

## Part II

# PHYSICAL PROPERTIES EVALUATION OF COMPOUNDS AND MATERIALS

## Viscosity and Thermal Conductivity of Water: Gaseous and Liquid States

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Experimental viscosity and thermal conductivity measurements reported in the literature for the gaseous and liquid states of water have been comprehensively reviewed and used to develop reduced state correlations for these transport properties. Values at atmospheric pressure have been used to establish relationships between the transport properties  $\mu^*$  and  $k^*$  and reduced temperature. For the dense gaseous and liquid regions residual viscosities,  $\mu - \mu^*$ , and residual thermal conductivities,  $k - k^*$ , were related to the corresponding density of water to generate continuous relationships for each of these transport properties. These relationships enabled the establishment of these transport properties at the critical point to be as follows:

$$\mu_c = 4300 \times 10^{-5} \text{ cp.} \quad \text{and} \quad k_c = 55.3 \times 10^{-5} \text{ cal./sec. cm. } ^\circ \text{K.}$$

From this information reduced state plots for the viscosity and thermal conductivity of water were established. Values resulting from the reduced state correlations, when compared with experimental values, produce average deviations of 1.61% for 120 viscosity points and 2.31% for 150 thermal conductivity points covering the conditions of temperature and pressure reported by the various investigators.

IN THIS STUDY, an attempt is made to present the viscosity and thermal conductivity of gaseous and liquid water over a wide range of temperatures and pressures. Although water has received considerable attention from an experimental standpoint, its properties have yet to be clearly defined over extremes of conditions. This is partly due to the fact that water possesses an unusual chemical structure which produces a high polarity within the molecule and a corresponding strong hydrogen bonding effect. From a theoretical point of view, these effects reflect to produce force interactions between molecules which become complex in nature and consequently have a pronounced effect on its transport properties.

As water possesses characteristics that are unique, this substance must be treated separately in order to establish its behavior in the gaseous and liquid states. Interest in the properties of water has recently been stimulated by

modern technological advances and particularly because the advent of nuclear energy has forced the frontiers of conditions to extremes. In this study attempts have been made to obtain the fragmentary information on water which, although valuable, is found to be restricted to the conditions of the experimental study.

Considerable attention has recently been given to the critical compressibility factor,  $z_c$ , as a correlating parameter (32). With the exception of helium, hydrogen, and neon, the critical compressibility factors of other substances decrease with increasing molecular complexities. The inert gases, argon, krypton, and xenon, and the diatomic gases, nitrogen, oxygen, and carbon monoxide represent the upper limit of the  $z_c$ -spectrum, while highly hydrogen bonded and polar substances represent the other extreme. Examples of substances representing this lower limit are water ( $z_c = 0.231$ ), methanol ( $z_c = 0.222$ ), and hydrogen cyanide

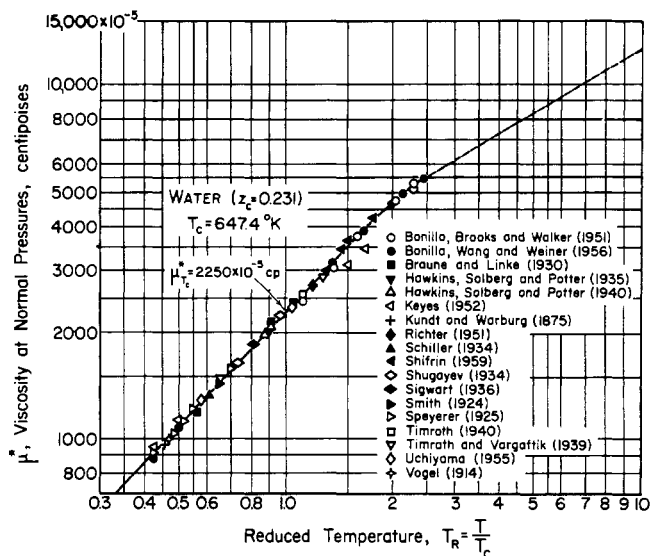


Figure 1. Viscosity-temperature relationship for water vapor at atmospheric pressure

( $z_c = 0.197$ ). Since water is representative of molecules possessing complex behavior, a study of its viscosity and thermal conductivity was undertaken in order to establish these transport properties and compare them with the corresponding properties of normally behaving substances.

#### VISCOSITY AT NORMAL PRESSURES

Experimental viscosities for water vapor at atmospheric pressure were obtained from 18 references ranging from the early work of Kundt and Warburg (29) in 1875 to the most recent data of Shifrin (43) reported in 1959. The most extensive work is that reported by Bonilla, Wang, and Weiner (5) for the temperature range  $T_R = 0.427$  to  $T_R = 2.43$ . Their values were not only internally consistent, but were also in agreement with the measurements of other investigators. The viscosities of Bonilla, Wang, and Weiner and those of other investigators are plotted *vs.* reduced temperature on log-log coordinates as presented in Figure 1 (4, 6, 16, 45, 48, 57, 66). The resulting relationship is linear with temperature below  $T_R = 1.50$ . For this region, the viscosity of water vapor at normal pressures (0.2 to 5 atm.) becomes

$$\mu^* = 2250 \times 10^{-5} T_R^{1.07} \quad T_R \leq 1.5 \quad (1)$$

Above  $T_R = 1.50$ , the relationship of Figure 1 exhibits a change in slope. Bonilla, Wang, and Weiner present extrapolated viscosity values for water for  $T_R = 3.20$ . These extrapolated values were used to extend the curve of Figure 1 to  $T_R = 3.2$ , and since in this high temperature region the relationship between  $\mu^*$  and  $T_R$  was essentially linear, the curve of Figure 1 was extended up to  $T_R = 10$ . This extrapolation assumes that water does not dissociate at these high temperatures.

Itean, Glueck, and Svehla (22) assumed that the angle dependent term,  $\Omega$  of the Stockmayer potential has its maximum value of 2 and calculated collision integrals,  $\Omega(T_N, \delta^*)$ , for polar molecules for use in the following theoretical relationship for gaseous viscosity (20):

$$\mu^* = 2.6693 \times 10^{-5} \frac{(MT)^{1/2}}{\sigma^2 \Omega(T_N, \delta^*)} \quad (2)$$

where  $T_N = T/(\epsilon/\mu)$  and  $\sigma$ ,  $\epsilon$ , and  $\delta^*$  are parameters for the Stockmayer potential. The collision integrals calculated previously by Krieger (28) for the same model were found

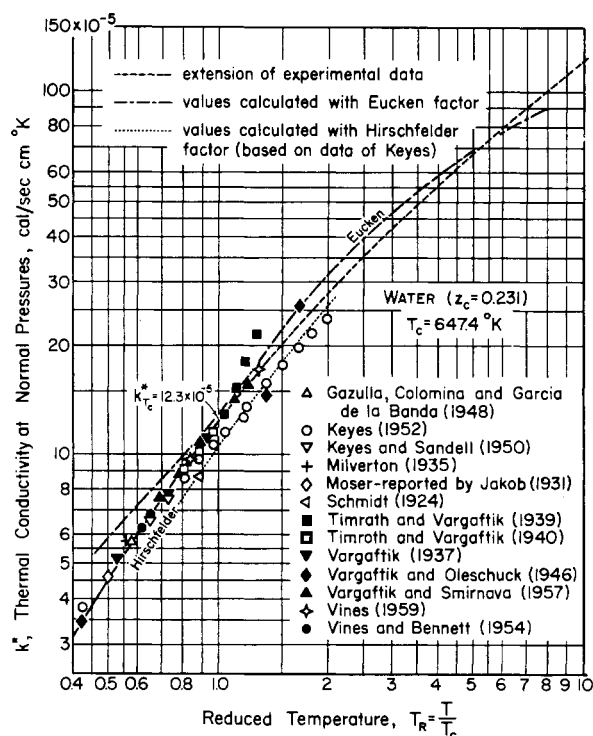


Figure 2. Thermal conductivity-temperature relationship for water vapor at atmospheric pressure

by Itean *et al.* to be in error. Itean *et al.* concluded that satisfactory viscosity values could not be calculated with their collision integrals for such highly polar substances as water. Monchick and Mason (35) have recently calculated collision integrals for the complete Stockmayer potential with the assumption that the relative orientation of the molecules is fixed during a collision and that each such orientation was equally probable. Monchick and Mason experienced difficulty in obtaining reliable values of  $\sigma$ ,  $\epsilon$ , and  $\Delta^*$  from the available experimental data and concluded that the Stockmayer potential does not accurately represent the complex nature of water. Therefore, no theoretical calculations were made to confirm the atmospheric viscosity behavior presented in Figure 1.

#### THERMAL CONDUCTIVITY AT NORMAL PRESSURES

Experimental thermal conductivities were not as plentiful as viscosity values, and as a result, reliable thermal conductivities were limited to temperatures up to  $T_R = 1.30$  (840° K.). A total of 13 references were used to establish the dependence of thermal conductivity at atmospheric pressure on temperature in the interval  $T_R = 0.425$  (275° K.) to  $T_R = 1.30$  (840° K.) as shown in Figure 2 (11, 26, 34, 41, 56, 60, 62, 64, 65). In order to extend this thermal conductivity relationship, the accepted high temperature viscosities of Figure 1 were used in conjunction with the expression:

$$k_m^* = \frac{15}{4} \frac{R}{M} \mu^* \quad (3)$$

to produce thermal conductivities which would be exhibited by water, if it were considered to behave as a monatomic molecule. To account for the polyatomic nature of the water molecule, the Eucken factor,  $\frac{3}{5}(C_v/R) + \frac{3}{5}$ , was applied to the results of Equation 3. The results are presented in Figure 2 and show that this simplified approach proves inadequate in defining the thermal conductivity of water. This conclusion is confirmed by Vargaftik (59) who

points out that the Eucken factor is dependable only for diatomic gases and fails to produce reliable results for higher order molecules.

An improvement on the Eucken factor has been presented by Hirschfelder (19), who develops the relationship

$$\frac{k_p^*}{k_m^*} = (1 - \delta_f) + \delta_f \frac{C_p}{C_{pf}} \quad (4)$$

where  $\delta_f$  is a dimensionless diffusion coefficient,  $nDC_{pf}/k_m^*$ . The variables in this coefficient are:  $n$ , the number of molecules/cm<sup>3</sup>,  $D$ , the selfdiffusion coefficient and  $C_{pf}$ , the heat capacity remaining when all rotations, vibrations, and electronic degrees of freedom in the molecule are frozen. Thus  $C_{pf} = \frac{5}{2}R$ .

For nonpolar substances, Hirschfelder (19) points out that  $\delta_f \approx 0.885$  and is independent of temperature. However, for water, this coefficient is temperature dependent and is considerably smaller. Specifically, these values for water are given as,

$t, ^\circ\text{C.}$	$\delta_f$
100	0.17
120	0.20
140	0.24
300	0.19
500	0.28
800	0.36
1000	0.39

These values, in conjunction with the heat capacity data reported by Wagman (67) permitted the calculation of  $k_p^*/k_m^*$ . When this ratio is multiplied by the results of Equation 3, thermal conductivities for water are obtained which account for the polyatomic and polar nature of this substance up to  $T_R = 2.0$  (1000° C.). A plot of  $\delta_f$  vs.  $T_R$  on log-log coordinates made possible the extension of  $\delta_f$  values to  $T_R = 4.0$ .

The thermal conductivities resulting from Equation 4 produce a relationship parallel to the experimental data for temperatures up to  $T_R = 1.30$ . The reason that these calculated values do not coincide with the majority of the reported data results from the fact that the  $\delta_f$  values of Hirschfelder were calculated using the data of Keyes (27) which are inconsistent with the values presented by the majority of the other investigators. The data of Keyes are also parallel to the accepted values and are displaced by the same amount as are the values calculated by Equation 4. Therefore, a superimposition of the thermal conductivities resulting from Equation 4 upon the experimental data has permitted the extension of the experimental curve up to  $T_R = 4.0$ .

The thermal conductivities between  $T_R = 1.0$  and  $T_R = 4.0$  were then used to delineate the  $k^*$  vs.  $T_R$  relationship by means of the expression,

$$\left(\frac{k^*}{k^*_{T_c}}\right)^{1.25} = 1.85 T_R - 0.85 \quad (5)$$

where  $k^*_{T_c} = 12.3 \times 10^{-5}$  cal./sec. cm. °K. Equation 5 has permitted the extension of the thermal conductivity relationship of Figure 2 up to  $T_R = 10$ . This extension again assumes that no dissociation occurs at these higher temperatures. This extrapolation is linear in this high temperature range and is therefore consistent with the behavior of the monatomic and diatomic gases (36, 39).

#### VISCOSITY AND THERMAL CONDUCTIVITY IN THE DENSE GAS AND LIQUID REGIONS

To correlate experimental viscosities and thermal conductivities available for the dense gas and liquid regions,

the densities of water must be available at the corresponding temperatures and pressures. Density values were obtained from an enlarged plot of the reduced density correlations presented in Figures 3 and 4. The construction of these figures has been based on the experimental  $P$ - $V$ - $T$  data of water compiled by Byrne and Thodos (8). A total of 26 references (8) was used to establish reduced density values which were extended by plotting pressure versus temperature for lines of constant reduced density. These constant density lines were essentially linear in the high pressure region and consistent with the  $P$ - $V$ - $T$  studies of Martin and Hou (33). These plots permitted the extrapolation of density values up to  $T_R = 10$  and  $P_R = 20$ .

Viscosities and thermal conductivities in the dense gas and liquid regions have been shown to become independent of temperature and pressure when the residual of these transport properties is related to the corresponding density (1, 7, 36, 44). The residual viscosity,  $\mu - \mu^*$ , and the residual thermal conductivity,  $k - k^*$ , were obtained from the high pressure and liquid state data by subtracting from them the corresponding  $\mu^*$  and  $k^*$  values obtained from Figures 1 and 2, respectively. Thus the residual quantities represent the isothermal effect of pressure on these transport properties.

Altogether the information of 25 literature sources was used to establish the residual viscosity-reduced density relationship shown in Figure 3 (2, 3, 14, 15, 18, 21, 37, 38, 47, 52). Many of these references not presented in this figure were restricted to viscosity measurements of liquid water at normal temperatures and therefore add no new information. The complete listing of these literature sources may be found elsewhere (51). The data of Timroth (53) for liquid water and water vapor made possible the establishment of the  $\mu - \mu^*$  vs.  $\rho_R$  relationship. These data cover the range of temperature, 18° to 600° C., for pressures up to 300 atm. Timroth's gaseous and liquid state data produce a single continuous curve which agrees well with the data of other investigators in the liquid region.

In the low density region, a considerable scattering exists for the data of Hawkins, Solberg, and Potter (17), Schiller (40), Sigwart (46), Speyerer (49), and Timroth and Vargaftik (54) and these data do not correlate properly with liquid phase values. Timroth (50) points out that discrepancies exist in the data of Hawkins, Solberg, and Potter (17) and those of Timroth and Vargaftik (54). Because of these disparities, these data were not considered in the development. In the liquid region, the residual relationship increases rapidly for small changes in density and therefore requires an expanded density scale in order to obtain exact residual viscosities in this region. Such an expanded plot is shown in the insert of Figure 3. This residual viscosity relationship permits the establishment of the viscosity of water at the critical point. This is made possible from the residual viscosity value at  $\rho_R = 1.00$ . The value  $(\mu - \mu^*)_c = 2050 \times 10^{-5}$  cp., in conjunction with the corresponding viscosity at normal pressures,  $\mu^*_{T_c} = 2250 \times 10^{-5}$  cp. produces a critical viscosity of  $\mu_c = 4300 \times 10^{-5}$  cp.

The same approach was used to develop the corresponding residual thermal conductivity-density relationship presented in Figure 4. The data of Timroth and Vargaftik (55) and Vargaftik and Tarzemanov (63) were used to establish this relationship in the dense gas region. When extended to the liquid state, this relationship goes through the liquid state data of a number of other investigators. This residual thermal conductivity plot made possible the determination of the thermal conductivity at the critical point. At  $\rho_R = 1.00$ ,  $(k - k^*)_c = 43.0 \times 10^{-5}$  cal./sec. cm. °K. produces the critical thermal conductivity for water,  $k_c = 55.3 \times 10^{-5}$  cal./sec. cm. °K.

For this residual thermal conductivity correlation, the experimental data of 50 references for liquid water were

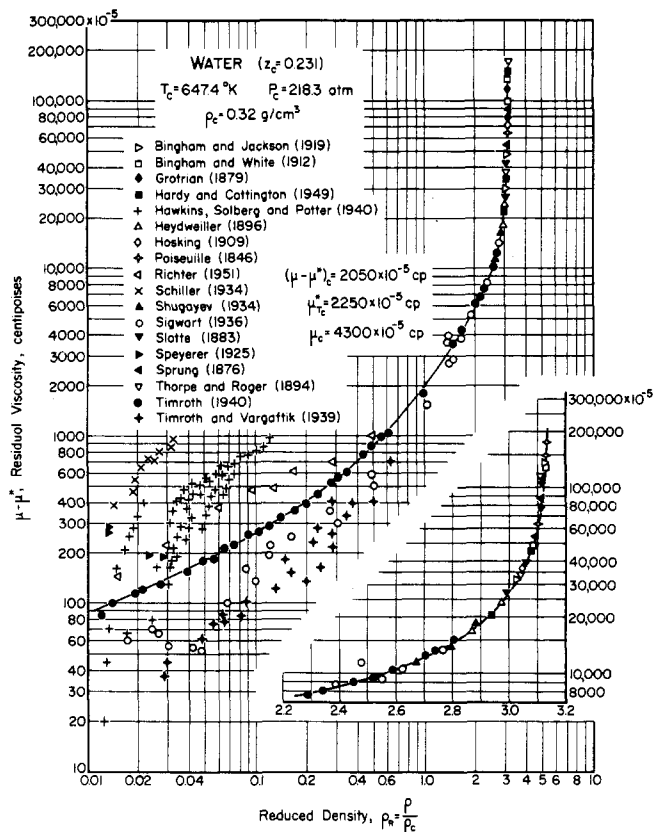


Figure 3. Residual viscosity-reduced density relationship for water

consulted. Only the most significant of these have been presented in Figure 4 (9, 23, 24, 31). A complete list of references may be found elsewhere (51). Thermal conductivities for liquid water have been measured up to pressures of 8000 atm. (30) and up to temperatures of 350° C. (61).

These data show a consistent and unusual behavior in that the thermal conductivity for liquid water reaches a maximum and then rapidly decreases with increasing density. For elevated pressures, this general trend continues to exist and produces similar relationships displaced to a somewhat higher value. Of particular interest are the data of Lawson, Lowell, and Jain (30). No such behavior has yet been observed for other substances (13, 25, 36, 39). However, Lawson *et al.* point out that this behavior may not be necessarily restricted to water if extremely high pressures are applied to other substances. More specifically, Lawson *et al.* predict that for organic liquids this reversal in thermal conductivity may occur at pressures in the range of 3000 atm. To define more clearly the high pressure region for liquid water, this section of the curve has been expanded and is shown in the insert of Figure 4.

#### REDUCED STATE CORRELATIONS

The residual viscosity relationship of Figure 3 in conjunction with a reduced density plot for water (8) have permitted the construction of a reduced state viscosity correlation for gaseous and liquid water. The critical viscosity,  $\mu_c = 4300 \times 10^{-5}$  cp. and Figure 1 permitted the construction of the base isobar,  $P_R \approx 0$ . The effect of pressure has been included through the use of the residual viscosity curve and the corresponding density of water. Each isobar was established by obtaining densities for the

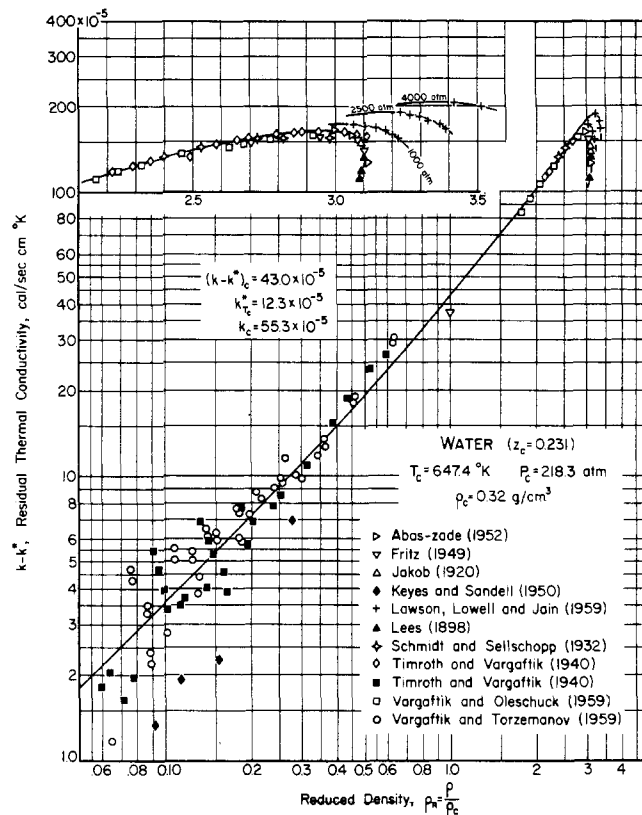


Figure 4. Residual thermal conductivity-reduced density relationship for water

entire range of temperatures, which were used to obtain residual viscosities,  $\mu - \mu^*$ , from Figure 3. The viscosity at the pressure of the isobar and a specified temperature was then calculated from this residual viscosity and the corresponding viscosity at normal pressures from Figure 1. These values when reduced produced the isobars presented in Figure 5. The reduced viscosity correlation for water given in this figure is applicable to the gaseous and liquid states for temperatures up to  $T_R = 10$  and pressures up to  $P_R = 20$ . The high pressure isobars for the liquid state rise sharply with decreasing temperature and exhibit a reversal in curvature in the vicinity of the critical point.

The same approach was adopted for thermal conductivity to produce the reduced state correlation for water presented in Figure 6. This figure also covers temperatures up to  $T_R = 10$  and pressures up to  $P_R = 20$ . The unusual behavior of water in both the saturated and compressed liquid regions is well defined by the maxima occurring for all the isobars of Figure 6. These maxima occur at approximately 130° C. and have been substantiated by a number of investigators (30, 42, 55, 61). In discussing this phenomenon, Lawson, Lowell, and Jain (30) point out that the reason for this behavior is unknown, but that it may be associated with the collapse of ice-like water molecules at higher temperatures. Another hypothesis advanced to explain this phenomenon relates to the hydrogen bonding effects inherent in water. Gierer and Wirtz (12) point out that if the hydrogen bonds in the water molecule are broken, the  $H^+$ ,  $H_3O^+$ , and  $OH^-$  ions would diffuse under a temperature gradient and carry with them an energy of activation necessary for their formation. It may be significant that the mobility of these groups increases and passes through a maximum at approximately the same temperature that the thermal conductivity becomes a maximum. The exact

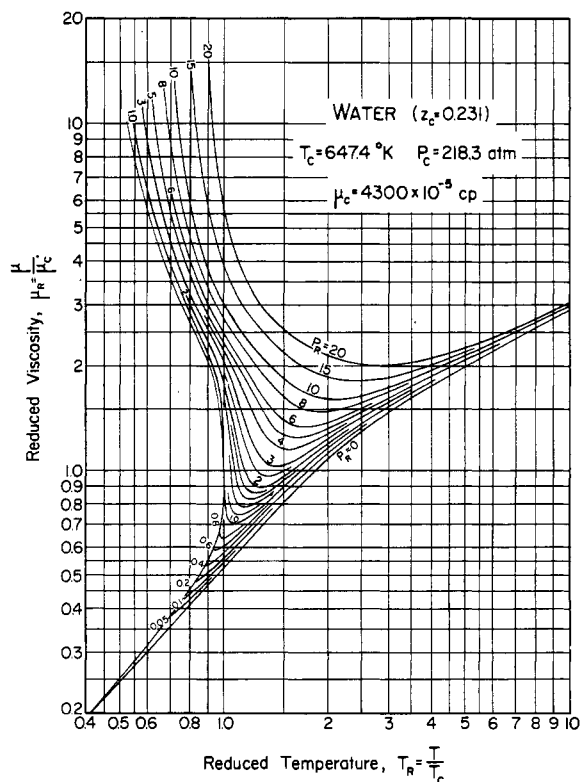


Figure 5. Reduced state correlation for the viscosity of water

explanation of this phenomenon requires further investigation. In the other regions, the thermal conductivity of water follows a normal behavior.

## CONCLUSIONS

As a result of this study, the following basic properties for water have been established:

$$\begin{aligned} \mu^* T_c &= 2250 \times 10^{-5} \text{ cp.} \\ (\mu - \mu^*)_c &= 2050 \times 10^{-5} \\ \mu_c &= 4300 \times 10^{-5} \\ k^* T_c &= 12.3 \times 10^{-5} \text{ cal./sec. cm. }^\circ\text{K.} \\ (k - k^*)_c &= 43.0 \times 10^{-5} \\ k_c &= 55.3 \times 10^{-5} \end{aligned}$$

The development of the correlations in Figures 5 and 6 are based on these  $\mu_c$  and  $k_c$  values.

Comparisons of viscosity and thermal conductivity values calculated from Figures 5 and 6 have been made with experimental data from all of the references consulted in this study. For viscosity and average deviation of 1.61% resulted from a consideration of 120 experimental points covering the range of available gaseous and liquid state data. Similar comparisons for thermal conductivity produced an average deviation of 2.31% for over 150 experimental points considered.

Because of the unusual nature of the water molecule, this substance does not adhere to the principle of corresponding states. Values calculated from the generalized viscosity correlation of Uyehara and Watson (58) produced an over-all average deviation of 68% for 40 points considered. Specifically for the gaseous state, the average deviation is 22.8% for 20 points while attempting to predict viscosities for the liquid state, deviations as high as 130% were encountered. The average deviation for the liquid state was 113% for 20 points.

The Gamson generalized plot for the thermal conductivity of gases (10) is capable of reproducing experi-

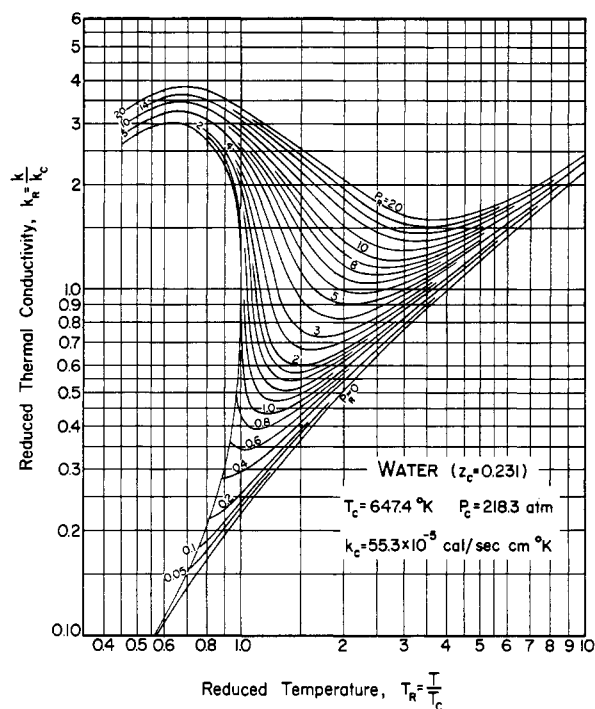


Figure 6. Reduced state correlation for the thermal conductivity of water

mental values with an average deviation of 9.3% for 40 points examined. Gamson's generalized correlation is limited in that it is only applicable to the gaseous state.

These comparisons indicate that the correlations of Figures 5 and 6 are capable of producing these transport properties accurately despite the deviations encountered in the experimental values reported in the literature. The development of Figures 5 and 6 assumes that no dissociation of water occurs for temperatures up to  $T_R = 10$ .

## NOMENCLATURE

- $\delta^*$  = reduced dipole energy modulus,  $\frac{1}{2} (\mu^2 / \epsilon \sigma^3)$
- $\delta_i$  = dimensionless diffusion coefficient,  $nDC_{pl} / k_m^*$
- $D$  = self-diffusion coefficient,  $\text{cm.}^2/\text{sec.}$
- $\epsilon$  = maximum energy of attraction for Lennard-Jones potential, ergs
- $\kappa$  = Boltzmann constant,  $1.38047 \times 10^{-16} \text{ ergs}/^\circ\text{K.}$
- $\mu$  = dipole moment, Debye units
- $\mu$  = viscosity, centipoises
- $\mu^*$  = viscosity at normal pressures, centipoises
- $\mu^* T_c$  = viscosity at normal pressures and the critical temperature, centipoises
- $\mu_c$  = viscosity at the critical point, centipoises
- $\rho$  = density,  $\text{g./cm.}^3$
- $\rho_c$  = density at the critical point,  $\text{g./cm.}^3$
- $\rho_R$  = reduced density,  $\rho / \rho_c$
- $\sigma$  = collision diameter for Lennard-Jones potential, A.
- $\Psi[T_N]$  = reduced collision integral for the Lennard-Jones potential,
- $\Omega[T_N, \delta^*]$  = reduced collision integral for the Stockmayer potential,
- $C_p$  = molar heat capacity at constant pressure,  $\text{cal./g.-mole }^\circ\text{K.}$
- $C_{pl}$  = molar heat capacity for monatomic gas,  $5R/2$ ,  $\text{cal./g.-mole }^\circ\text{K.}$
- $C_v$  = molar heat capacity at constant volume,  $\text{cal./g.-mole }^\circ\text{K.}$
- $k$  = thermal conductivity,  $\text{cal./sec. cm. }^\circ\text{K.}$
- $k^*$  = thermal conductivity at normal pressures,  $\text{cal./sec. cm. }^\circ\text{K.}$

$k_m^*$  = thermal conductivity at monatomic gas at normal pressures, cal./sec. cm. °K.  
 $k_p^*$  = thermal conductivity for polyatomic gas at normal pressures, cal./sec. cm. °K.  
 $k^*_T$  = thermal conductivity at normal pressures and the critical temperature, cal./sec. cm. °K.  
 $k_c$  = thermal conductivity at the critical point, cal./sec. cm. °K.  
 $M$  = molecular weight  
 $n$  = number of molecules/cm.<sup>3</sup>  
 $P$  = pressure, atm.  
 $P_c$  = critical pressure, atm.  
 $P_R$  = reduced pressure,  $P/P_c$   
 $R$  = gas constant  
 $t$  = temperature, °C.  
 $T$  = absolute temperature, °K.  
 $T_N$  = normalized temperature,  $T/(t/\mu)$   
 $T_c$  = critical temperature, °K.  
 $T_R$  = reduced temperature,  $T/T_c$   
 $V$  = molar volume, cm.<sup>3</sup>/g.-mole  
 $z_c$  = critical compressibility factor,  $P_c v_c / RT_c$   
 $v_c$  = critical volume, cm.<sup>3</sup>/g.-mole

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