empirical methods, the corresponding-states method is meritable as it requires only pure liquid critical parameters as input. Further, the current model design features the use of methane to predict viscosities of more complex mixtures. As improvements are made, i.e., reference fluid updated to propane, better agreement between predicted and experiental values is anticipated.

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