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Thermal Conductivity and Viscosity of Biphenyl and the Terphenyls

W. H. HEDLEY,[1] M. V. MILNES,[2] and W. H. YANKO
Monsanto Research Corp., Dayton, Ohio

The thermal conductivities and viscosities of liquid biphenyl and the three terphenyls were measured over a range of temperatures using the hot-wire and capillary viscometer methods. These properties were also calculated as a function of temperature in the vapor phase to provide data for calculation of the performance of these compounds in heat transfer and other applications. Equations for the thermal conductivity of vapors at low pressure were formulated directly from a general equation fitted to the tabulated data of Bromley and Wilke.

THE LIQUID AND VAPOR VISCOSITY and thermal conductivity values of biphenyl and the three isomeric terphenyls were determined to provide data for use in heat transfer calculations and engineering design of heat transfer equipment. Because of their good radiation and thermal stability, the polyphenyls are useful as nuclear reactor coolants and in other high temperature heat transfer applications.

The purity (*12, 13*) of the samples used in the viscosity and thermal conductivity measurements was:

Biphenyl	99.95%
o-Terphenyl	99.9%
m-Terphenyl	99.5%
p-Terphenyl	99.99%

The samples of biphenyl, *o*-, and *m*-terphenyl were prepared by an initial vacuum fractional distillation of commercial materials, and then these batches were refined further by repeated fractional freezing. The *p*-terphenyl was purified by recrystallization from benzene. The purity of these materials was checked by vapor phase chromatography with a flame ionization detector.

All experimental temperatures quoted were measured with platinum resistance thermometers which had been calibrated by the National Bureau of Standards.

VISCOSITY

Liquid Viscosity. The viscosities of liquid biphenyl, *o*-terphenyl, and *m*-terphenyl were measured on 1-ml. samples using a Cannon-Manning semimicroviscometer; with *p*-terphenyl a Cannon-Ubbelohde semimicroviscometer was used. Viscosity measurements were made using constant temperature baths controlled to ±0.006° C. at 98.9° C. and ±0.02° C. at higher temperatures. The viscometer calibration runs with water and the measurements on the polyphenyls were done in accordance with ASTM standard D 445-52T, and the observed kinematic viscosities are listed in Table I.

These data are fitted to the following Andrade equations:

Biphenyl
$$\log \eta = -1.7080 + 634.8/T \text{ for } 370 \leq T \leq 480^\circ \text{ K.} \tag{1}$$

o-Terphenyl
$$\log \eta = -2.4604 + 1153.6/T \text{ for } 370 \leq T \leq 560^\circ \text{ K.} \tag{2}$$

m-Terphenyl
$$\log \eta = -2.0444 + 977.0/T \text{ for } 370 \leq T \leq 590^\circ \text{ K.} \tag{3}$$

p-Terphenyl
$$\log \eta = -1.6965 + 791.0/T \text{ for } 490 \leq T \leq 620^\circ \text{ K.} \tag{4}$$

The kinematic viscosity values can be converted to dynamic viscosities in centipoises by multiplying by the following liquid densities, ρ_L (*5, 11*):

Biphenyl
$$\rho_L = 1.0145 - 0.0328 \times 10^{-2} t - 0.0138 \times 10^{-4} t^2 \text{ for } 100 \leq t \leq 450^\circ \text{ C.} \tag{5}$$

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[1] Present address, Monsanto Enviro-Chem Systems, Inc., Box 8, Station B, Dayton, Ohio 45407

[2] Present address, Aerospace Systems Division, Bendix Corp., 3300 Plymouth Road, Ann Arbor, Mich. 48107

Table I. Viscosity of Liquid Biphenyl and the Terphenyls

Compound	Temp., ° C.	Viscosity, Centistokes
Biphenyl	98.9	0.996
	204.4	0.418
o-Terphenyl	98.9	4.564
	204.4	0.794
	287.8	0.429
m-Terphenyl	98.9	3.955
	204.4	0.917
	315.6	0.435
p-Terphenyl	220.6	0.818
	220.9	0.799
	269.0	0.571
	269.1	0.573
	320.2	0.434
	320.2	0.436
	340.4	0.393

o-Terphenyl $\rho_L = 1.087 - 0.068 \times 10^{-2}t - 0.0030 \times 10^{-4}t^2$ for $100 \leq t \leq 350°$ C. (6)

m-Terphenyl $\rho_L = 1.101 - 0.065 \times 10^{-2}t - 0.0025 \times 10^{-4}t^2$ for $100 \leq t \leq 400°$ C. (7)

p-Terphenyl $\rho_L = 1.089 - 0.052 \times 10^{-2}t - 0.0045 \times 10^{-4}t^2$ for $225 \leq t \leq 400°$ C. (8)

The dynamic viscosities of the polyphenyls based upon our data are compared with values from the literature in Tables II, III, IV, and V. The agreement between the new values and those in the literature is good for biphenyl and *p*-terphenyl but only fair for *o*- and *m*-terphenyl.

The viscosities of biphenyl–*o*-terphenyl, *m*-terphenyl–*o*-terphenyl, and *m*-terphenyl–biphenyl mixtures were measured at 98.89° C. (Table VI).

Table II. Viscosity Data for Biphenyl

	Dynamic Viscosity, Cp.				
Temp., ° C.	This work	Anderson (*2*)	Gercke (*14*)	McEwen (*20*)	Stone *et al.* (*24*)
100	0.95	0.94		1.00	
125	0.73	0.72		0.75	0.71
150	0.57	0.57	0.57	0.58	0.55
175	0.46	0.46	0.46	0.47	0.44
200	0.38	0.38	0.37	0.38	0.37
225	0.32	0.32	0.31	0.32	0.31

Table III. Viscosity Data for o-Terphenyl

	Dynamic Viscosity, Cp.				
Temp., ° C.	This work	Andrews and Ubbelohde (*3*)	Bowring *et al.* (*4*)	McEwen (*20*)	McLaughlin and Ubbelohde (*21*)
100	4.34	4.38	4.35	4.8	
125	2.73	2.24	2.29	2.6	
150	1.80		1.53	1.8	
175	1.24		1.08	1.3	
200	0.89		0.80	0.92	0.84
225	0.66		0.62	0.74	0.66
250	0.50		0.50	0.59	0.53
275	0.39		0.42	0.49	0.44
300	0.30		0.35	0.42	0.36

Table IV. Viscosity Data for *m*-Terphenyl

	Dynamic Viscosity, Cp.			
Temp., ° C.	This work	Andrews and Ubbelohde (*3*)	Bowring *et al.* (*4*)	Stone *et al* (*24*)
100	3.87	3.62		
125	2.61	2.30	2.98	2.15
150	1.83	1.55	1.66	1.55
175	1.34		1.21	1.16
200	1.01		0.91	0.89
225	0.78		0.72	0.70
250	0.61		0.58	0.57
275	0.49		0.49	0.47
300	0.40		0.41	0.39
325	0.34		0.35	0.33

Table V. Viscosity Data for *p*-Terphenyl

	Dynamic Viscosity, Cp.				
Temp., ° C.	This work	Andrews and Ubbelohde (*3*)	Bowring *et al.* (*4*)	Burns *et al.* (*9*)	Stone *et al.* (*24*)
225	0.74	0.74	0.74	0.75	
250	0.61	0.61	0.61	0.62	0.60
275	0.51	0.52	0.52	0.52	0.50
300	0.43	0.45	0.45	0.45	0.43
325	0.37	0.39	0.39	0.39	0.37
350	0.32		0.35	0.35	0.32

In all cases the measured viscosity was lower than that which would be calculated from the pure components assuming ideal behavior. As shown in Table VI, the deviation from ideality was very significant for the biphenyl–*o*-terphenyl and *m*-terphenyl–biphenyl mixtures.

Low-Pressure Vapor Viscosity. The viscosities of polyphenyl vapors at low pressures ($\mu°$) were calculated using our simple but valuable modification of the method of Bromley and Wilke (*8*). These calculations normally require that values of the viscosity temperature function, $f\left(\frac{kT}{\varepsilon}\right)$, be obtained from tables in which it is listed as a function of $\frac{kT}{\varepsilon}$. The values of $\frac{kT}{\varepsilon}$ are customarily

Table VI. Viscosity of Terphenyl-Biphenyl Solutions at 98.89° C.

Composition, Mole %			
Biphenyl	*o*-Terphenyl	Kinematic Viscosity, Cs.	% Deviation
0.75	99.25	4.537	−0.3
1.48	98.52	4.456	−1.5
22.99	77.01	3.059	−18.5
49.26	50.74	2.065	−26.6
74.93	25.07	1.458	−23.1
m-Terphenyl	*o*-Terphenyl		
50.01	49.99	4.225	−0.9
m-Terphenyl	Biphenyl		
50.74	49.26	2.046	−18.0

calculated from the reduced temperature, using Equation 9 (*8*).

$$\frac{kT}{\varepsilon} = 1.33\ T_r \tag{9}$$

During this work it was found that the viscosity temperature function values could be fitted accurately to a simple equation (Equation 10) over a wide temperature range. Using Equation 10 the tabulated values can be predicted with a maximum error of 0.5% over the range of reduced temperature from 0.34 to 0.67 and with a maximum error of 2.0% over the range of T_r from 0.28 to 1.30.

$$f\left(\frac{kT}{\varepsilon}\right) = 0.8335\ T_r \tag{10}$$

The advantage of using Equation 10 in preference to the tabulated values is that the Bromley-Wilke equation can be rewritten in the form of Equation 11.

$$\mu^\circ = \frac{0.002776\ (M)^{1/2}\ T}{(V_c)^{2/3}\ (T_c)^{1/2}} \tag{11}$$

This equation requires only the substitution of known or easily calculated physical properties of molecules for the derivation of simple viscosity equations which are much more convenient for use in computer calculation of viscosity values.

Equation 11 was used to calculate the following low-pressure viscosity equations:

Biphenyl $\quad \mu^\circ = 1.83 \times 10^{-5}t + 5.00 \times 10^{-3}$ (12)

o-Terphenyl $\quad \mu^\circ = 1.71 \times 10^{-5}t + 4.67 \times 10^{-3}$ (13)

m-Terphenyl $\quad \mu^\circ = 1.65 \times 10^{-5}t + 4.51 \times 10^{-3}$ (14)

p-Terphenyl $\quad \mu^\circ = 1.66 \times 10^{-5}t + 4.53 \times 10^{-3}$ (15)

These equations are also valid for the saturated vapors up to temperatures of 450° C. for biphenyl, 520° C. for *o*-terphenyl, and 540° C. for *m*- and *p*-terphenyl.

The critical properties used in calculating Equations 12 to 15 are given in Table VII.

Saturated Vapor Viscosity. The saturated vapor viscosities (μ_s) of the polyphenyls were calculated using the method of Grunberg and Nissan (*15*). The ratios of the saturated vapor viscosities to those at low pressure for *m*- and *p*-terphenyl were found equal to 1.00 at temperatures up to 538° C. The ratios of the saturated vapor viscosities to those at low pressure for biphenyl and *o*-terphenyl are listed in Table VIII.

Table VIII. Ratio of Saturated Vapor Viscosity to Low-Pressure Vapor Viscosity

Biphenyl		*o*-Terphenyl	
Temp., ° C.	μ_s/μ°	Temp., ° C.	μ_s/μ°
449	1.00	521	1.00
460	1.22	527	1.13
471	1.36	532	1.16
482	1.49	538	1.19
493	1.69		

THERMAL CONDUCTIVITY

Liquid Thermal Conductivity. The thermal conductivities of the liquid polyphenyls were determined using a hot-wire type thermal conductivity cell (*16*). The hot-wire cell was constructed with a 10.1-cm. length of 1-mil diameter platinum wire suspended along the axis of an aluminum sample cup having a radius of 0.80 cm. The thermal conductivity of the liquid is calculated using Equation 16. Since this equation

$$k_L = \frac{Q \ln a/b}{2\pi L\ (t_a - t_b)} \tag{16}$$

is dependent only on measurable quantities and constants of the cell, the hot-wire method is an absolute method.

Table VII. Critical Properties

	T_c, ° K.	V_c, Cc.-G. Mole^{-1}	Ref.
Biphenyl	772.3	558.3	(*11*)
o-Terphenyl	891	752	(*18*)
m-Terphenyl	925	767	(*18*)
p-Terphenyl	926	762	(*18*)

While making measurements the cell was immersed in a constant temperature bath (Hallikainen Catalog No. 1139-C) which controlled the temperature to ±0.002° C. A Mueller bridge (Leeds & Northrup, Type G-2, Model 8069B) was used for the measurements of cell resistance with biphenyl, *o*-terphenyl, and *m*-terphenyl. A 6-dial potentiometer (Leeds & Northrup, Model 7556-A1) was used for the *p*-terphenyl measurements. Cell current was determined by measuring the voltage across a thermostated standard 1-ohm resistor connected in series with the cell. Values of Q were calculated using the cell resistance and current readings; they were chosen to keep t_a-t_b well below 0.2° C. to prevent convection. Values of t_b were measured with a platinum resistance thermometer (Leeds & Northrup, Model 8163). Values of t_a were obtained by using the hot wire as a platinum resistance thermometer and calculating its temperature using resistance measurements at two or more current readings, and t_b equal to the wire temperature at zero current.

To verify that the apparatus was performing properly, initial measurements made using this cell were compared with those of other investigators on glycerol and toluene at 80° C. The agreement as shown in Table IX is good.

Table IX. Thermal Conductivities of Glycerol and Toluene

Compound	Thermal Conductivity at 80° C. $10^6 \times k$, Cal. – Cm.$^{-1}$ – Sec.$^{-1}$ – °C.$^{-1}$
Glycerol	
Mason (*22*)	743
McCready (*19*)	708
This work	715
Toluene	
McCready (*19*)	299
Riedel (*23*)	282
Ziebland and Burton (*26*)	280
This work	280

The values obtained for polyphenyls with our apparatus are listed in Table X.

The data for the liquid thermal conductivity of biphenyl and the three terphenyls were fitted to the following least-squares equations:

Biphenyl $10^6 k_L = 348 - 0.286t$ for $80 \leq t \leq 210°$ C. (17)

o-Terphenyl $10^6 k_L = 338 - 0.239t$ for $100 \leq t \leq 210°$ C. (18)

m-Terphenyl $10^6 k_L = 341 - 0.196t$ for $90 \leq t \leq 210°$ C. (19)

p-Terphenyl $10^6 k_L = 344 - 0.094t$ for $210 \leq t \leq 260°$ C. (20)

These data are compared with literature data on thermal conductivity of liquid polyphenyls in Figures 1, 2, 3, and 4. Agreement between the literature values and those from this work is good for biphenyl and o-terphenyl, but only fair for m- and p-terphenyl.

Low-Pressure Vapor Thermal Conductivity. The low-pressure vapor thermal conductivities (extrapolated to zero pressure) of biphenyl and the terphenyls were calculated using the method of Bromley (*7*). Specific heat data for these calculations were obtained by the method of Anderson, Beyer, and Watson (*1*).

Table X. Thermal Conductivity of Liquid Biphenyl and Terphenyls

Biphenyl		o-Terphenyl		m-Terphenyl		p-Terphenyl	
t, ° C.	k_L[a]	t, ° C.	k_L[a]	t, ° C.	k_L[a]	t, ° C.	k_L[a]
84.5	322	102.7	313	99.8	316	213.3	324
99.7	323	102.7	313	102.5	325	253.6	321
151.2	303	197.9	289	151.2	309		
153.7	295	203.4	290	153.8	316		
203.2	290			197.8	300		
205.7	291			203.3	304		
				203.3	299		
				205.7	300		

[a] $10^6 \times$ cal.–cm.$^{-1}$–sec.$^{-1}$–° C.$^{-1}$

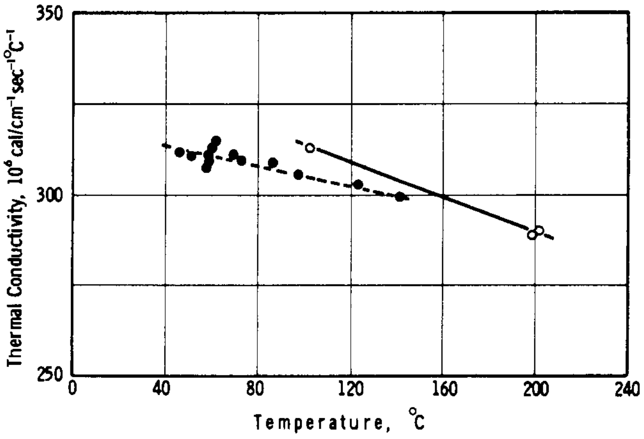

Figure 2. Thermal conductivity of liquid o-terphenyl

○ This work
● Horrocks and MacLaughlin (*17*)

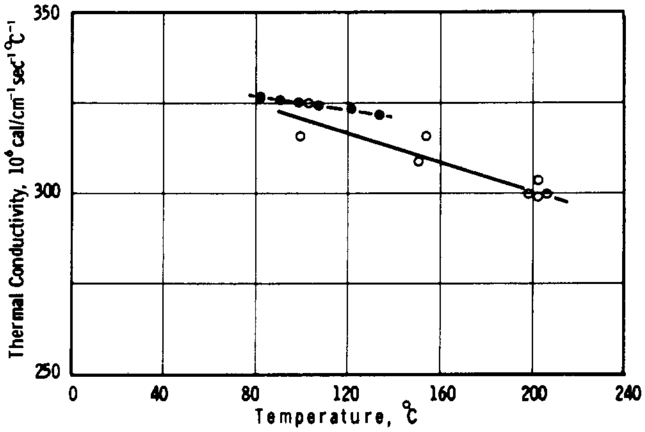

Figure 3. Thermal conductivity of liquid m-terphenyl

○ This work
● Horrocks and MacLaughlin (*17*)

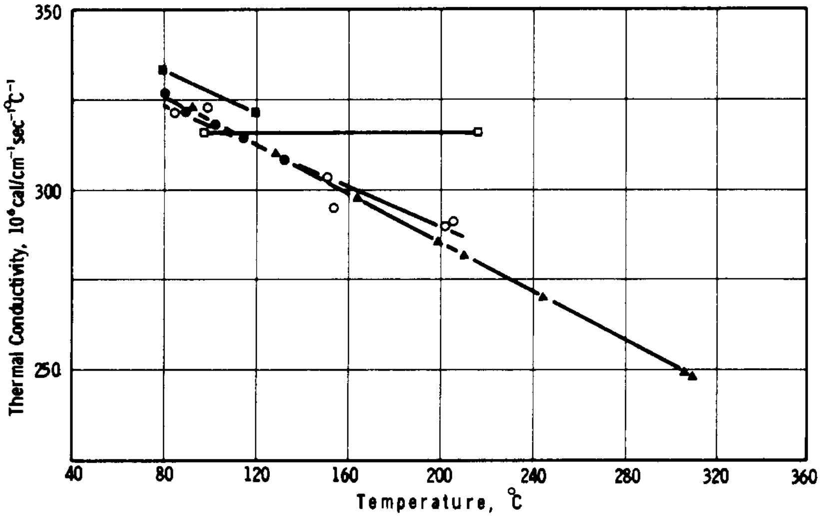

Figure 1. Thermal conductivity of liquid biphenyl

○ This work
■ Briggs (*6*)
□ Cecil, Koerner, and Munch (*10*)
● Horrocks and MacLaughlin (*17*)
▲ Ziebland and Burton (*26*)

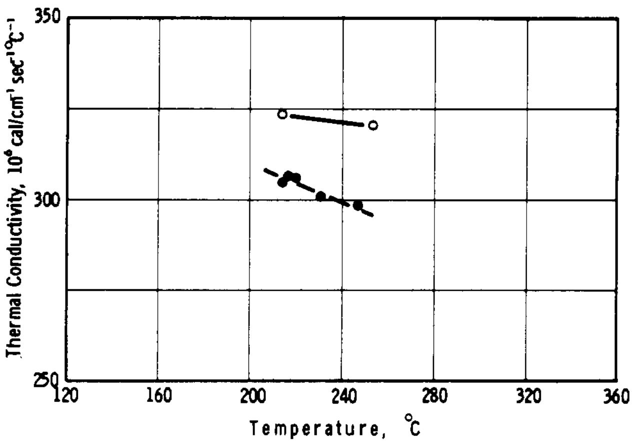

Figure 4. Thermal conductivity of liquid p-terphenyl

○ This work
● Horrocks and MacLaughlin (17)

Biphenyl $C_p° = 36.54 + 0.1206t - 5.225 \times 10^{-5}t^2$ (21)

o-Terphenyl $C_p° = 56.01 + 0.1704t - 7.028 \times 10^{-5}t^2$ (22)

m-Terphenyl $C_p° = 54.37 + 0.1758t - 7.522 \times 10^{-5}t^2$ (23)

p-Terphenyl $C_p° = 54.02 + 0.1761t - 7.544 \times 10^{-5}t^2$ (24)

The resulting thermal conductivity equations are:

Biphenyl $k° = 14.65 \times 10^{-6} + 11.68 \times 10^{-8}t + 1.856 \times 10^{-10}t^2 - 1.372 \times 10^{-13}t^3$ (25)

o-Terphenyl $k° = 14.41 \times 10^{-6} + 9.94 \times 10^{-8}t + 1.461 \times 10^{-10}t^2 - 0.680 \times 10^{-13}t^3$ (26)

m-Terphenyl $k° = 13.47 \times 10^{-6} + 9.58 \times 10^{-8}t + 1.448 \times 10^{-10}t^2 - 0.702 \times 10^{-13}t^3$ (27)

p-Terphenyl $k° = 13.47 \times 10^{-6} + 9.61 \times 10^{-8}t + 1.460 \times 10^{-10}t^2 - 0.707 \times 10^{-13}t^3$ (28)

Saturated Vapor Thermal Conductivity. The thermal conductivities of saturated polyphenyl vapors, k_s, were calculated using a method of corresponding pressure states (*16*). The ratio of saturated vapor thermal conductivity to that at low pressure is tabulated in Table XI.

Other Properties. Equations for the thermal conductivities of the polyphenyls in the unsaturated vapor state were calculated (*16*) using the method of Lenoir, Junk, and Comings. These values at reduced temperatures above 1.0 and a reduced pressure of 0.3 are approximately 10% higher than the corresponding values at zero pressures.

Table XI. Ratio of Saturated Vapor Thermal Conductivity to Low-Pressure Vapor Thermal Conductivity for Polyphenyls

Temp., ° C.	$k_s/k°$			
	Biphenyl	*o*-Terphenyl	*m*-Terphenyl	*p*-Terphenyl
93	1.00	1.00	1.00	. . .
204	1.08	1.03	1.02	. . .
316	1.13	1.09	1.08	1.07
427	1.17	1.14	1.12	1.12
538	. . .	1.20	1.18	1.17

The relative over-all heat transfer performances of polyphenyls were calculated in both the liquid and vapor states (*16*) by inserting the physical property values in the Dittus-Boelter equation (*25*).

$$h = 0.313 \times 10^{-6} \left(\frac{V^{0\cdot84}}{D^{0\cdot16}}\right) \left(\frac{k_L{}^{0\cdot60} \quad \rho_L{}^{0\cdot84} \; c_p{}^{0\cdot40}}{\mu^{0\cdot44}}\right) \qquad (29)$$

In the liquid state, biphenyl is the best coolant among the pure polyphenyls at temperatures up to 300° C., whereas *o*-terphenyl is superior at higher temperatures. However, certain liquid biphenyl-terphenyl mixtures appear to be superior as coolants to the pure polyphenyls. In the vapor state biphenyl is superior in heat transfer performance to the terphenyls at all temperatures.

NOMENCLATURE

a = radius of sample cup, cm.
b = radius of wire, cm.
c_p = specific heat of fluid, cal.-g.$^{-1}$-° C.$^{-1}$
$C_p°$ = specific heat of vapor at a constant low pressure, cal.-g. mole^{-1}-° K.$^{-1}$
D = diameter, cm.
$f(kT/\varepsilon)$ = viscosity temperature function, dimensionless
h = heat transfer coefficient, B.t.u.-ft.$^{-1}$-hr.$^{-1}$-° F.$^{-1}$
k = Boltzmann constant, cal.-molecule^{-1}-° K.$^{-1}$
k_L = thermal conductivity of liquid, cal.-cm.$^{-1}$-sec.$^{-1}$-° C.$^{-1}$
k_s = thermal conductivity of saturated vapor, cal.-cm.$^{-1}$-sec.$^{-1}$-° C.$^{-1}$
$k°$ = low pressure vapor thermal conductivity, cal.-cm.$^{-1}$-sec.$^{-1}$-° C.$^{-1}$
L = length of cell wire, cm.
M = molecular weight, dimensionless
Q = heat input to cell, cal.-sec.$^{-1}$
T = temperature, ° K.
t = temperature, ° C.
t_a = temperature of wire, ° C.
t_b = temperature of bath, ° C.
T_c = critical temperature, ° K.
T_r = reduced temperature, dimensionless
V = velocity of fluid, cm.-sec.$^{-1}$
V_c = critical volume, cc.-g. mole^{-1}
ε = intermolecular potential parameter, cal.-molecule^{-1}
η = kinematic viscosity of liquid, centistokes
μ = viscosity of liquid, centipoises
$\mu°$ = viscosity of low pressure vapor, centipoises
μ_s = viscosity of saturated vapor, centipoises
ρ_L = density of liquid, g.-cc.$^{-1}$

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Viscosity of Solutions of Some Electrolytes in Heavy Water

ANATOL SELECKI, BOGDAN TYMINSKI, and ANDRZEJ G. CHMIELEWSKI
Department of Nuclear Chemical Engineering, Warsaw Technical University, Warsaw, Poland

The viscosities of solutions of nine electrolytes—KCl, KI, KBr, $KMnO_4$, LiCl, Li_2SO_4, Na_2CO_3, NaF, and $MnSO_4$—in heavy water have been determined at 25°, 35°, 45°, 60°, 75°, and 90° C. at various concentrations up to near saturation.

INVESTIGATIONS of viscosity of electrolyte solutions can give some information about their structure (*1, 4*). There are also relationships between viscosity and heat of evaporation (*2*) or activity coefficients of solution components (*3*). This paper investigates the differences that may be caused in viscosity and solution structure by replacing ordinary water with heavy water.

EXPERIMENTAL

The viscosities of solutions were measured with an Ostwald viscosimeter (Figure 1). The time of flow was four minutes for ordinary water at 25° C. Because of the viscosimeter design, the solutions did not come into contact with the atmosphere, thus protecting the heavy water from dilution by air moisture.

Viscosity was calculated according to the formula

$$\eta = \eta_0 \frac{\rho t}{\rho_0 t_0} \qquad (1)$$

where η = viscosity, ρ = density, t = time of flow, and 0 refers to pure heavy water, which was used as a reference standard.

The ratio ρ/ρ_0 was measured in two manometers (Figure 2). One of the manometers was filled with the solution under investigation and the second one with pure heavy water, which was also used for preparing the solution. The ratio $\rho/\rho_0 = \Delta h_0/\Delta h$, where Δh is the difference of liquid levels in the arms of the manometer. The level of liquid was measured with a cathetometer.

Measurements were made in each case, and the ratio ρ/ρ_0 was determined for each solution. The viscosimeter and manometers were thermostated with an accuracy of ±0.05° C.

MATERIALS

The heavy water used in these experiments was produced in the USSR. Its concentration was determined by the picnometric method. For measurements 1 to 27, the concentration of heavy water was 99.8 mole % D_2O; for 28 to 35, 98.8 mole % D_2O; and for 36 to 43, 98.5 mole % D_2O. Before preparation of the solutions, the heavy water was distilled twice.

The salts used were of analytical purity and were dried but not purified before use.

RESULTS

Measurements were made for the solutions of nine salts—KCl, KI, KBr, $KMnO_4$, LiCl, Li_2SO_4, Na_2CO_3, NaF, and $MnSO_4$—at 25°, 35°, 45°, 60°, 75°, and 90° C.