Prediction of Viscosity of Liquid Hydrocarbon Mixtures

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Despite the concentrated efforts directed at understanding the mechanism of the molecular momentum transfer in liquids, the progress on the development of a sound and reliable predictive theory for the viscosity of liquids and their mixtures has been rather slow. Currently available models of the liquid state emphasize similarity either with gas-like or solid-like behavior. Unfortunately, neither of these approaches has had much success in yielding the values of viscosity for pure liquids and mixtures as functions of temperature. Most of the progress in this area has therefore been made through the use of semi-empirical/empirical considerations. Consequently, there is no scarcity of predictive expressions available in the literature, and most of these, for pure components, have been reviewed and evaluated by Przezdziecki and Sridhar (1985), Reid et al. (1987), and more recently by Mehrotra (1991a), whereas the pertinent literature on the prediction of viscosities of liquid mixtures has been summarized by Liu et al. (1991). Most predictive schemes available for estimating the viscosity of liquid mixtures (for example, Reid et al., 1987; Liu et al., 1991) entail unknown interaction parameters which sometime show further dependence on composition, temperature, and/or the system itself. Thus, the need to develop a simple method for predicting the viscosity of liquid mixtures is obvious.

In a recent article, Mehrotra (1991b) has presented the following single-parameter equation for estimating the viscosity of pure liquids (primarily heavy hydrocarbons) as the function of temperature:

$$\log(\mu + 0.8) = 100\ (0.01T)^b \tag{1}$$

In Eq. 1, μ is the viscosity in m·Pa·s at temperature T(K), and b is a characteristic constant for each substance. Although the parameter b seems to show systematic variation with the logarithm of molar mass as well as with the reciprocal of boiling point (at 10 mm Hg), Mehrotra (1991b) obtained the best values of b via nonlinear regression of the viscosity-temperature data. Indeed, he showed that the discrepancy between the predicted and experimental values of viscosity seldom exceeded 7–8% for 273 individual components including n-Paraffins, 1-Olefins, branched paraffins and olefins, nonfused aromatics, fused and nonfused ring aromatics, and naphthenes. The range of applications shows the extremely versatile nature of this approach. Thus, the temptation to extend this approach to the mixtures of liquids is irresistible. In this communication, we examine this contention and develop mixture rules for the prediction of b without introducing any further adjustable parameter. We conclude this article by presenting extensive comparisons between the predictions and experimental values of viscosity for scores of liquid mixtures.

Mixing Rules for Prediction of b

Numerous expressions of varying forms but involving only the values of b for pure components and the composition of mixture (mass/volume/mole basis) were chosen to test their effectiveness in yielding the values of mixture viscosities. Some of the forms tested are:

$$b_{\text{mix}} = \Sigma x_i b_i \tag{2a}$$

$$b_{\text{mix}} = b_1^{x_1} b_2^{x_2} b_3^{x_3} \ldots\ldots\ldots \tag{2b}$$

$$b_{\text{mix}} = \Sigma (x_i / b_i) \tag{2c}$$

where x_i, molar composition, was replaced by mass and volume fractions, respectively. Unfortunately, none of the aforementioned expressions proved to be completely satisfactory. Finally, the following general form for b_{mix} was postulated:

$$b_{\text{mix}} = [x_1 (b^n)_1 + x_2 (b^n)_2]^{1/n} \tag{3}$$

For a prechosen value of n, the viscosity of a large number of binary mixtures comprising a wide variety of organic compounds was calculated and compared with the corresponding experimental value at different temperatures. The parameter n was varied from -5 to $+5$ in steps of 0.05. Based on this approach, $n = -2$ was found to give the overall minimum average and maximum errors in the predicted viscosity for scores of mixtures. Thus, the mixing rule for the prediction of b_{mix} becomes:

$$b_{\text{mix}} = \left\{ \frac{x_1}{(b^2)_1} + \frac{x_2}{(b^2)_2} \right\}^{-1/2} \tag{4}$$

where x_1, x_2 are the mole fractions of the two components,

respectively. Obviously, only the negative root of Eq. 4 is meaningful to depict the correct temperature dependence of viscosity. In the next section, the utility and effectiveness of this simple method in predicting the values of mixture viscosities are demonstrated.

Database and Comparison with Predictions

Experimental data on the viscosity of liquid mixtures at different levels of temperature and concentration are scant as compared to the corresponding data on pure components. Nor is there any comprehensive database available on liquid mixtures. Therefore, in this work, experimental data on viscosities of liquid mixtures have been culled from the open literature. Altogether, there are 135 binary mixtures involving 57 different compounds and nearly 2,125 individual data points, albeit a bulk of it pertains to 298 K.

The values of b for pure components have been taken from the compilation of Mehrotra (1991b), whereas for the systems not listed therein, these were extracted from the pure-component viscosity-temperature data. Hence, from a knowledge of pure-component b values, temperature and the molar composition, the value of b for a mixture was calculated using Eq. 4 which in turn permitted the calculation of the mixture viscosity from Eq. 1. Table 1 summarizes results and the resulting average and maximum % errors for each system. The resulting overall average deviation between predictions and experiments is 7% for 2,125 data points, whereas only about 50 systems display average errors larger than 10%. While assessing the comparisons shown in Table 1, the following factors must be borne in mind. First, the viscosity values quoted by different investigators under nominally identical conditions (temperature, composition, and purity) often do not agree with one another (for instance, see the discrepancy in the results for mixtures of paraffins at 298.15 K reported by different investigators). Secondly, Eq. 1 itself entails errors up to 50-55% in predicting the viscosity of pure components. There is no discernible trend present in Table 1 to identify the problematic cases. In view of the aforementioned factors coupled with the fact that the scheme proposed herein does not contain any adjustable parameter, the correspondence between the calculated and experimental values is regarded to be satisfactory and acceptable. Finally, it would be desirable to extend this method to ternary and quaternary mixtures. Though it is our belief that Eq. 3 itself would still be applicable but whether $n=2$ will lead to accurate predictions or not is a matter for further studies in this area.

Based on the single-parameter equation of Mehrotra (1991b), a simple method has been developed for predicting the viscosity of liquid mixtures. The scheme proposed herein necessitates only the pure-component values of the viscosity equation parameter and the molar composition of the mixture. The new method predicts about 2,125 data points for about 135 binary mixtures with an average error of 7%. However, more data, especially at high temperatures, are needed to generalize this conclusion over a wide range of conditions.

Table 1. Summary of Results

System	T(K)	N	% Error Avg.	% Error Max.	Data Source
n-Heptane/*n*-Hexane	293.15	9	0.81	1.10	1
	297.85	8	20.15	20.75	2
	298.15	9	18.67	20.04	3
		9	0.91	1.23	1
	308.15	9	1.03	1.41	
n-Heptane/*n*-Octane	293.15	9	0.60	0.82	
	298.15	9	15.48	19.48	3
		9	0.44	0.60	1
	308.15	9	0.56	0.76	
	313.15	9	0.69	0.94	
n-Octane/*n*-Nonane	298.15	9	8.21	8.77	3
n-Nonane/*n*-Decane	298.15	9	3.13	6.25	
n-Hexane/*n*-Octane	293.15	9	2.59	3.50	1
	298.15	9	15.75	17.82	3
		9	2.74	3.75	1
	308.15	9	3.25	4.44	
n-Heptane/*n*-Nonane	298.15	9	15.93	20.10	3
n-Octane/*n*-Decane	293.15	9	1.21	1.65	1
	298.15	9	4.44	8.01	3
		8	1.17	1.60	1
	308.15	9	1.29	1.77	
	313.15	9	1.86	2.55	
n-Decane/*n*-Dodecane	298.15	3	3.66	5.53	3
n-Dodecane/*n*-Tetradecane	298.15	3	8.05	9.31	
n-Tetradecane/*n*-Hexadecane	293.15	9	0.43	0.59	1
	298.15	3	9.18	9.73	3
		9	0.65	0.88	1
	308.15	9	0.59	0.80	
	313.15	9	0.54	0.74	
n-Hexane/*n*-Nonane	298.15	9	17.51	20.39	3
n-Heptane/*n*-Decane	293.15	9	3.14	4.33	1
	298.15	9	14.50	25.84	3
		9	3.28	4.47	1
	308.15	9	3.58	4.85	
	313.15	9	3.93	5.38	
n-Nonane/*n*-Dodecane	298.15	3	3.89	6.43	3
n-Hexane/*n*-Decane	293.15	9	7.24	9.93	1
	298.15	9	16.53	21.80	3
		9	7.67	10.3	1
	308.15	9	8.78	12.13	
n-Octane/*n*-Dodecane	298.15	3	5.36	9.46	3
n-Decane/*n*-Tetradecane	298.15	3	3.21	7.09	
n-Dodecane/*n*-Hexadecane	298.15	3	5.55	6.30	
n-Heptane/*n*-Dodecane	293.15	9	7.2	9.86	1
	298.15	3	15.12	22.78	3
		9	7.45	10.22	1
	308.15	9	7.86	11.00	
	313.15	9	8.23	11.36	
n-Nonane/*n*-Tetradecane	298.15	3	5.15	7.48	3
n-Hexane/*n*-Dodecane	298.15	3	18.90	25.59	3
n-Octane/*n*-Tetradecane	293.15	9	8.01	10.92	1
	298.15	3	7.40	12.45	3
		9	8.16	11.15	1
	303.16	5	10.73	15.05	4
	308.16	5	11.87	16.14	
		9	8.36	11.48	1
	313.15	9	8.23	11.26	

Table 1. Continued

System	T(K)	N	% Error Avg.	% Error Max.	Data Source
n-Decane/*n*-Hexadecane	298.15	3	2.84	3.56	3
n-Heptane/*n*-Tetradecane	293.15	9	12.50	17.27	1
	298.15	3	17.68	26.59	3
		9	7.10	9.64	1
	308.15	9	13.4	18.63	
	313.15	9	13.7	19.10	
n-Nonane/*n*-Hexadecane	298.15	3	8.35	12.34	3
n-Hexane/*n*-Tetradecane	298.15	3	23.22	31.98	
n-Octane/*n*-Hexadecane	298.15	3	12.52	17.09	
n-Heptane/*n*-Hexadecane	298.15	3	23.63	31.42	
n-Hexane/*n*-Hexadecane	298.15	3	28.48	36.57	
Anisole/Benzene	298.15	9	20.05	39.66	4
	303.15	9	17.88	35.59	
	308.15	9	15.25	31.61	
	313.15	9	16.50	33.99	
Anisole/Methyl Alcohol	298.15	9	5.89	9.30	
	303.15	9	5.50	8.97	
	308.15	9	5.30	8.89	
	313.15	9	5.22	8.55	
Anisole/Cyclohexane	298.15	9	13.51	19.43	5
	303.15	9	12.14	17.85	
	308.15	9	11.25	16.84	
	313.15	9	10.63	15.90	
Anisole/Nitrobenzene	298.15	9	3.62	9.73	5
	303.15	9	3.30	9.60	
	308.15	9	3.21	9.28	
Anisole/1,2 Dichloroethane	298.15	9	1.51	2.56	
	303.15	9	1.46	2.51	
	308.15	9	1.23	2.13	
	313.15	9	1.13	1.80	
Anisole/Chlorobenzene	298.15	9	2.22	2.50	
	303.15	9	2.18	2.59	
	308.15	9	1.94	2.43	
	313.15	9	2.21	2.55	
Anisole/Carbontetrachloride	298.15	9	1.37	2.29	
	303.15	9	1.49	2.46	
	308.15	9	1.30	2.15	
	313.15	9	1.39	2.27	
n-Methylacetamide/Methyl Alcohol	303.15	9	3.58	5.02	6
n-Methylacetamide/Ethyl Alcohol	303.15	9	5.28	7.20	
n-Methylacetamide/1-Propanol	303.15	7	8.02	10.25	
n-Methylacetamide/2-Propanol	303.15	9	6.13	9.35	
n-Methylacetamide/1-Butanol	303.15	9	7.45	10.81	
n-Methylacetamide/2-Methyl-2-Propanol	303.15	10	10.46	22.10	
n,n Dimethylacetamide/Methyl Alcohol	303.15	9	8.74	11.37	7
n,n Dimethylacetamide/Ethyl Alcohol	303.15	9	6.42	9.37	
n,n Dimethylacetamide/1-Propanol	303.15	9	13.14	19.10	
n.n Dimethylacetamide/2-Propanol	303.15	9	20.00	30.20	
n,n Dimethylacetamide/1-Butanol	303.15	9	14.59	20.58	
n,n Dimethylacetamide/2-Methyl-2-Propanol	303.15	9	24.89	36.75	
Methyl Alcohol/Ethyl Alcohol	298.15	8	2.65	3.81	8
Methyl Alcohol/2-Propanol	298.15	8	16.06	22.68	
Ethyl Alcohol/2-Propanol	298.15	8	7.09	12.10	
Acetone/Ethyl Alcohol	298.15	8	18.07	22.49	
Acetone/Methyl Alcohol	298.15	8	6.57	8.19	
n-Hexane/Ethyl Alcohol	298.15	8	15.46	19.54	
Acetone/*n*-Hexane	298.15	8	2.93	7.67	
2-Butanone/*n*-Hexane	297.85	9	6.72	15.92	2
n-Hexane/1-Hexanol	303.15	9	5.23	17.34	9
	313.15	9	4.71	16.54	
	323.15	9	3.76	9.35	
	333.15	9	2.29	5.89	
Ethyl Alcohol/*n*-Heptane	298.15	10	7.84	12.00	10
n-Heptane/2-Butanone	297.85	8	7.36	16.64	2
n-Heptane/Cyclohexanone	298.15	9	7.13	16.25	11
Benzene/Ethyl Alcohol	298.15	9	13.26	18.16	10
Benzene/2-Butanone	297.85	8	2.77	3.57	2
Cyclohexanone/Benzene	298.15	9	5.71	22.96	11
Cyclohexanone/Toluene	298.15	9	5.93	14.65	
Triethylamine/Methyl Alcohol	298.15	10	6.73	12.63	12
Triethylamine/Ethyl Alcohol	298.15	8	3.48	5.56	
Triethylamine/1-Propanol	298.15	8	7.25	14.51	
Benzene/Hexane	297.85	8	6.29	14.36	2
Benzene/Heptane	297.85	13	7.10	16.27	10
	298.15	11	6.27	15.51	
Toluene/*n*-Hexane	298.15	3	7.34	12.57	11
	303.15	9	11.86	17.64	9
	313.15	9	12.38	15.26	
	323.15	9	8.51	10.03	
	333.15	9	5.76	6.98	
n-Hexane/Chlorobenzene	303.15	9	10.37	17.37	
	313.15	9	10.48	16.25	
	323.15	9	8.13	12.32	
	333.15	9	6.90	9.71	
Carbon Tetrachloride/*n*-Propyl Acetate	303.15	10	1.73	2.65	13
	313.15	5	1.61	2.15	
Carbon Tetrachloride/*n*-Butyl Acetate	303.15	10	1.49	1.94	
	313.15	5	1.24	1.81	
Carbon Tetrachloride/Ethyl Burate	303.15	11	1.66	2.79	
	313.15	5	1.96	2.65	
Carbon Tetrachloride/Ethyl Acetate	293.15	9	1.57	3.13	14
Acetone/Carbon Tetrachloride	298.15	8	1.62	3.07	8
Cyclohexanone/Carbon Tetrachloride	298.15	9	3.49	5.13	11
Cyclohexane/Carbon Tetrachloride	293.15	9	1.94	3.35	14
	298.15	5	3.87	4.28	15
Carbon Tetrachloride/Benzene	293.15	9	1.75	3.64	14
	298.15	7	1.31	1.82	15
Chloroform/Dimethyl Sulfoxide	298.15	8	20.93	28.82	8
Chloroform/*n*-Hexane	298.15	8	13.34	17.05	

Table 1. Continued

System	T(K)	N	% Error Avg.	% Error Max.	Data Source
Chloroform/Methyl Alcohol	298.15	8	10.56	15.66	
Chloroform/Methyl Acetate	298.15	8	13.15	15.57	
Trichloro Ethylene/Methyl Ethyl Ketone	298.15	8	5.78	8.60	16
	308.15	8	4.99	7.00	
	318.15	8	4.93	7.02	
Trichloro Ethylene/Diethyl Ketone	298.15	9	5.39	7.46	
	308.15	9	3.99	5.49	
Trichloro Ethylene/Methyl Isobutyl Ketone	298.15	9	4.47	6.05	8
	308.15	9	2.81	5.10	
	318.15	9	1.70	2.42	
Trichloro Ethylene/Cyclohexanone	298.15	9	5.07	7.28	
	308.15	9	6.21	9.28	
	318.15	9	5.97	8.57	
Trichloro Ethylene/1,4 Dioxane	298.15	9	3.13	5.17	
	308.15	9	2.46	4.17	
	318.15	9	4.12	7.24	
Bromoform/Dimethyl Sulfoxide	318.15	6	22.30	28.54	17
Bromoform/Cyclohexane	318.15	5	2.24	3.20	
Bromoform/Bromobenzene	318.15	5	2.06	2.51	
Bromoform/Dimethyl Formamide	318.15	6	22.20	28.72	
Bromoform/Methyl Ethyl Ketone	318.15	5	14.79	18.81	
Bromoform/Ethyl Acetate	318.15	3	10.98	13.33	
Bromoform/Methyl Alcohol	318.15	5	20.91	28.96	
n-Pentane/1-Chlorobutane	298.15	9	6.55	8.48	18
n-Decane/1-Chlorobutane	298.15	9	2.83	4.20	
n-Dodecane/1-Chlorobutane	298.15	9	5.48	8.31	
n-Pentane/1-Chlorooctane	298.15	9	17.84	23.93	
n-Decane/1-Chlorooctane	298.15	9	3.42	4.42	
n-Dodecane/1-Chlorooctane	298.15	9	4.85	6.57	
n-Decane/1-Chlorooctadecane	298.15	9	13.35	18.19	
n-Dodecane/1-Chlorooctadecane	298.15	9	5.00	8.47	
Acetonitrile/1-Propanol	293.15	11	21.94	32.55	19
	303.15	11	19.37	29.08	
	313.15	11	17.41	26.75	
	323.15	11	14.44	22.25	
Benzene/Cyclohexane	298.15	8	13.44	17.93	15
Benzene/Hexafluorobenzene	323.15	1	4.65	4.65	20
	348.15	1	6.72	6.72	
	373.15	1	10.76	10.76	
Tetrahydrofuran/γ-Butyrolactone	278.15	6	4.81	6.44	21
	283.15	6	3.47	5.91	
	288.15	6	4.05	5.98	
	293.15	6	3.38	4.92	
	298.15	6	3.04	4.51	
Cyclohexanone/*n*-Hexane	298.15	9	8.96	23.25	15
Cyclohexanone/2,2,4 Trimethylpentane	298.15	9	23.00	32.57	
n-Methylaniline/Tetrachloroethane	303.15	9	3.13	4.78	22
n-Methylaniline/Dimethyl Sulfoxide	303.15	9	10.49	16.67	
Ethyl Alcohol/Cyclohexane	298.15	8	7.40	10.42	8
Acetone/Cyclohexane	298.15	8	16.99	23.10	8
n-Hexane/Cyclohexane	298.15	8	7.48	14.82	
Carbontetrachloride/Cyclohexane	298.15	7	3.43	4.29	8
Carbontetrachloride/2-Propanol	298.15	8	15.74	20.63	
Triethylamine/Chloroform	298.15	8	27.54	35.29	
Methyl Alcohol/Dimethyl Sulfoxide	298.15	8	14.90	19.57	
Triethylamine/Methyl Alcohol	298.15	8	23.28	29.35	
Triethylamine/Chlorobenzene	298.15	8	4.16	6.27	
Methyl Acetate/*n*-Hexane	298.15	8	8.57	28.84	
Cyclohexane/2-Propanol	298.15	8	18.54	24.32	
Toluene/Acetonitrile	298.15	3	5.72	6.79	23
	323.15	3	7.03	8.69	
Bromoform/Carbontetrachloride	318.15	5	0.89	1.25	14
Benzene/Cyclohexane	293.15	9	13.72	19.75	
Ethyl Acetate/Cyclohexane	293.15	9	17.52	25.07	
Ethyl Acetate/Benzene	293.15	9	2.20	3.82	
Toluene/Acetonitrile	288.15	9	14.42	21.88	24
	298.15	9	10.82	17.40	
	308.15	9	7.68	14.40	
Benzene/*n*-Propyl Acetate	303.15	11	2.37	3.87	13
	315.15	5	1.10	1.68	
Benzene/Ethyl Burate	303.15	11	1.29	2.19	
	313.15	5	0.61	0.80	
Methyl Alcohol/Acetone	298.15	13	7.84	14.50	25
Trimethyl Pentane/*n*-Decane	298.15	5	8.66	10.73	2
Trimethyl Pentane/*n*-Hexadecane	298.15	5	8.24	24.90	
2-Methyl Pentane/*n*-Hexadecane	298.15	5	8.24	22.52	
Toluene/*n*-Octane	293.15	9	3.17	4.50	26
	298.15	9	5.58	7.76	
	308.15	9	5.43	7.35	
	313.15	9	5.21	7.11	
Toluene/*n*-Decane	293.15	9	3.17	4.50	
	298.15	9	3.05	4.32	
	303.15	9	2.59	3.90	
	313.15	9	2.29	3.45	
Toluene/*n*-Dodecane	293.15	9	1.40	2.05	
	298.15	9	1.21	1.91	
	308.15	9	1.72	2.64	
	313.15	9	1.85	2.63	
Toluene/*n*-Tetradecane	293.15	9	6.48	8.90	
	298.15	9	6.44	8.89	
	308.15	9	7.37	10.10	
	313.15	9	7.31	10.21	
Toluene/*n*-Hexadecane	293.15	9	13.26	18.50	
	298.15	9	14.24	19.63	
	308.15	9	9.00	19.22	
	313.15	9	14.40	19.90	
Ethylbenzene/*n*-Octane	293.15	9	5.15	7.00	
	298.15	9	4.99	6.82	
	308.15	9	4.72	6.33	
	313.15	9	4.56	6.05	

Table 1. Continued

System	T(K)	N	% Error Avg.	% Error Max.	Data Source	System	T(K)	N	% Error Avg.	% Error Max.	Data Source
Ethylbenzene/*n*-Tetradecane	293.15	9	3.33	4.63	26	Ethylbenzene/*n*-Hexadecane	293.15	9	9.16	12.50	26
	298.15	9	3.80	5.14			298.15	9	9.24	12.61	
	308.15	9	3.95	5.36			308.15	9	12.12	17.42	
	313.15	9	4.22	5.98			313.15	9	9.35	13.20	

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