

Thermal Conductivity and Density of Toluene in the Temperature Range 273–373 K at Pressures up to 250 MPa

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New experimental data on the thermal conductivity and the density of liquid toluene are presented in the temperature range 0–100°C at pressures up to 250 MPa. The measurements of thermal conductivity were performed with a transient hot-wire apparatus on an absolute basis with an inaccuracy less than 1.0%. The density was measured with a high-pressure burette method with an uncertainty within 0.1%. The experimental results for both properties are represented satisfactorily by the Tait-type equations, as well as empirical polynomials, covering the entire ranges of temperature and pressure. Furthermore, it is found that simple relations exist between the temperature dependence of thermal conductivity and the thermal expansion coefficient, and also between the pressure dependence of thermal conductivity and the isothermal compressibility, as are suggested theoretically.

KEY WORDS: density; isothermal compressibility; Tait equation; thermal conductivity; thermal expansion coefficient; toluene; transient hot-wire method.

1. INTRODUCTION

The thermal conductivity of toluene is of scientific and technical importance, especially as one of the reference liquids in various relative measurements. Since toluene is chemically stable and its pure reagent is obtainable easily anywhere, it has been selected and recommended frequently as a suitable standard fluid in the calibration of thermal conductivity cells. However, the thermal conductivity values of toluene have not been definitive up to the present time. Although over 50 investigations have been reported in the literature, the discrepancy among the reported values of

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different authors is extremely large, even near room temperature at atmospheric pressure. There is a marked tendency for the recent data, obtained by advanced experimental techniques, to be lower than those obtained earlier. Among several sets of recommended values [1-4], the same tendency is observed. Furthermore, there is no reliable work on the effect of temperature and pressure upon the thermal conductivity of toluene, as well as its density in wide ranges of temperature and pressure. The purpose of this paper is to present new experimental data on the thermal conductivity and density of toluene, obtained by new measuring techniques, covering wide ranges of temperature and pressure.

2. EXPERIMENTAL

2.1. Thermal Conductivity Measurements

Recent rapid progress in the electronics field has made it possible for the transient hot-wire method to become a technique of high precision. Its advantages were described in a previous paper [5]. A thin, long, straight platinum wire, suspended vertically in a sample fluid, provides a line source of constant heat generation by an electric current. The wire also acts as a thermometer as a result of the change in its resistance. Using the Fourier equation, the temperature rise ΔT at the surface of the wire of radius r and at time θ is

$$\Delta T(r, \theta) = \frac{q}{4\pi\lambda} \ln\left(\frac{4a\theta}{r^2C}\right) \quad (1)$$

where q is the heat flux per unit length of the wire, λ is the thermal conductivity, a is the thermal diffusivity, and $\ln C$ is Euler's constant. From Eq. (1), a linear relation between ΔT and $\ln\theta$ should hold under an experimental condition, and λ can be determined from the slope of the straight line. That is, differentiating Eq. (1), λ should be represented by

$$\lambda = \frac{q}{4\pi} \Big/ \frac{d\Delta T}{d\ln\theta} \quad (2)$$

where q and $(d\Delta T/d\ln\theta)$ could be obtained precisely by electrical measurements.

In the present apparatus, the hot wire made of platinum (30 μm in diameter and 150 mm long) was placed tautly in an inner cylinder as shown in Fig. 1. Two potential leads were welded on the wire, the distance between the welding points being 107.05 mm. The resistance change of the wire due to its temperature rise was determined from the voltage between

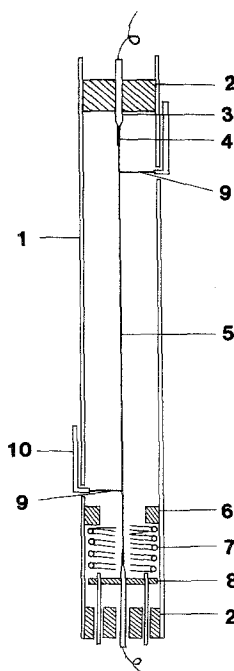


Fig. 1. Structure of the hot-wire cell: 1, inner cylinder; 2, insulator rings; 3, electrodes; 4, copper leads; 5, platinum hot wire; 6, spring supporter; 7, spring; 8, electrode supporting disk; 9, platinum potential leads; 10, copper leads.

the potential leads, which was measured with a precision double bridge. The current in the wire was also determined from the voltage across a standard resistor (100.000Ω). These electric measurements were controlled and recorded by a microcomputer (NEC PC-8001). The electric circuit for the present measurements is shown schematically in Fig. 2. The unbalanced voltage of the bridge, amplified 1800 times, and the potential across the standard resistor were read alternately by sample-holding circuits (S/H) at intervals of 0.1 s during 10 s from the start of one measurement. Then the signals in S/H were transmitted to the computer through an analog-digital converter (A/D) and a computer interface. The final results were displayed and printed after all the computations. In one measurement, 100 sets of data of ΔT and θ were recorded and the term $d\Delta T/d\ln\theta$ in Eq. (2) was determined from every 10 sets of data. Finally, a constant value of the term was selected at the optimum region where the linearity condition between ΔT and $\ln\theta$ was satisfied.

It was confirmed that the thermal conductivity under a given condition of temperature and pressure was reproducible within 0.1%, even when

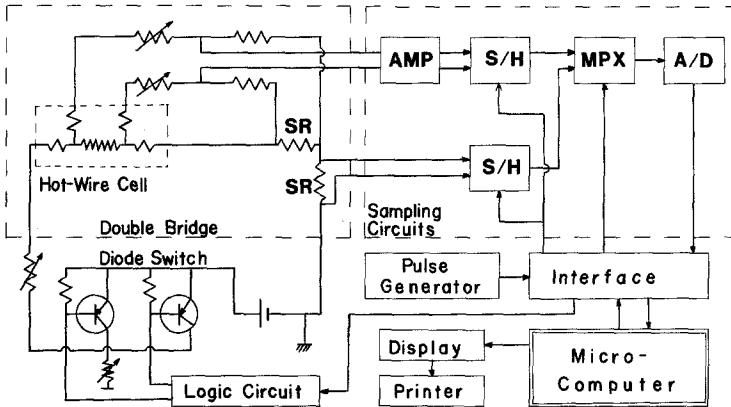


Fig. 2. Schematic diagram of the electrical circuits: AMP, amplifier, *S/H*, sample hold; MPX, multiplexer; *A/D*, analog-digital converter; SR, standard resistor (100 Ω).

the values of q and ΔT , or the current in the wire, were changed several-fold. The end effect of the wire was eliminated by use of the potential leads. The onset of convection was also detected by the gradual departure from the linear relation between ΔT and $\ln \theta$. Therefore, the results obtained should be free from errors due to natural convection. There was no evidence of heat transfer by radiation in the measurements, since the surface area of the wire and the temperature difference were relatively small. Therefore, no correction due to radiation was applied to the thermal conductivity results.

The hot-wire cell shown in Fig. 1 was mounted in a high pressure vessel made of Cr-Mo steel (SCM 3), which was placed vertically in an oil bath thermostatically controlled to within ± 10 mK. Temperature was determined with a standard resistance thermometer calibrated by the National Research Laboratory of Metrology (Japan). The pressure in the cell was generated through a mercury *U*-tube by an oil pump and measured by Bourdon tube gauges calibrated against a dead-weight gauge within an error of 0.1%. Based on the above discussion of errors and reproducibility of the measurements, the maximum uncertainty in the measured thermal conductivity was estimated to be less than 1.0%.

2.2. Density Measurements

The density of toluene was measured utilizing a new "high-pressure burette" method, whose details are described elsewhere [6, 7]. The volume change of a sample liquid (about 95 cm³) with pressure and temperature was measured by the displacement of mercury level in the high-pressure

burette connected to the sample vessel. The position of a magnetic float on the surface of the mercury column was detected outside the burette by a differential linear transformer. The uncertainty of the density measurements does not exceed 0.1% over the entire experimental range.

2.3. Material

Pure toluene used was supplied as "chromatographic grade" reagent by Wako Pure Chemical Industries, Ltd. The reported purity was better than 99.5%. Before each measurement, it was purified twice by fractional distillation. Thus, small amount of impurities would not have affected the present results.

3. EXPERIMENTAL RESULTS

3.1. Thermal Conductivity of Toluene

The experimental results for the thermal conductivity of toluene are presented in Table I. Although measurements have been performed 6 to 10 times under the same experimental conditions, only the mean values of each run are listed in the table. The thermal conductivity of toluene at atmospheric pressure can be expressed by a linear function of temperature as follows:

$$\lambda_0 = 0.2166 - 2.88 \times 10^{-4}T \quad (3)$$

where λ_0 is in $\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ and T is in K. This equation is valid in the temperature range from 273 to 373 K. The deviations of a number of literature values from Eq. (3) were shown in a previous paper [5].

The variation of thermal conductivity of toluene with temperature and pressure was found to be relatively simple over the entire range in the measurements. The thermal conductivity at a constant pressure decreases almost linearly with increasing temperature. That is, the temperature coefficient $(\partial\lambda/\partial T)_P$ is always negative, nearly constant independent of temperature, and increases gradually with increasing pressure. The pressure coefficient $(\partial\lambda/\partial P)_T$ is always positive and becomes larger with increasing temperature. Each isotherm could be fitted by the following polynomial equation:

$$\lambda = A_0 + A_1P + A_2P^2 \quad (4)$$

where λ is in $\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ and P is pressure in MPa. The coefficients A_0, A_1, A_2 are listed in Table II, where the average and maximum deviations of the experimental data from Eq. (4) are also given.

Table I. Thermal Conductivity and Density of Toluene^a

<i>P</i>	ρ	λ	<i>P</i>	ρ	λ
<i>T</i> = 0°C (273.15 K)			125.0	922.2	0.1611
0.10	885.6	0.1379	137.5	927.4	0.1630
24.99	901.4	0.1445	150.0	932.4	0.1660
49.95	915.2	0.1507	162.5	937.2	0.1688
74.98	927.4	0.1568	175.0	941.8	0.1714
100.0	938.4	0.1625	200.0	950.5	0.1758
112.5	943.5	0.1643	212.5	954.7	0.1780
125.0	948.4	0.1673	225.0	958.7	0.1791
150.0	957.6	0.1713	240.0	963.3	0.1817
162.5	962.0	0.1740	<i>T</i> = 50°C (323.15 K)		
175.0	966.2	0.1754	0.10	838.9	0.1235
200.0	974.2	0.1800	24.89	859.2	0.1313
214.9	978.7	0.1821	34.57	866.2	0.1349
225.0	981.7	0.1830	50.05	876.5	0.1391
<i>T</i> = 25°C (298.15 K)			74.98	891.3	0.1464
0.10	862.3	0.1306	99.98	904.2	0.1524
12.51	871.7	0.1345	125.0	915.6	0.1585
24.99	880.2	0.1388	150.0	925.9	0.1636
37.47	888.2	0.1413	175.0	935.6	0.1689
49.95	895.7	0.1453	200.0	945.1	0.1734
62.50	902.7	0.1484	224.9	954.9	0.1773
74.98	909.2	0.1515	<i>T</i> = 75°C (348.15 K)		
87.46	915.4	0.1551	0.10	815.0	0.1164
100.0	921.2	0.1580	24.99	838.3	0.1262
112.5	926.7	0.1598	49.95	857.6	0.1330
125.0	931.9	0.1630	74.98	873.9	0.1403
137.5	936.9	0.1652	100.0	887.8	0.1469
150.0	941.7	0.1683	112.5	894.0	0.1506
162.5	946.3	0.1704	125.0	899.8	0.1530
175.0	950.8	0.1729	150.0	910.7	0.1587
187.5	955.3	0.1746	162.5	915.8	0.1616
200.0	959.8	0.1769	175.0	920.8	0.1644
214.9	965.1	0.1793	188.2	926.1	0.1674
225.0	968.8	0.1810	200.0	930.9	0.1690
240.0	974.3	0.1833	212.5	936.1	0.1716
<i>T</i> = 40°C (313.15 K)			225.0	941.6	0.1738
0.10	848.4	0.1263	237.6	947.3	0.1764
12.51	858.6	0.1305	249.9	953.3	0.1790
24.99	867.9	0.1347	<i>T</i> = 100°C (373.15 K)		
37.47	876.4	0.1385	0.10	790.1	0.1096
49.95	884.3	0.1423	12.51	804.3	0.1137
62.50	891.6	0.1455	24.99	816.8	0.1188
74.98	898.4	0.1488	37.47	828.2	0.1225
87.46	904.9	0.1519	49.95	838.6	0.1264
100.0	910.9	0.1546	62.50	848.1	0.1304
112.5	916.7	0.1576			

Table I. (Continued)

P	ρ	λ
74.98	856.7	0.1339
100.0	871.9	0.1412
125.0	884.8	0.1480
149.5	896.0	0.1545
175.0	906.8	0.1605
200.0	917.4	0.1654
225.0	928.5	0.1705
237.6	934.6	0.1726
250.0	941.0	0.1754

^a P , pressure in MPa; ρ , density in $\text{kg} \cdot \text{m}^{-3}$; λ , thermal conductivity in $\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$.

Table II. Coefficients of Polynomial Equation (4) for Thermal Conductivity of Toluene

Temp. (K)	A_0	$10^3 A_1$	$10^6 A_2$	Av. dev. (%)	Max. dev. (%)
273.15	0.1378	0.275	-0.328	0.14	0.29
298.15	0.1309	0.300	-0.349	0.18	0.40
313.15	0.1267	0.321	-0.383	0.18	0.35
323.15	0.1236	0.330	-0.408	0.12	0.27
348.15	0.1173	0.330	-0.342	0.23	0.75
373.15	0.1097	0.353	-0.366	0.15	0.41

The results of a few investigations on the thermal conductivity of toluene under high pressure are available in the literature [8–11]. In Fig. 3, the literature values are compared with the present measurements. Since the early values at atmospheric pressure were considerably higher than those in the present work, the relative thermal conductivity, λ_p/λ_0 , where subscripts p and 0 denote high pressure and atmospheric pressure, respectively, is plotted in Fig. 4. It is found that the present results agree well with those of Rastorguev and Pugach [10] on the effect of pressure on thermal conductivity at both 40 and 100°C. Although Bridgman's data [8] show a small pressure effect, the absolute values are extremely high. On the other hand, Kandiyoti et al. [11] reported a large pressure effect different from that of the present work. The data of Akhundov and Gasanova [9] at 107°C are reasonable although the pressure range is somewhat small.

3.2. Density of Toluene

The density of toluene was measured in the temperature range from 10 to 100°C and at pressures up to 205 MPa. It was found that the density at a constant temperature increases with increasing pressure, that is, $(\partial\rho/\partial P)_T$

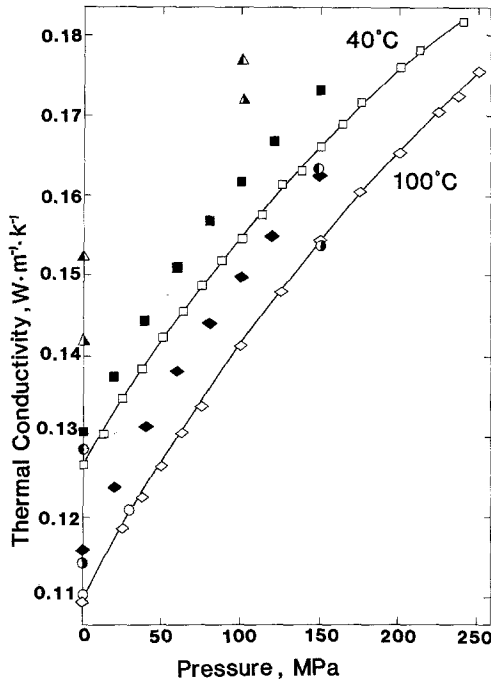


Fig. 3. Comparison of thermal conductivity of toluene with those in earlier works: \square 40°C, \diamond 100°C, this work; \triangle 30°C, Δ 75°C of Bridgman [8]; \circ 107°C of Akhundov and Gasanova [9]; \blacksquare 40°C, \blacklozenge 100°C of Rastorguev and Pugach [10]; \bullet 30.8°C, \circ 91.5°C of Kandiyoti et al. [11].

is always positive. It also increases with temperature, and decreases with pressure. The temperature coefficient $(\partial\rho/\partial T)_P$ is always negative, and its change becomes smaller with increasing pressure, which is well known for normal liquids.

The raw data² (114 sets) were used to obtain some empirical equations, after confirmation of the data's accuracy by means of the Hudleston equation. Each isotherm was fitted to the following polynomial equation:

$$\rho = B_0 + B_1P + B_2P^2 + B_3P^3 \quad (5)$$

where ρ is in $\text{kg} \cdot \text{m}^{-3}$ and P is in MPa. The coefficients are listed in Table III, where the deviations of the raw data from Eq. (5) are also given. This equation was used to compare the present results with the values reported by Mopsik [12], who measured the density of toluene at temperatures below

²The raw data on the density of toluene are available from T. Makita by direct request.

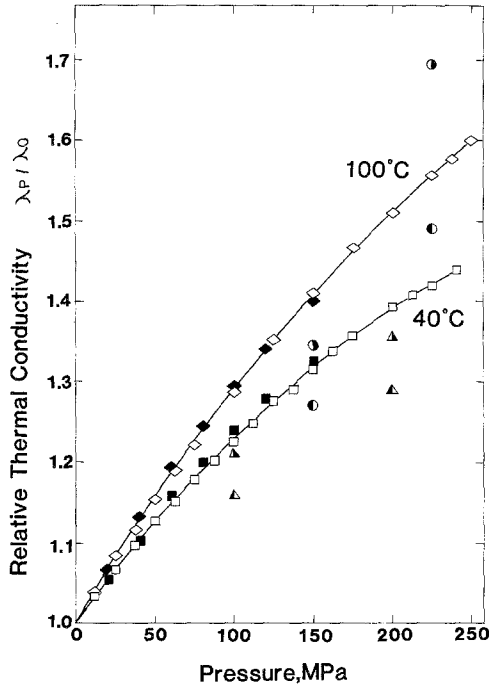


Fig. 4. Comparison of relative thermal conductivity of toluene with those in earlier works. Key is the same as in Fig. 3.

298.15 K and at pressures up to 200 MPa. It is found that Mopsik's data at 298.15 K are consistent with the calculated values from Eq. (5) within a maximum deviation of 0.07%.

The Tait equation has been used to represent the high pressure isotherms of liquids:

$$\frac{\rho - \rho_0}{\rho} = C \ln\left(\frac{D + P}{D + P_0}\right) \tag{6}$$

Table III. Coefficients of Polynomial Equation (5) for Density of Toluene

Temp. (K)	B_0	B_1	$10^3 B_2$	$10^6 B_3$	Av. dev. (%)	Max. dev (%)
283.15	876.70	0.69695	- 1.7951	3.013	0.02	0.04
298.15	862.52	0.75843	- 2.0686	3.540	0.02	0.04
323.15	839.16	0.86702	- 2.6403	4.775	0.02	0.05
348.15	815.45	0.99449	- 3.3382	6.266	0.04	0.08
373.15	790.65	1.1438	- 4.0761	7.626	0.05	0.10

Table IV. Coefficients of Tait Equation (7) for Density of Toluene

Temp. (K)	ρ_0 ($\text{kg} \cdot \text{m}^{-3}$)	C	D (MPa)	Av. dev. (%)	Max. dev. (%)
273.15	885.56	0.09143	117.2		
283.15	876.52	0.09143	109.8	0.02	0.04
298.15	862.34	0.09143	99.1	0.02	0.06
313.15	848.42	0.09143	89.2		
323.15	838.89	0.09143	83.0	0.01	0.03
348.15	814.95	0.09143	68.7	0.03	0.04
373.15	790.09	0.09143	56.4	0.02	0.05

where ρ_0 is the density at a reference pressure P_0 , often taken at atmospheric pressure, and C and D are empirical constants. Although the optimum values of C and D could be determined for each isotherm from experimental data, it is more convenient to treat C as a constant and D as a function of temperature. Furthermore, in order to calculate the density under an arbitrary condition, the rearranged form is as follows:

$$\rho = \rho_0(T) / \left[1 - C \ln \frac{D(T) + P}{D(T) + P_0} \right] \quad (7)$$

where P_0 is taken as 0.1 MPa. The values of C and D , as well as ρ_0 , are listed in Table IV, in which the deviations of the raw data from this equation are also shown. The values at 273.15 and 313.15 K are determined by extrapolation and interpolation, respectively. The values of ρ_0 and D could be also represented as a function of temperature by

$$\rho_0(T) = 1103.06 - 0.68074T - 0.4229 \times 10^{-3}T^2 \quad (8)$$

$$D(T) = 440.47 - 1.604T + 1.539 \times 10^{-3}T^2 \quad (9)$$

where $\rho_0(T)$ is in $\text{kg} \cdot \text{m}^{-3}$, $D(T)$ is in MPa, and T is in K. The density values in Table I were calculated by Eq. (7) using the values of P_0 , C and $D(T)$ in Table IV.

4. DISCUSSION

4.1. Density Dependence of Thermal Conductivity of Toluene

The effect of pressure on many physical properties of fluids could be interpreted in terms of density (13). That is, the changes in properties with pressure have been frequently expressed by simple functions of density,

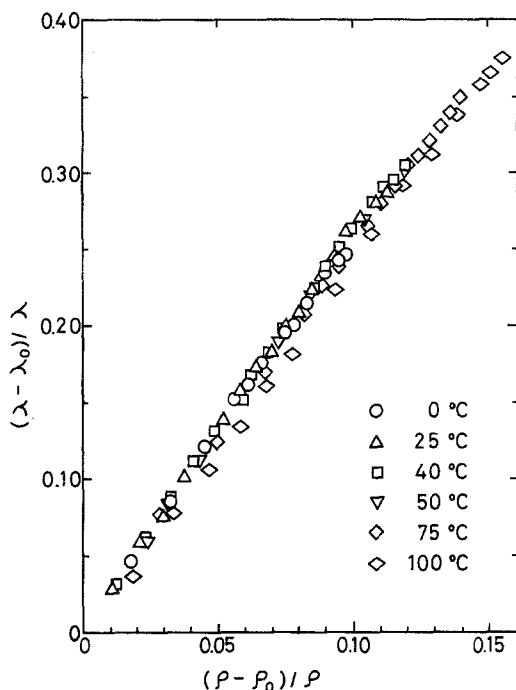


Fig. 5. Plots of $(\lambda - \lambda_0)/\lambda$ vs. $(\rho - \rho_0)/\rho$ of toluene.

instead of pressure. In view of this, the present data on thermal conductivity of toluene have been examined by several relations to the density. One of the interesting results is shown in Fig. 5, where $(\lambda - \lambda_0)/\lambda$ is plotted against $(\rho - \rho_0)/\rho$ over the entire range of the present experimental conditions. It is found that a nearly linear relation exists between these quantities, and the difference among isotherms almost disappears. Namely, all the values fall on a straight line within 10%. This scatter is reasonable because of the existence of the difference term $(\lambda - \lambda_0)$ and the present experimental uncertainty. The relation in Fig. 5 suggests that the pressure effect on thermal conductivity could be represented in the same way as that of the density.

4.2. The Tait-Type Equation for Thermal Conductivity

As described in Section 3.2, the variation of density with temperature and pressure is well represented by the Tait equation (6). Therefore, the present values of the thermal conductivity of liquid toluene were used to

Table V. Coefficients of Equation (10) for Thermal Conductivity of Toluene

Temp. (K)	<i>E</i>	<i>F</i> (MPa)	Av. dev. (%)	Max. dev. (%)
273.15	0.1968	87.8	0.18	0.39
298.15	0.2043	76.6	0.20	0.59
313.15	0.2058	68.8	0.19	0.53
323.15	0.2163	71.6	0.22	0.51
348.15	0.2149	61.0	0.17	0.45
373.15	0.2427	66.4	0.31	0.67

examine the same formula as follows:

$$\frac{\lambda - \lambda_0}{\lambda} = E \ln \left(\frac{F + P}{F + P_0} \right) \quad (10)$$

where *E* and *F* are empirical constants. It is found that each isotherm can be represented by this formula with the errors comparable to Eq. (4). The coefficients *E* and *F* are given in Table V, in which the deviations of experimental data from Eq. (10) are also shown.

The rearranged form of Eq. (10), which should be more convenient to use, is also derived in the same way as in the case of Eq. (7) for the density, as follows:

$$\lambda = \lambda_0(T) / \left[1 - E \ln \frac{F(T) + P}{F(T) + P_0} \right] \quad (11)$$

The values of $\lambda_0(T)$, *E*, and *F*(*T*) determined are listed in Table VI, as well as the deviations of the raw data. Although the deviations are somewhat larger than those in Table V, the maximum deviation is within the experimental uncertainty. The values of $\lambda_0(T)$ are calculated from Eq. (3), and

Table VI. Coefficients of Equation (11) for Thermal Conductivity of Toluene

Temp. (K)	λ_0 (W · m ⁻¹ · K ⁻¹)	<i>E</i>	<i>F</i> (MPa)	Av. dev. (%)	Max. dev. (%)
273.15	0.1379	0.2138	99.9	0.21	0.65
298.15	0.1306	0.2138	82.5	0.23	0.54
313.15	0.1263	0.2138	73.3	0.19	0.75
323.15	0.1235	0.2138	70.2	0.21	0.54
348.15	0.1164	0.2138	60.4	0.17	0.47
373.15	0.1096	0.2138	53.0	0.58	0.94

$F(T)$ is also given by the relation

$$F(T) = 510.72 - 2.269T + 2.795 \times 10^{-3}T^2 \quad (12)$$

where $F(T)$ is in MPa and T is in K.

In comparing Eq. (10) for the thermal conductivity with Eq. (6) for the density, it may be seen that the ratio of the coefficients E to C depends mainly on the slope of the line in Fig. 5, and the coefficient F is expected to be identical with D . However, the values of F are not the same as those of D , as seen in Tables IV, V, or VI, although the behavior of F and D with temperature is qualitatively similar. Therefore, the pressure effects on the thermal conductivity are not completely identical with those on the density.

4.3. Temperature and Pressure Dependences

Another relation between the thermal conductivity and the PVT relations was derived theoretically by Horrocks and McLaughlin [14] and Kamel and McLaughlin [15], assuming that heat conduction in liquids is performed by the vibration mechanism in a lattice structure. Their results in the first approximation are written as:

$$\frac{1}{\lambda} \left(\frac{\partial \lambda}{\partial T} \right)_P = -\alpha \left[\frac{1}{3} - \left(\frac{\partial \ln \nu}{\partial \ln v} \right)_P \right] \quad (13)$$

$$\frac{1}{\lambda} \left(\frac{\partial \lambda}{\partial P} \right)_T = \beta \left[\frac{1}{3} - \left(\frac{\partial \ln \nu}{\partial \ln v} \right)_T \right] \quad (14)$$

where $(\partial \ln \nu / \partial \ln v)$ is called Gruneisen's constant, α is coefficient of thermal expansion, and β is the isothermal compressibility. These equations show that the important factor in controlling the temperature dependence of thermal conductivity is the coefficient of thermal expansion and that the pressure dependence depends on the isothermal compressibility. Using the present experimental data, the temperature and the pressure coefficients of λ are plotted against α and β in Figs. 6 and 7, respectively.

As seen in Fig. 6, $-(1/\lambda)(\partial \lambda / \partial T)_P$ is nearly linear in α and decreases with increasing pressure, since α decreases with pressure. The curve (or line) does not pass through the origin, and therefore, the sign of the temperature coefficient is expected to reverse at a higher pressure, to which the present work could not attain. On the other hand, the pressure coefficient of λ is also linear in β and decreases with decreasing temperature and increasing pressure, as seen in Fig. 7. As the curve should pass through the origin, the pressure coefficient of λ is expected to be always positive.

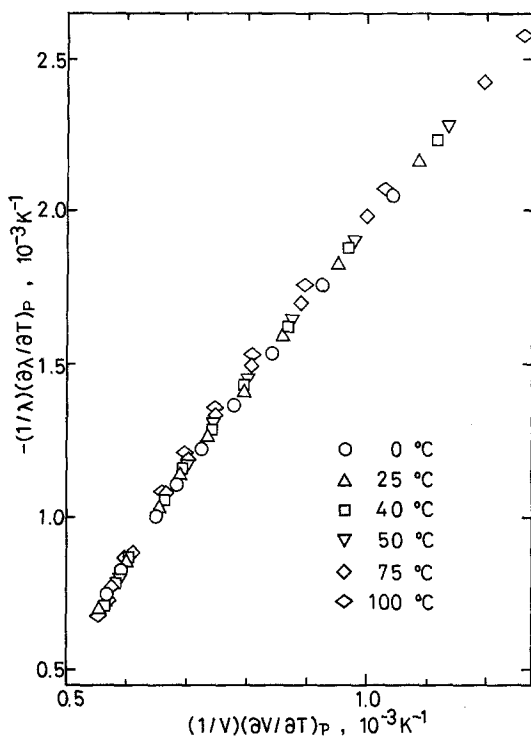


Fig. 6. Plots of the temperature coefficient of thermal conductivity vs. thermal expansion coefficient of toluene.

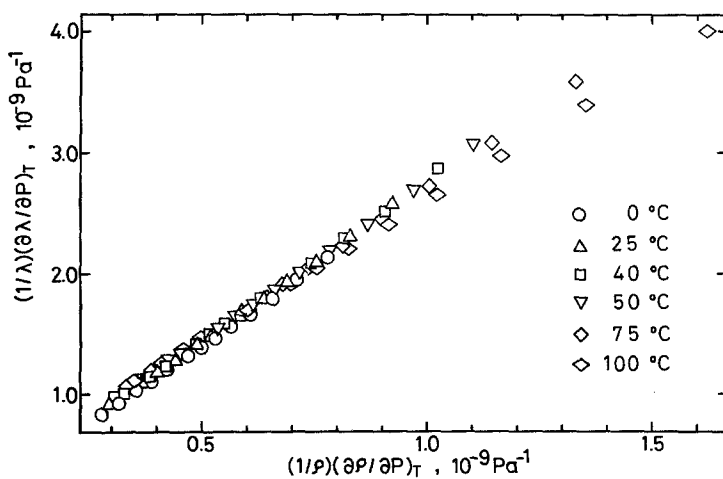


Fig. 7. Plots of the pressure coefficients of thermal conductivity vs. isothermal compressibility of toluene.

5. CONCLUSIONS

New experimental data on the thermal conductivity and the density of liquid toluene have been presented, covering extensive ranges of temperature and pressure. The numerical values of both properties are expressed by empirical equations over the entire ranges of this work.

It is essential to determine standard values of thermophysical properties of some selected reference materials on an international basis. Such values should be decided by the consensus of many investigators in the field, based on the agreement of the data obtained by various investigators utilizing different methods and instruments. The present work represents an attempt in this direction by measuring thermal conductivity utilizing a computer-aided transient hot-wire technique.

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