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The Viscosity of Liquid Toluene at Elevated Pressures

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This paper reports new measurements on the viscosity of toluene at pressures up to 500 MPa at 299 K and pressures up to 435 MPa at 323 K. The results are in close agreement with recent viscosity measurements from a vibrating-wire viscometer at pressures up to 250 MPa but confirm earlier measurements from a falling-body viscometer which show that the recently recommended polynomial expression for the representation of the pressure dependence of the viscosity of toluene up to 250 MPa underestimates the viscosity if applied at 500 MPa by about 30%. Modification of this polynomial expression by inclusion of one additional term is found to represent the data satisfactorily over the whole pressure range.

KEY WORDS: high pressure; toluene; viscosity.

1. INTRODUCTION

The recent development of the vibrating-wire viscometer [1] has led to a series of accurate measurements on the viscosity of certain liquid hydrocarbons at 303, 323, and 348 K for pressures up to 250 MPa [2]. These data have been represented by polynomial expressions in terms of pressure for the purposes of interpolation. In this paper, the application of these expressions to higher pressures is investigated in the case of toluene, which would make an excellent reference material for liquid viscosity at elevated pressures. New measurements on a falling-body instrument are reported at 299 K at pressures up to 500 MPa and at 323 K up to 435 MPa to provide data for a more accurate polynomial representation to cover this wider pressure range.

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2. VISCOSITY REPRESENTATION

Oliveira and Wakeham [2] represented their viscosity data for pressures up to 250 MPa by a polynomial in pressure which, in the case of toluene, was quadratic:

$$\eta_P = \sum_{i=0}^{2} a_i(P/P')^i \tag{1}$$

where P' is a scaling parameter for each temperature. At 323.15 K, $P' = 126.1$ MPa, $a_0 = 0.4227$, $a_1 = 0.4170$, and $a_2 = 0.1001$, and at 348.15 K, $P' = 125.6$ MPa, $a_0 = 0.3394$, $a_1 = 0.3241$, and $a_2 = 0.0735$, for η_P in mPa · s.

In view of the need for a reference material for liquid viscosity up to elevated pressures, for which toluene would be a good choice, it is important to determine how well these equations can represent higher pressure data. Measurements up to 500 MPa have been made by a falling-body method [3], and although these are of a lower accuracy ($\sim \pm 4\%$) than the vibrating-wire values, they can provide a good test of the applicability of these equations at higher pressures.

Figure 1 shows a comparison of the falling-body results for toluene at 323 and 348 K, the two common temperatures, with the values given by Eq. (1). The equation underestimates the viscosities by up to 30% at 500 MPa.

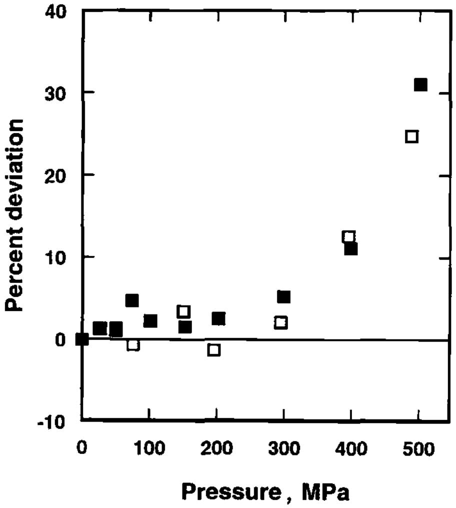

Fig. 1. Percentage deviation, $100(\eta_{\mathrm{exp}} - \eta_{\mathrm{calc}})/\eta_{\mathrm{calc}}$, between the viscosity data of Dymond et al. [3] (□, 323 K; ■, 348 K) and values calculated from Eq. (1).

To confirm the falling-body measurements and provide additional data to develop an equation valid up to 500 MPa, new measurements have been made.

3. EXPERIMENTS

Viscosities were determined from measurements of the fall time of a self-centering hemispherically nosed sinker down a vertical cylindrical tube. As the sinker passes through two coils, the piece of ferrite embedded in it produces a change in inductance which is used to start and stop a timer. Details of the operation have been described in outline earlier [3].

The viscosity is related to the fall time t by the equation

$$\eta_P = \frac{t(1 - \rho_L/\rho_S)}{A[1 + 2\alpha(T - T_o)][1 - 0.666\beta(P - P_o)]} \tag{2}$$

where η_P is the viscosity at pressure P and ρ_L and ρ_S are the densities of liquid and sinker at temperature T and pressure P. T_o is the reference temperature, taken as 298.15 K, P_o is atmospheric pressure, α is the linear coefficient of thermal expansion, β is the bulk compressibility coefficient, and A is the viscometer constant.

A new sinker/tube combination was employed in this work and calibrated using *n*-octane, *n*-hexadecane, and Shell Vitrea No. 21 oil at temperatures from 298 to 373 K at atmospheric pressure, for which viscosities had been measured in suspended-level glass viscometers with an estimated accuracy of ±0.5%. The calibration curve was represented for $t^* < 175$ [where $t^* = t(1 - \rho_L/\rho_S)$] by the equation

$$A = 22.933 - 0.001811t^* \tag{3}$$

where t is in s and A has units of mPa^{-1}.

4. RESULTS

Measurements of the sinker fall times were made for toluene at 298.61 K at pressures up to 500 MPa, and at 323.14 and 323.29 K at pressures up to 435 MPa. The toluene was purchased from Aldrich Chemical Co. Ltd., Gillingham, U.K., with a stated minimum purity of 99.9 mol% and was used as received.

The viscosities given in Table I were calculated by the "ratio" method. Equation (2) was applied at pressure P and at atmospheric pressure to give the viscosity ratio for these pressures in terms of the corresponding ratio of the measured fall times, the ratio of the density terms, the ratio of A values,

Table I. The Viscosity of Liquid Toluene

T (K)	P (MPa)	t (s)	Density ($kg \cdot m^{-3}$)	Viscosity ($mPa \cdot s$)
298.61	0.1	14.338	861.7	0.5492
	51.8	20.958	896.3	0.799
	76.6	24.640	909.6	0.938
	101.2	28.505	921.3	1.084
	150.7	37.672	942.0	1.429
	200.4	48.535	959.6	1.838
	316.0	84.871	994.9	3.208
	413.0	129.905	1018.0	4.910
	492.4	183.064	1034.4	6.931
323.14	0.1	10.866	838.8	0.4211
	29.6	13.835	862.9	0.534
	50.4	16.035	876.9	0.618
	98.5	21.122	903.9	0.812
	149.2	27.671	926.7	1.061
	199.9	35.163	945.9	1.345
	340.4	62.922	987.7	2.398
	435.4	90.236	1010.2	3.435
323.29	0.1	10.705	838.7	0.4205
	48.9	15.698	875.9	0.614
	99.4	21.315	904.2	0.830
	190.5	33.719	942.5	1.307
	330.0	60.184	985.0	2.325
	425.0	87.528	1007.9	3.377

and a small pressure correction. From consideration of the fluctuations in pressure during a run, which were usually less than 0.5 MPa, and the temperature variations, which were at most 0.03 K, it is estimated that the precision of the viscosity results is ±2%. The accuracy is estimated to range from ±3% at the lowest pressures to ±5% at 500 MPa.

In Fig. 2, a comparison is made with the earlier falling-body viscosity measurements by Dymond et al. [3]. The agreement is well within the combined estimated uncertainties, except for the highest pressure at 298 K, where the earlier result is considered to be too high. This confirms the conclusion from Fig. 1 that Eq. (1) must be modified to account for the viscosity data at pressures from 250 to 500 MPa. In view of the superior accuracy of the measurements of Oliveira and Wakeham [2], Eq. (1) has been altered simply by the addition of a term in a higher power of pressure. At the common temperature of 323 K, the ratio of viscosity at elevated pressure to that at atmospheric pressure can be represented by

$$\eta_P/\eta_{0.1} = 1.0000 + 0.007824P + 1.489 \times 10^{-5}P^2 + 3.35 \times 10^{-19}P^7 \quad (4)$$

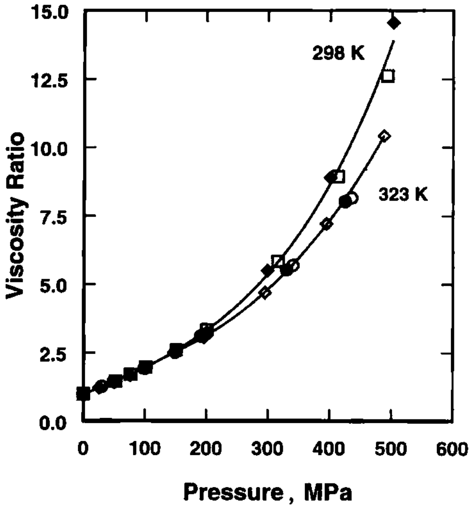

Fig. 2. Comparison of experimental high-pressure viscosity ratios $\eta_P/\eta_{0.1}$ for liquid toluene. Data of Dymond et al. [3]: ◆, 298 K; ◇, 323 K. Present results: □, 298.61 K; ○, 323.14 K; ●, 323.29 K.

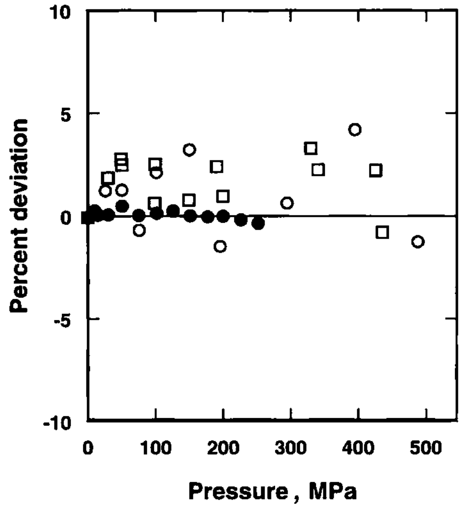

Fig. 3. Percentage deviation, $100(\eta_{\text{exp}} - \eta_{\text{calc}})/\eta_{\text{calc}}$, between experimental viscosity data (●, Oliveira and Wakeham [2]; ○, Dymond et al. [3]; □, present work) at 323 K and values calculated from Eq. (4).

where P is pressure/MPa. The first three terms are identical to those given by Oliveira and Wakeham [2] for $\eta_{0.1} = 0.4227$ mPa · s.

The deviations of the experimental data from this equation are shown in Fig. 3. The agreement is within the estimated experimental uncertainties over the whole pressure range.

5. CONCLUSION

An equation is given for the pressure dependence of the viscosity of toluene at 323 K up to 500 MPa which reproduces the vibrating-wire measurements up to 250 MPa within the stated uncertainty of ±0.5% and also gives good agreement with the falling-body measurements at higher pressures. It is recommended that this equation be considered for use of toluene as a reference material in the calibration and testing of high-pressure viscometers.

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