

# Densities and Viscosities of the Regular Quinary System: Toluene (1) + Octane (2) + Ethylbenzene (3) + Tetradecane (4) + Hexadecane (5) and Its Quaternary Subsystems at (308.15 and 313.15) K

Abdulghanni H. Nhaesi and Abdul-Fattah A. Asfour\*

Environmental Engineering, University of Windsor, Windsor, Ontario, Canada N9B 3P4

The densities and kinematic viscosities of the quaternary subsystems of the regular quinary system toluene (1) + octane (2) + ethylbenzene (3) + tetradecane (4) + hexadecane (5) were measured over the entire composition range at 308.15 K and 313.15 K. The viscosity deviations from the mole fraction-averaged viscosity of the mixture were calculated from the absolute viscosity data. All of the viscosity deviations are negative. The viscosity data reported herein were used to test the predictive capabilities of the predictive version of the McAllister model, which was reported by Nhaesi and Asfour and the Grunberg–Nissan equation. The obtained results showed that the McAllister model predicts the viscosity data better than the Grunberg–Nissan equation.

## Introduction

The densities and viscosities of liquid mixtures are important from a practical standpoint because these properties are required for design calculations. In addition, they are also of great interest from a theoretical viewpoint because they may lead to a better understanding of liquid mixture structure. Unfortunately, viscosity data on multicomponent liquid mixtures at different temperatures are very scarce in the literature.

In the present work, the viscosities and densities of a quinary regular system and all of its corresponding quaternary subsystems at 308.15 K and 313.15 K are reported. The quinary system consists of toluene, octane, ethylbenzene, tetradecane, and hexadecane. The pure components of the selected system are of the nonassociating type with significant differences in shape and structure. To the best of our knowledge, similar data have not been reported in the literature.

The objective of the present study is to report the viscosity and density data of the above-named quinary system and all of its quaternary subsystems at 308.15 K and 313.15 K. In addition, the data were used to test some literature viscosity models.

## Experimental Section

**Materials.** The components used in preparing the systems investigated in the present study and the chemicals used for density meter calibration were purchased from Aldrich Chemical Co. The liquids were used as received without any further purification. Their purity was stated by the manufacturer to be higher than 99+%. A chromatographic test of the chemical purity, using a 5 m × 0.53 mm methyl silicone capillary column and a flame ionization detector (FID), confirmed that the purities of the chemicals were better than their stated values (cf. Table 1).

**Preparation of Solutions.** Solutions were prepared gravimetrically by weighing the components of the mixtures in vials fitted with Teflon disks and aluminum seals

**Table 1. Results of Chromatographic Analysis of Reagent Purity**

compound	specification, mol %	GC analysis, mass%
toluene	99.8	99.9
octane	99+	99.5
ethylbenzene	99.8	99.9
tetradecane	99	99.8
hexadecane	99+	99.2

to minimize evaporation losses. A Mettler HK 160 balance with a stated precision of  $\pm 2 \times 10^{-7}$  kg was employed.

**Density Measurements.** Densities were measured at (308.15 ± 0.01) K and (313.15 ± 0.01) K by using an Anton Paar DMA 60/602 density meter. The stated uncertainty in the density meter measurements was estimated to be  $\pm 1.5 \times 10^{-5}$  kg L<sup>-1</sup>. The density meter was placed in a temperature-controlled environment. The density meter was connected to a Haake N4-B circulator fitted with a calibrated platinum temperature sensor (IPTS-68). The temperature in the bath was also measured by a DP95 digital RTD thermometer (ITS-90) supplied by Omega. The temperature fluctuations within the density meter measuring cell were kept to within ±0.01 K. Densities were determined from the density meter readings by the equation

$$\rho = \frac{A\tau^2}{1 - B\tau^2} - C \quad (1)$$

where  $\rho$  is the density and  $\tau$  is the period of oscillation indicated by the density meter. The values of parameters  $A$ ,  $B$ , and  $C$  were determined at (308.15 and 313.15) K by using compounds with accurately known densities at those temperatures that are not used to constitute the systems under investigation. The densities of the compounds used for calibration were selected such that their densities cover the range of the densities of the components constituting the liquid mixtures under investigation.

Error analysis showed that at the maximum fluctuation in density meter readings an uncertainty of less than  $1.5 \times 10^{-4}$  kg L<sup>-1</sup> in the density would result. Also, a temper-

\* Corresponding author. E-mail: asfour@uwindsor.ca or aasfour@cogeco.net. Fax: (519) 735-6112.

**Table 2. Pure Component Densities and Kinematic Viscosities and Their Comparison with Literature Values at (308.15 and 313.15) K**

compound	$\rho/\text{g}\cdot\text{cm}^{-3}$		$10^6 \nu/\text{m}^2\cdot\text{s}^{-1}$	
	exptl	lit	exptl	lit
$T = 308.15 \text{ K}$				
toluene	0.8529	0.8577 <sup>9</sup>	0.5811	0.5741 <sup>9</sup>
octane	0.6904	0.69042 <sup>10</sup>	0.6603	0.6614 <sup>9</sup>
ethylbenzene	0.8539	0.8548 <sup>11</sup>	0.6632	0.6614 <sup>9</sup>
tetradecane	0.7521	0.75212 <sup>12</sup>	2.2770	2.3050 <sup>9</sup>
hexadecane	0.7631	0.7632 <sup>13</sup>	3.2250	3.252 <sup>9</sup>
$T = 313.15 \text{ K}$				
toluene	0.8484	0.4885 <sup>9</sup>	0.5509	0.5465 <sup>9</sup>
octane	0.6864	0.6863 <sup>9</sup>	0.6254	0.6286 <sup>9</sup>
ethylbenzene	0.8496	0.8495 <sup>9</sup>	0.6265	0.6286 <sup>9</sup>
tetradecane	0.7487	0.7491 <sup>9</sup>	2.0920	2.1180 <sup>9</sup>
hexadecane	0.7597	0.7595 <sup>9</sup>	2.9280	2.9530 <sup>9</sup>

ature variation of  $\pm 0.01 \text{ }^\circ\text{C}$  would result in an uncertainty of  $3 \times 10^{-6} \text{ kg L}^{-1}$  in the density.

**Viscosity Measurements.** Kinematic viscosity measurements were performed by using a set of five Cannon-Ubbelohde type viscometers with an estimated uncertainty in measurement of  $\pm 0.1\%$ . The set consists of two viscometers of size 24A (range of  $(0.5\text{--}2.0) \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$ ), two of size 50B (range of  $(0.8\text{--}4.0) \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$ ), and one of size 75J (range of  $(1.6\text{--}8.0) \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$ ). The range of viscosities of the investigated systems varied from  $(0.55\text{--}4.5) \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$ . Kinematic viscosities were obtained from the measured efflux time,  $t$ , and the equation

$$\nu = Et - \frac{F}{t^2} \quad (2)$$

where  $E$  and  $F$  are calibration constants. These calibration constants were determined by using calibration standards purchased from Cannon Instrument Company. The range of the standards covers the viscosity range of interest. The viscometers were placed in a model CT-1000 constant-temperature bath supplied by Cannon. The bath temperature was controlled to within  $\pm 0.01 \text{ K}$  by using silicone oil as a bath medium. The unit was connected to an external refrigerating unit for measurements below the laboratory temperature. To monitor the temperature setting inside the bath, a DP95 digital RTD thermometer (ITS-90) purchased from Omega was used. Temperature fluctuations within the CT-1000 were kept to within  $\pm 0.005 \text{ }^\circ\text{C}$ . An electronic stopwatch accurate to within  $0.01 \text{ s}$  was used to measure the efflux times. The uncertainty in the case of the kinematic viscosities was estimated, through error analysis, to be better than  $5.5 \times 10^{-9} \text{ m}^2 \text{ s}^{-1}$ .

## Results and Discussion

The experimental density and kinematic viscosity of the pure components employed in this study are listed in Table 2 along with the corresponding literature values. As can be seen from Table 2, the reported experimental values of the densities and kinematic viscosities conform closely to their corresponding literature values.

The measured densities and the kinematic viscosities were used to calculate absolute viscosities and the viscosity deviations from a mole fraction-averaged viscosity of the mixture ( $\Delta\eta$ ). The viscosity deviation is defined as

$$\Delta\eta = \eta - \sum_{i=1}^n x_i \eta_i \quad (3)$$

In the above equation,  $\eta$  is the absolute viscosity,  $x$  is the component mole fraction, and subscript  $i$  stands for pure

component  $i$ . As can be seen from Tables 3 and 4, the values of  $\Delta\eta$  are always negative.

The experimental kinematic viscosities of the investigated systems at 308.15 and 313.15 K are listed in Tables 3 and 4. For each measured viscosity value, the corresponding efflux time was the average of three measurements with a reproducibility of better than  $0.1\%$ . The maximum expected uncertainty is less than  $\pm 3 \times 10^{-10} \text{ m}^2 \text{ s}^{-1}$ .

The viscosity–composition data presented in Tables 3 and 4 were utilized to test the predictive capabilities of the McAllister model and the Grunberg–Nissan equation. In our earlier work,<sup>1</sup> the McAllister equation for binary systems was extended to treat  $n$ -component liquid systems. The following equation was reported:

$$\ln \nu_m = \sum_{i=1}^n x_i^3 \ln(\nu_i M_i) + 3 \sum_{i=1}^n \sum_{\substack{j=1 \\ i \neq j}}^n x_i^2 x_j \ln(\nu_{ij} M_{ij}) + 6 \sum_{i=1}^n \sum_{\substack{j=1 \\ i \neq j}}^n \sum_{\substack{k=1 \\ i \neq j \neq k}}^n x_i x_j x_k \ln(\nu_{ijk} M_{ijk}) - \ln(M_{av}) \quad (4)$$

Because of the assumption of three-body interactions,<sup>2</sup> only two types of interaction parameters are involved: the binary interaction parameters and the ternary interaction parameters,  $\nu_{ijk}$ . The number of those parameters depends on the number of components in a liquid mixture. The McAllister equation has been extensively used in the literature as a correlative technique. In other words, to use the McAllister equation, viscosity–composition data are needed. It is not always possible, however, to find reliable experimental values of this property for liquid mixtures of interest in the literature, nor is it practical to measure the property as the need arises. Consequently, reliable predictive models for the dependence of the viscosity of liquid systems can alleviate the need for costly and time-consuming experimental work.

In the present work, the binary interaction parameters were predicted by using the technique proposed by Asfour et al.<sup>3</sup> Accordingly, the binary interaction parameters can be predicted as follows:

$$\frac{\nu_{ij}}{(\nu_i^2 \nu_j)^{1/3}} = 1 + 0.044 \frac{(N_j - N_i)^2}{(N_i^2 N_j)^{1/3}} \quad (5)$$

$$\nu_{ji} = \nu_{ij} \left( \frac{\nu_j}{\nu_i} \right)^{1/3} \quad (6)$$

In the above equations, ( $i < j$ ),  $N_i$  and  $N_j$  are the effective carbon numbers of pure components of the regular mixture or carbon numbers in the case of  $n$ -alkane mixtures. To determine the numerical value of the effective carbon number, the following equation was suggested by Nhaesi and Asfour<sup>4</sup>

$$\ln \nu = -1.943 + 0.193N \quad (7)$$

where  $\nu$  is the kinematic viscosity at 308.15 K in cSt. and  $N$  is the effective carbon number. The ternary interaction parameter was estimated by means of the following equation, which was reported by Nhaesi and Asfour<sup>1</sup>

$$\frac{\nu_{ijk}}{(\nu_i \nu_j \nu_k)^{1/3}} = 0.9637 + \frac{(N_k - N_i)^2}{N_j} \quad (8)$$

where  $i < j < k$ .

**Table 3. Viscosity and Density Data for the Quaternary Subsystems of the Quinary Mixture: Toluene (1) + Octane (2) + Ethylbenzene (3) + Tetradecane (4) + Hexadecane (5) at (308.15 and 313.15) K**

mole fraction							mole fraction						
$x_1$	$x_2$	$x_3$	$\rho/\text{g}\cdot\text{cm}^{-3}$	$10^6\nu/\text{m}^2\cdot\text{s}^{-1}$	$10^3\eta/\text{Pa}\cdot\text{s}$	$10^3\Delta\eta/\text{Pa}\cdot\text{s}$	$x_1$	$x_2$	$x_3$	$\rho/\text{g}\cdot\text{cm}^{-3}$	$10^6\nu/\text{m}^2\cdot\text{s}^{-1}$	$10^3\eta/\text{Pa}\cdot\text{s}$	$10^3\Delta\eta/\text{Pa}\cdot\text{s}$
$T = 308.15\text{ K}$							$T = 313.15\text{ K}$						
Toluene (1) + Octane (2) + Ethylbenzene (3) + Tetradecane (4)							Toluene (1) + Octane (2) + Ethylbenzene (3) + Tetradecane (4)						
1.0000	0.0000	0.0000	0.8529	0.5811	0.4956	0.0000	1.0000	0.0000	0.0000	0.8484	0.5509	0.4674	0.0000
0.0000	1.0000	0.0000	0.6904	0.6603	0.4559	0.0000	0.0000	1.0000	0.0000	0.6864	0.6254	0.4293	0.0000
0.0000	0.0000	1.0000	0.8539	0.6632	0.5826	0.0000	0.0000	0.0000	1.0000	0.8496	0.6265	0.5323	0.0000
0.0000	0.0000	0.0000	0.7521	2.2770	1.7126	0.0000	0.0000	0.0000	0.0000	0.7487	2.0920	1.5662	0.0000
0.7011	0.1065	0.0932	0.8092	0.7024	0.5684	-0.0518	0.6905	0.1092	0.0999	0.8044	0.6674	0.5369	-0.0426
0.0982	0.6999	0.0923	0.7224	0.7622	0.5506	-0.0586	0.0951	0.6997	0.1012	0.7188	0.7135	0.5129	-0.0481
0.0327	0.1539	0.7089	0.8113	0.7713	0.6257	-0.0526	0.0943	0.1203	0.6719	0.8043	0.7253	0.5834	-0.0437
0.2545	0.2244	0.2701	0.7742	0.9329	0.7223	-0.0934	0.2492	0.2314	0.2615	0.7688	0.8893	0.6837	-0.0737
0.1389	0.0971	0.1127	0.7602	1.5805	1.2015	-0.0927	0.1368	0.0859	0.1421	0.7588	1.4484	1.0990	-0.0715
0.1609	0.2909	0.2330	0.7646	0.9737	0.7445	-0.1434	0.1575	0.2918	0.2774	0.7601	0.9075	0.6898	-0.0831
0.2256	0.1028	0.2961	0.7793	1.1169	0.8704	-0.1039	0.2125	0.1104	0.2891	0.7735	1.0689	0.8268	-0.0797
$T = 308.15\text{ K}$							$T = 313.15\text{ K}$						
Toluene (1) + Octane (2) + Ethylbenzene (3) + Hexadecane (4)							Toluene (1) + Octane (2) + Ethylbenzene (3) + Hexadecane (4)						
1.0000	0.0000	0.0000	0.8529	0.5811	0.4956	0.0000	1.0000	0.0000	0.0000	0.8484	0.5509	0.4674	0.0000
0.0000	1.0000	0.0000	0.6904	0.6603	0.4559	0.0000	0.0000	1.0000	0.0000	0.6864	0.6254	0.4293	0.0000
0.0000	0.0000	1.0000	0.8539	0.6632	0.5663	0.0000	0.0000	0.0000	1.0000	0.8496	0.6265	0.5323	0.0000
0.0000	0.0000	0.0000	0.7631	3.2250	2.4609	0.0000	0.0000	0.0000	0.0000	0.7597	2.9280	2.2243	0.0000
0.0911	0.6896	0.1085	0.7266	0.8205	0.5962	-0.0992	0.0911	0.6896	0.1085	0.7227	0.7740	0.5593	-0.0828
0.1127	0.0924	0.6862	0.8146	0.8211	0.6689	-0.0964	0.1127	0.0924	0.6862	0.8105	0.7738	0.6272	-0.0681
0.0939	0.3026	0.3154	0.7665	1.1719	0.8983	-0.1790	0.0939	0.3026	0.3154	0.7627	1.0908	0.8320	-0.1486
0.2004	0.0921	0.2988	0.7827	1.4489	1.1340	-0.1871	0.2004	0.0921	0.2988	0.7790	1.3610	1.0603	-0.1393
0.1144	0.1175	0.1015	0.7658	2.1365	1.6362	-0.1736	0.1144	0.1175	0.1015	0.7623	1.9736	1.5046	-0.1355
0.6840	0.1055	0.1163	0.8112	0.7503	0.6086	-0.0780	0.6840	0.1055	0.1163	0.8070	0.7233	0.5837	-0.0520
0.2535	0.2405	0.2490	0.7752	1.0932	0.8474	-0.1652	0.2535	0.2405	0.2490	0.7714	1.0269	0.7922	-0.1321
$T = 308.15\text{ K}$							$T = 313.15\text{ K}$						
Toluene (1) + Ethylbenzene (2) + Tetradecane (3) + Hexadecane (4)							Toluene (1) + Ethylbenzene (2) + Tetradecane (3) + Hexadecane (4)						
1.0000	0.0000	0.0000	0.8529	0.5811	0.4956	0.0000	1.0000	0.0000	0.0000	0.8484	0.5509	0.4674	0.0000
0.0000	1.0000	0.0000	0.8539	0.6632	0.5663	0.0000	0.0000	1.0000	0.0000	0.8496	0.6265	0.5323	0.0000
0.0000	0.0000	1.0000	0.7521	2.2770	1.7126	0.0000	0.0000	0.0000	1.0000	0.7487	2.0920	1.5662	0.0000
0.0000	0.0000	0.0000	0.7631	3.2250	2.4609	0.0000	0.0000	0.0000	0.0000	0.7597	2.9280	2.2243	0.0000
0.0723	0.6724	0.1276	0.8093	1.0380	0.8400	-0.1203	0.0723	0.6724	0.1276	0.8054	0.9733	0.7839	-0.0877
0.6492	0.1446	0.1480	0.8129	0.8871	0.7211	-0.0815	0.6492	0.1446	0.1480	0.8088	0.8343	0.6748	-0.0660
0.0588	0.1832	0.6975	0.7649	1.8211	1.3930	-0.0863	0.0588	0.1832	0.6975	0.7614	1.6858	1.2836	-0.0674
0.3226	0.2836	0.2954	0.7915	1.2252	0.9698	-0.1034	0.3226	0.2836	0.2954	0.7877	1.1566	0.9111	-0.0705
0.0882	0.3423	0.3630	0.7788	1.5906	1.2387	-0.1342	0.0882	0.3423	0.3630	0.7752	1.4840	1.1503	-0.0989
0.2452	0.3328	0.3864	0.7885	1.2175	0.9600	-0.1047	0.2452	0.3328	0.3864	0.7848	1.1375	0.8927	-0.0815
0.1304	0.2266	0.1763	0.7761	1.9381	1.5042	-0.1428	0.1304	0.2266	0.1763	0.7726	1.7957	1.3873	-0.1071
$T = 308.15\text{ K}$							$T = 313.15\text{ K}$						
Octane (1) + Ethylbenzene (2) + Tetradecane (3) + Hexadecane (4)							Octane (1) + Ethylbenzene (2) + Tetradecane (3) + Hexadecane (4)						
1.0000	0.0000	0.0000	0.6904	0.6603	0.4559	0.0000	1.0000	0.0000	0.0000	0.6864	0.6254	0.4293	0.0000
0.0000	1.0000	0.0000	0.8539	0.6632	0.5663	0.0000	0.0000	1.0000	0.0000	0.8496	0.6265	0.5323	0.0000
0.0000	0.0000	1.0000	0.7521	2.2770	1.7126	0.0000	0.0000	0.0000	1.0000	0.7487	2.0920	1.5662	0.0000
0.0000	0.0000	0.0000	0.7631	3.2250	2.4609	0.0000	0.0000	0.0000	0.0000	0.7597	2.9280	2.2243	0.0000
0.0940	0.7166	0.0945	0.8034	0.9281	0.7456	-0.1101	0.0940	0.7166	0.0945	0.7995	0.8714	0.6967	-0.0799
0.7033	0.1116	0.0877	0.7225	0.9265	0.6694	-0.1062	0.7033	0.1116	0.0877	0.7187	0.8687	0.6243	-0.0904
0.1082	0.2866	0.3004	0.7668	1.7250	1.3227	-0.1582	0.1082	0.2866	0.3004	0.7632	1.6008	1.2217	-0.1240
0.1022	0.1046	0.6917	0.7544	1.9268	1.4535	-0.0884	0.1022	0.1046	0.6917	0.7509	1.7867	1.3416	-0.0665
0.0792	0.1074	0.0991	0.7626	2.4981	1.9051	-0.1211	0.0792	0.1074	0.0991	0.7592	2.2950	1.7423	-0.0922
0.1772	0.1004	0.2883	0.7600	2.0660	1.5701	-0.1312	0.1772	0.1004	0.2883	0.7564	1.9075	1.4428	-0.1032
0.2349	0.2686	0.2434	0.7556	1.4930	1.1280	-0.1752	0.2349	0.2686	0.2434	0.7521	1.3933	1.0479	-0.1385
$T = 308.15\text{ K}$							$T = 313.15\text{ K}$						
Toluene (1) + Octane (2) + Tetradecane (3) + Hexadecane (4)							Toluene (1) + Octane (2) + Tetradecane (3) + Hexadecane (4)						
1.0000	0.0000	0.0000	0.8529	0.5811	0.4956	0.0000	1.0000	0.0000	0.0000	0.8484	0.5509	0.4674	0.0000
0.0000	1.0000	0.0000	0.6904	0.6603	0.4559	0.0000	0.0000	1.0000	0.0000	0.6864	0.6254	0.4293	0.0000
0.0000	0.0000	1.0000	0.7521	2.2770	1.7126	0.0000	0.0000	0.0000	1.0000	0.7487	2.0920	1.5662	0.0000
0.0000	0.0000	0.0000	0.7631	3.2250	2.4609	0.0000	0.0000	0.0000	0.0000	0.7597	2.9280	2.2243	0.0000
0.0872	0.7236	0.1025	0.7185	0.9164	0.6584	-0.1036	0.0872	0.7236	0.1025	0.7143	0.9164	0.6546	-0.0502
0.1012	0.1184	0.7011	0.7524	1.8844	1.4178	-0.0822	0.1012	0.1184	0.7011	0.7489	1.8844	1.4113	-0.0387
0.1068	0.0863	0.1152	0.7614	2.4577	1.8713	-0.1205	0.1068	0.0863	0.1152	0.7580	2.4577	1.8629	-0.0569
0.1095	0.2916	0.3030	0.7485	1.6986	1.2714	-0.1629	0.1095	0.2916	0.3030	0.7449	1.6986	1.2653	-0.0438
0.1717	0.1083	0.3310	0.7600	2.0103	1.5277	-0.1309	0.1717	0.1083	0.3310	0.7565	2.0103	1.5207	-0.0103
0.2473	0.2720	0.2270	0.7550	1.4466	1.0922	-0.1675	0.2473	0.2720	0.2270	0.7513	1.4466	1.0869	-0.0653
0.6792	0.0956	0.1287	0.7939	0.9178	0.7266	0.1094	0.6792	0.0956	0.1287	0.7900	0.9178	0.7250	-0.0497

Equation 8 was tested by using the viscosity data of quinary and quaternary regular systems generated in our laboratory. First, the values of  $\nu_{ijk}$  for each system were calculated by means of eq 8, and the binary interaction parameters, for the binary subsystems of the quinary and

quaternary systems under consideration, were predicted by eqs 5–7. The values of the predicted parameters were then substituted into the McAllister three-body model for  $n$ -component liquid systems, eq 4, to obtain the predicted viscosity. The calculated and experimental viscosities were

**Table 4. Viscosity and Density Data for The Quinary System: Toluene(1) + Octane(2) + Ethylbenzene(3) + Tetradecane(4) + Hexadecane(5) at (303.15 and 313.15) K**

$x_1$	$x_2$	$x_3$	$x_4$	$\rho/\text{g}\cdot\text{cm}^{-3}$	$10^6\nu/\text{m}^2\cdot\text{s}^{-1}$	$10^3\eta/\text{Pa}\cdot\text{s}$	$10^3\Delta\eta/\text{Pa}\cdot\text{s}$
$T = 308.15\text{ K}$							
1.0000	0.0000	0.0000	0.0000	0.8529	0.5811	0.4956	0.0000
0.0000	1.0000	0.0000	0.0000	0.6904	0.6603	0.4559	0.0000
0.0000	0.0000	1.0000	0.0000	0.8539	0.6632	0.5663	0.0000
0.0000	0.0000	0.0000	1.0000	0.7521	2.2770	1.7126	0.0000
0.0000	0.0000	0.0000	0.0000	0.7631	3.2250	2.4609	0.0000
0.4716	0.1968	0.1993	0.0781	0.7904	0.7888	0.6235	-0.0832
0.0517	0.5990	0.1756	0.1122	0.7338	0.8804	0.6461	-0.0984
0.4050	0.2941	0.0855	0.0486	0.7689	0.9749	0.7496	-0.1287
0.1205	0.0407	0.4157	0.1403	0.7866	1.3884	1.0921	-0.1645
0.0723	0.1052	0.0655	0.2002	0.7610	2.2386	1.7036	-0.1314
0.1704	0.0650	0.6029	0.0580	0.8109	0.8862	0.7187	-0.1012
0.1777	0.3792	0.0931	0.3080	0.7481	1.1075	0.8285	-0.1175
0.0780	0.1611	0.2941	0.3994	0.7667	1.3134	1.0070	-0.1264
0.2810	0.0388	0.1701	0.1050	0.7799	1.6342	1.2746	-0.1582
0.0715	0.0433	0.0603	0.6177	0.7588	2.0852	1.5822	-0.0759
$T = 313.15\text{ K}$							
1.0000	0.0000	0.0000	0.0000	0.8484	0.5509	0.4674	0.0000
0.0000	1.0000	0.0000	0.0000	0.6864	0.6254	0.4293	0.0000
0.0000	0.0000	1.0000	0.0000	0.8496	0.6265	0.5323	0.0000
0.0000	0.0000	0.0000	1.0000	0.7487	2.0920	1.5662	0.0000
0.0000	0.0000	0.0000	0.0000	0.7597	2.9280	2.2243	0.0000
0.6703	0.0806	0.1087	0.0834	0.8038	0.7416	0.5961	-0.0664
0.1053	0.5587	0.1805	0.1023	0.7351	0.7884	0.5796	-0.0831
0.1810	0.0632	0.6065	0.0538	0.8089	0.8130	0.6576	-0.0700
0.1153	0.1463	0.3038	0.3721	0.7667	1.1649	0.8932	-0.1053
0.1864	0.1596	0.1485	0.1091	0.7708	1.4665	1.1304	-0.1560
0.1661	0.1905	0.1980	0.2468	0.7690	1.2509	0.9619	-0.1300

**Table 5. Testing the McAllister Model and Grunberg–Nissan Equation by Using Viscosity Data on Regular Quaternary Systems at (308.15 and 313.15) K**

system	$T/\text{K}$	McAllister model		Grunberg–Nissan eq	
		AAD	MAX	AAD	MAX
toluene (1) + octane (2) + ethylbenzene (3) + tetradecane (4)	308.15	1.32	-2.54	4.21	-8.81
	313.15	0.39	-1.22	4.17	-7.45
toluene (1) + octane (2) + ethylbenzene (3) + hexadecane (4)	308.15	0.59	-2.67	3.29	-6.86
	313.15	1.05	3.24	3.40	-6.47
toluene (1) + ethylbenzene (2) + tetradecane (3) + hexadecane (4)	308.15	0.64	2.42	1.05	2.40
	313.15	1.33	2.49	1.19	2.81
octane (1) + ethylbenzene (2) + tetradecane (3) + hexadecane (4)	308.15	0.48	-1.69	3.86	-6.04
	313.15	0.59	1.61	4.40	-7.61
toluene (1) + octane (2) + tetradecane (3) + hexadecane (4)	308.15	0.46	-1.73	1.45	-3.02
	313.15	0.49	-1.43	4.50	5.83
overall % AAD		0.75		3.15	

compared. The percent average absolute deviation (% AAD), which is defined by

$$\% \text{AAD} = \frac{1}{n} \sum_{i=1}^n \frac{|v_i^{\text{exptl}} - v_i^{\text{calcd}}|}{v_i^{\text{exptl}}} \times 100 \quad (9)$$

was calculated and reported.

The maximum deviation of the predicted data is determined by means of function max, which is defined as

$$\% \text{max} = \max \left( \frac{|v_i^{\text{exptl}} - v_i^{\text{calcd}}|}{v_i^{\text{exptl}}} \right) \times 100 \quad (10)$$

The results of testing the model are reported in Tables 5 and 6. Furthermore, the reported absolute viscosity data were used to test the predictive capability of the Grunberg–Nissan equation. The Grunberg–Nissan equation<sup>5</sup> has been recommended by Irving<sup>6</sup> as being the most effective

**Table 6. Testing the McAllister Model and Grunberg–Nissan Equation by Using Viscosity Data on the Regular Quinary System: Toluene (1) + Octane (2) + Ethylbenzene (3) + Tetradecane (4) + Hexadecane (5) at (308.15 and 313.15) K**

$T/\text{K}$	McAllister model		Grunberg–Nissan eq	
	AAD	MAX	AAD	MAX
308.15	0.37	2.06	2.34	-7.12
313.15	0.48	1.99	3.79	-8.54
overall % AAD	0.42	3.07		

equation in correlating viscosity data for binary liquid mixtures. This purely empirical equation is given by

$$\ln \eta_m = \sum_{i=1}^n x_i \ln \eta_i + \sum_{i<j}^n \sum_{j=1}^n x_i x_j G_{ij} \quad (11)$$

The model given by eq 11 contains binary adjustable parameters,  $G_{ij}$ . These adjustable parameters are determined by fitting experimental viscosity–composition data

to eq 11. Isdale et al.<sup>7</sup> reported a group contribution method to predict the binary adjustable parameter  $G_{ij}$ . The number of those parameters depends on the number of components in the system. In the present study, the calculation procedure outlined by Reid et al.<sup>8</sup> was followed. The results are listed in Tables 5 and 6.

A comparison of the McAllister model and the Grunberg–Nissan equation (Tables 5 and 6) for all the systems investigated in the present study clearly indicates that the McAllister model performed much better than the Grunberg–Nissan model over the entire composition range. The overall %AADs of the McAllister model are lower than those of the Grunberg–Nissan equation.

## Conclusions

The kinematic viscosities of a quinary regular system and all of its quaternary liquid subsystems at (308.15 and 313.15) K were measured over the entire composition range with a maximum estimated error of less than  $\pm 3 \times 10^{-10} \text{ m}^2 \cdot \text{s}^{-1}$ .

Using the pure component properties, the viscosity of liquid mixtures was predicted by using two different techniques, namely, the McAllister model and the Grunberg–Nissan equation. The obtained results indicate that the McAllister model predicts the data much better than the Grunberg–Nissan equation for the systems reported in this study.

## Literature Cited

- (1) Nhaesi, A. H.; Asfour, A. A. Prediction of the Viscosity of Multi-component Liquid Mixtures: A generalized McAllister Three-body Interaction Model. *Chem. Eng. Sci.* **2000**, *55*, 2861–2873.

- (2) McAllister, R. A. The viscosity of liquid mixtures. *AIChE J.* **1960**, *6*, 427–431.
- (3) Asfour, A. A.; Cooper, E. F.; Wu, J.; Zahran, R. Prediction of the McAllister Model Parameters from Pure Components Properties for Liquid Binary *n*-Alkane Systems. *Ind. Eng. Chem. Res.* **1991**, *30*, 1666–1669.
- (4) Nhaesi, A. H.; Asfour, A. A. Prediction of the McAllister Model Parameters from Pure Component Properties of Regular Binary Liquid Mixtures. *Ind. Eng. Chem. Res.* **1998**, *37*, 4893–4897.
- (5) Grunberg, L.; Nissan, A. H. Mixture Law for Viscosity. *Nature* **1949**, *164*, 799.
- (6) Irving, J. B. *The Effectiveness of Mixture Equations*; National Engineering Laboratory Report 630; National Engineering Laboratory: East Kilbride, Glasgow, Scotland, 1977.
- (7) Isdale, J. D.; MacGillivray, J. C. National Engineering Laboratory Report 631; National Engineering Laboratory: East Kilbride, Glasgow, Scotland, 1985.
- (8) Reid, R. C.; Praunitz, J. M.; Poling, B. E. *The Properties of Gases and Liquids*; McGraw-Hill: New York, 1987.
- (9) *TRC Thermodynamic Tables – Hydrocarbons*; Thermodynamic Research Center, Texas A&M University: College Station, TX, 1988; Supplement No. 92.
- (10) Inglese, A.; Grolier, J.; Wilhelm, E. Excess Volume of Mixtures of Oxolane, 1,3-Dioxolane, and 1,4-Dioxane with *n*-Alkanes at 298.15 and 318.15 K. *J. Chem. Eng. Data* **1983**, *28*, 124–127.
- (11) Puri P. S.; Raju, K. S. N. Vapor-Liquid Equilibria Systems *p*-Xylene–Furfural and Ethylbenzene–Furfural. *J. Chem. Eng. Data* **1970**, *15*, 480–483.
- (12) Aicart, E.; Tardajos, G.; Pena, M. D. Isothermal Compressibility of Cyclohexane + *n*-Decane, Cyclohexane + *n*-Dodecane, and Cyclohexane + *n*-Tetradecane. *J. Chem. Eng. Data* **1981**, *26*, 22–26.
- (13) Gomez-Ibanez, J. D.; Liu, C. The Excess Volume of Binary Mixtures of Normal Alkanes. *J. Phys. Chem.* **1963**, *67*, 1388–1391.

Received for review June 21, 2004. Accepted September 21, 2004. We acknowledge with thanks the financial support from the Natural Sciences and Engineering Research Council of Canada.

JE049768F