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Thermal Conductivity Measurements of Aqueous $SrCl_2$ and $Sr(NO_3)_2$ Solutions in the Temperature Range Between 293 and 473 K at Pressures up to 100 MPa[1]

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Accurate high-pressure thermal conductivity measurements have been performed on $H_2O + SrCl_2$ and $H_2O + Sr(NO_3)_2$ mixtures at pressures up to 100 MPa over a temperature range between 293 and 473 K using a parallel-plate apparatus. The concentrations studied were 0.025, 0.05, 0.10, 0.15, and 0.20 mass fraction of the salts. The estimated accuracy of the method is about ±1.6%. The pressure, temperature, and concentration dependences of the thermal conductivity have been studied. Measurements were made on six isobars, namely, 0.1, 20, 40, 60, 80, and 100 MPa. The thermal conductivity shows a linear dependence on pressure and concentration for all isotherms. Along each isobar, a given concentration shows the thermal-conductivity maximum at a temperature of about 413 K. The measured values of thermal conductivity at atmospheric pressure are compared with the results of other investigators. Literature data at atmospheric pressure reported by Ridel and by Zaitzev and Aseev agree with our thermal conductivity values within the estimated uncertainty.

KEY WORDS: aqueous solutions; density; high pressure; parallel-plate method; salt; thermal conductivity; water.

1. INTRODUCTION

Only limited experimental thermal-conductivity data over a wide range of temperatures, pressures, and concentrations are available in the literature

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for aqueous salt solutions. The thermal conductivity of $H_2O + SrCl_2$ and $H_2O + Sr(NO_3)_2$ solutions has been investigated previously [1, 2]. All measurements in these papers were performed at atmospheric pressure. In the paper of Ridel [1] the thermal conductivity of $H_2O + SrCl_2$ and $H_2O + Sr(NO_3)_2$ solutions were measured in a temperature range from 293 to 373 K at a pressure of 0.10 MPa for concentrations from 0.025 to 0.2 mass fraction of salt using a parallel-plate method. The thermal conductivity of liquid $H_2O + SrCl_2$ and $H_2O + Sr(NO_3)_2$ solutions at atmospheric pressure and at a temperature of 293 K has been reported by Zaitzev and Aseev [2] for concentrations from 0.05 to 0.25 mass fraction using a coaxial-cylinder method.

In this investigation, a parallel-plate cell was constructed for measuring the thermal conductivity of aqueous solutions of salts in the liquid and vapor phases at high temperatures (up to 473 K) and pressures up to 100 MPa.

2. EXPERIMENTAL

The apparatus and experimental procedures have been described in detail in previous publications [3–5], and only a brief description of the approach is given here. Essentially, the apparatus consists of a thermal-conductivity cell, a high-pressure vessel, and a liquid thermostat, dead-weight pressure gauge, a water-to-oil separator, and containers for degassed water and for solutions. The thermal-conductivity cell consists of three plates: guard plate, upper plate, and lower plate. The guard plate is surrounded by a guard heater. The thermal-conductivity cell has a cylindrical shape with a 21-mm height and 90-mm diameter. The cell is made from stainless steel. The fluid surrounds the cell and fills the gap between upper and lower plates. All plates were polished with powder of a small grain size (320 nm). In this way heat transport by radiation is small compared to the heat transport by conduction.

The thermal conductivity measurements were based on the measurement of the power transferred Q from the upper plate to the lower plate by conduction through the fluid layer, and the temperature difference, ΔT, across the fluid layer. The thermal conductivity λ of the fluid is deduced from the relation:

$$\lambda = \frac{QD}{S \Delta T} \tag{1}$$

where $S = (36.38 \pm 0.01)\ \mathrm{cm}^2$ is the effective area of the upper plate and d is the width of the measurement layer. The experimentally determined value of the power, Q_{exp}, was measured with an uncertainty of $\pm 0.12\%$. The temperature difference ΔT_{exp} was determined to better than $\pm 0.10\%$.

The thermal-conductivity cell is mounted inside a high-pressure vessel which is placed in a liquid thermostat. A molten salt mixture (45 wt% KNO_3 + 55 wt% $NaNO_3$) was used as the fluid in the thermostat. The fluid was circulated by a stirrer driven by a motor, and its temperature was maintained stable within ± 10 mK. The fluid temperature, which was detected by a platinum resistance thermometer, was automatically controlled. The measurements were performed with different values of ΔT in the range from 0.8 to 1.25 K. The pressures have been measured with an uncertainty of $\pm 0.05\%$.

Dimensions S and d of the thermal-conductivity cell vary with the applied temperature and pressure. The cell constant (d/S) has to be corrected for the thermal expansion and compression. For our apparatus this correction is given by

$$\frac{d}{S} = [1 - 0.152635 \times 10^{-4}(T - 293)] \left(\frac{d}{S}\right)_n \tag{2}$$

where $(d/S)_n = 0.0827684\ \mathrm{m}^{-1}$ is the value of the cell constant at a temperature of 293 K and at a pressure of 0.1 MPa. The effect of pressure is negligibly small in the pressure range of the experiments. The uncertainty of the cell constant is $\pm 0.3\%$. Taking into account the uncertainties in temperature, pressure, and concentration measurements, the total experimental uncertainty of thermal conductivity, $\overline{\delta\lambda}$, is less than $\pm 1.6\%$.

The parallel-plate method is the configuration in which convection is not likely to occur. Convective heat transfer increases with increasing values of the Rayleigh number [6]. The Rayleigh number Ra can be determined as follows:

$$\mathrm{Ra} = \frac{g\alpha_p\, \Delta T\, d^3 C_p \rho^2}{\lambda\eta} \tag{3}$$

where g is the acceleration of gravity, α_p is the thermal expansion coefficient of the fluid, ρ is the density, C_p is the specific heat at constant pressure, and η is the viscosity coefficient. To reduce the Ra, a small gap distance of $d = 301.0 \pm 0.1\ \mu\mathrm{m}$ was used, while the temperature difference, ΔT, employed in the measurements was 1 K. This made it possible to minimize the effect of convection. Convection could develop when the Ra exceeds a certain critical value Ra_c, which is about 2000 [7]. A possible cause of the convection is a temperature gradient in the layer in a horizontal direction. The absence of convection can be varified experimentally by

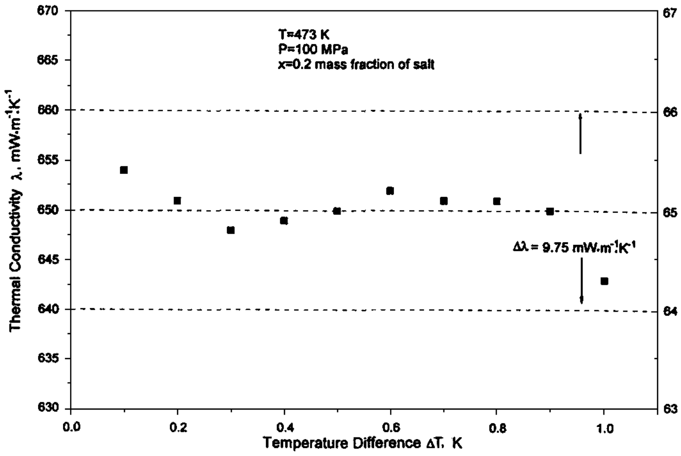

Fig. 1. Measured thermal conductivity as a function of the applied temperature difference ΔT between upper and lower plates for $H_2O + SrCl_2$ mixture ($x = 0.2$ mass fraction of $SrCl_2$, $P = 100$ MPa, and $T = 473$ K).

measuring the thermal conductivity with various temperature differences ΔT across the measurement gap. Examples of such series of measurements are shown in Fig. 1. The measured thermal conductivities were indeed independent of the applied temperature difference ΔT and power Q transferred from the upper plate to the lower plate.

The water was triply distilled and degassed, and had an electrical conductivity of about $10^{-4} \Omega^{-1} \cdot m^{-1}$. The solutions were prepared gravimetrically from reagent-grade $SrCl_2$ and $Sr(NO_3)_2$ (purity of 99.7%).

3. RESULTS AND DISCUSSION

Experimental thermal-conductivity data were obtained as a function of temperature along six isobars covering a range of pressures from 0.1 to 100 MPa. Some of the experimental results for $H_2O + SrCl_2$ and $H_2O + Sr(NO_3)_2$ solutions, including 300 thermal conductivity data for each system, are presented in Tables I and II. The average temperature in the fluid layer is equal to $T_L + 0.5\,\Delta T$, where T_L is the temperature of the lower plate.

The temperature dependence of the thermal conductivity of pure water and a $H_2O + SrCl_2$ solution along various isobars at a constant concentration are graphically shown in Fig. 2. Along each isobar, a given concentration shows a thermal-conductivity maximum at a temperatures of about

Table I. Experimental Thermal Conductivity Data of $H_2O + SrCl_2$

	Thermal conductivity λ ($mW \cdot m^{-1} \cdot K^{-1}$) at pressures P (MPa)					
T (K)	0.1	20	40	60	80	100
			$x = 0.025$ mass fraction of $SrCl_2$			
293	597	605	615	624	630	640
313	628	636	645	655	660	667
333	650	660	670	678	687	695
353	666	676	685	698	707	716
375	677	687	697	710	720	730
393	681	695	710	721	730	740
413	682	700	712	724	737	746
433	679	698	710	725	735	745
453	670	691	705	720	730	740
473	660	680	695	712	725	736
			$x = 0.05$ mass fraction of $SrCl_2$			
293	598	602	612	620	629	637
313	625	632	643	650	657	665
333	645	655	667	675	680	691
353	661	672	684	694	700	711
375	672	685	696	706	715	724
393	680	691	707	717	727	733
413	681	696	709	721	731	740
433	677	695	708	720	730	739
453	669	688	702	716	726	737
473	657	678	694	706	720	730
			$x = 0.10$ mass fraction of $SrCl_2$			
293	590	598	605	612	620	627
313	620	629	634	640	649	654
333	640	651	659	665	672	680
353	657	668	676	684	690	700
375	668	678	688	697	704	714
393	674	686	699	707	716	722
413	675	691	700	712	720	730
433	670	688	700	710	718	728
453	663	680	692	707	714	724
473	649	670	683	697	707	720
			$x = 0.15$ mass fraction of $SrCl_2$			
293	583	590	597	604	610	620
313	612	620	626	633	639	647
333	633	643	650	656	662	670
353	650	661	667	676	682	690
375	660	670	678	687	694	702
393	665	677	690	696	704	712
413	666	682	692	701	710	717
433	664	678	690	700	707	716
453	655	672	685	695	704	713
473	643	661	674	686	697	707

Table II. Experimental Thermal Conductivity Data of $H_2O + Sr(NO_3)_2$

	Thermal conductivity λ $(mW \cdot m^{-1} \cdot K^{-1})$ at pressures P (MPa)					
T(K)	0.1	20	40	60	80	100
			$x = 0.025$ mass fraction of $Sr(NO_3)_2$			
293	598	608	617	626	633	640
313	628	638	645	656	660	666
333	650	661	670	679	687	692
353	666	678	687	699	706	713
375	677	688	698	710	718	728
393	682	697	712	722	729	738
413	684	702	714	727	734	746
433	680	698	712	726	733	745
453	671	694	708	722	728	740
473	660	682	698	713	722	738
			$x = 0.05$ mass fraction of $Sr(NO_3)_2$			
293	596	604	612	619	626	635
313	624	635	640	650	655	663
333	648	655	665	672	680	688
353	660	673	680	691	700	709
375	672	682	693	703	712	724
393	680	690	705	714	724	733
413	680	696	707	719	729	738
433	675	692	705	718	728	739
453	667	689	701	713	723	735
473	658	679	690	704	716	730
			$x = 0.10$ mass fraction of $Sr(NO_3)_2$			
293	588	596	605	612	619	627
313	619	627	635	642	648	655
333	640	650	660	664	672	680
353	656	667	675	683	690	700
375	666	680	686	694	703	715
393	672	685	697	705	715	725
413	673	690	701	710	718	730
433	670	685	700	710	720	729
453	662	680	695	706	714	724
473	650	670	682	696	707	720
			$x = 0.15$ mass fraction of $Sr(NO_3)_2$			
293	583	591	600	605	612	619
313	612	621	629	635	640	645
333	636	642	652	660	665	670
353	650	660	670	678	685	690
375	660	670	680	688	695	705
393	666	677	690	698	705	712
413	667	682	694	704	710	720
433	663	680	692	702	710	717
453	655	674	688	699	705	713
473	643	662	677	688	700	708

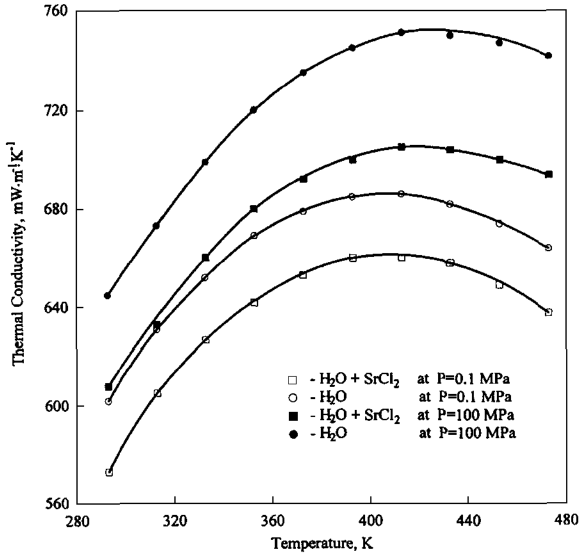

Fig. 2. Thermal conductivity of H_2O and $H_2O + SrCl_2$ solutions along various isobars as a function of temperature for a concentration of $x = 0.2$ mass fraction of $SrCl_2$.

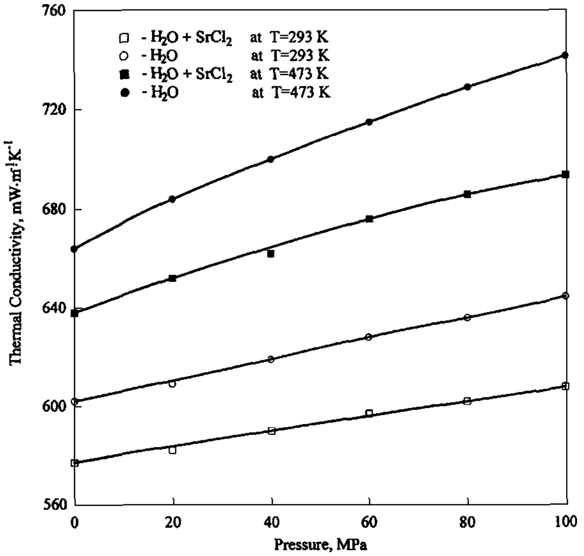

Fig. 3. Thermal conductivity of H_2O and $H_2O + SrCl_2$ solutions along various isobars as a function of pressure for a concentration of $x = 0.2$ mass fraction of $SrCl_2$.

413 K. The maximum in thermal conductivity at ≈413 K indicates that the temperature coefficient $(\partial\lambda/\partial T)_{P,X}$ has changed from a positive-to-negative value for all P and x. These observations are consistent with previous work on pure water and other aqueous salt solutions [3–5]. The dependence of the thermal conductivity on pressure P at various temperatures is graphically illustrated in Fig. 3. The thermal conductivity increases monotonically with increasing pressure along each isotherm for all concentrations. The pressure coefficient of the thermal conductivity, $(\partial\lambda/\partial T)_{T,X}$, is always positive and slowly changing for a given T and x. Variations of the thermal conductivities of $H_2O + SrCl_2$ solutions with concentration at constant temperature and pressure are shown in Fig. 4. From the data, it can be concluded that the derivative $(\partial\lambda/\partial T)_{P,X}$ is always negative and slowly changing for a given P and T.

The measurements were compared with values obtained by other authors [1, 2]. The deviations are shown in Figs. 5–7. The measurements of Zaitzev and Aseev [2] for $H_2O + SrCl_2$ and $H_2O + Sr(NO_3)_2$, which have been reported under the same conditions are in good agreement with the present results in the overlapping range. The average relative deviation was +0.3% for $H_2O + SrCl_2$, and ±1.2% for $H_2O + Sr(NO_3)_2$. Results

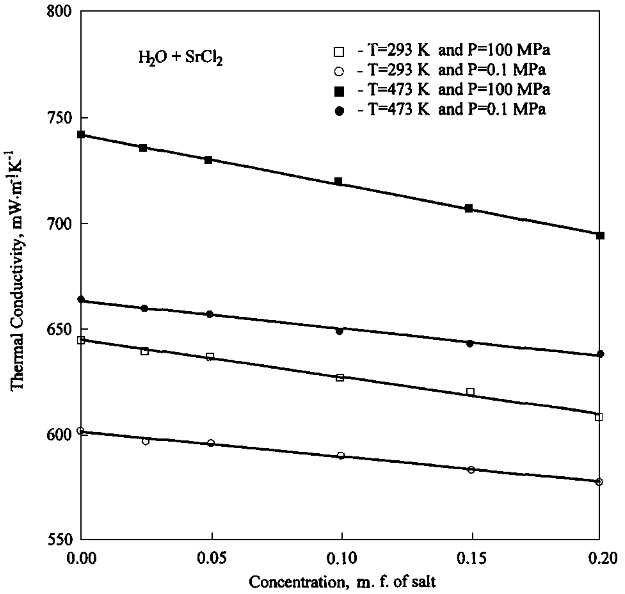

Fig. 4. Thermal conductivity of $H_2O + SrCl_2$ solutions at constant temperature and pressure as a function of concentration (m.f. = mass fraction).

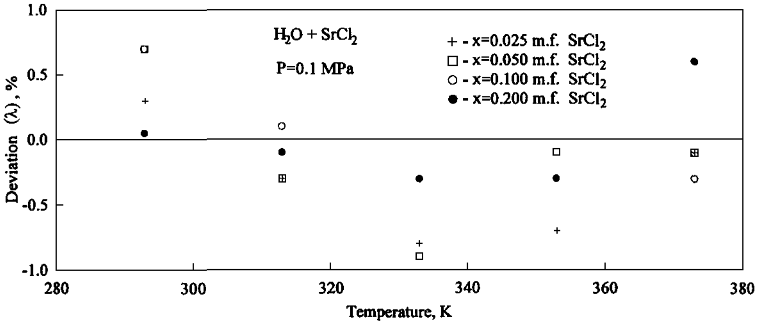

Fig. 5. Percentage deviations, $\delta\lambda = 100(\lambda_{exp} - \lambda_{this})/\lambda_{this}$, for thermal conductivity literature values [2] compared to present results at $P = 0.1$ MPa for $H_2O + SrCl_2$ (m.f. = mass fraction).

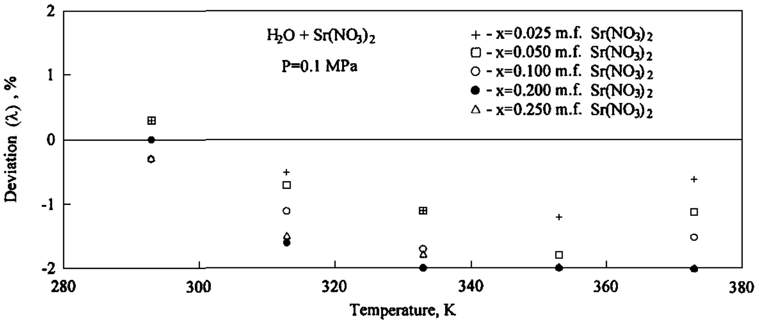

Fig. 6. Percentage deviations, $\delta\lambda = 100(\lambda_{exp} - \lambda_{this})/\lambda_{this}$, for thermal conductivity literature values [2] compared to present results at $P = 0.1$ MPa for $H_2O + Sr(NO_3)_2$ (m.f. = mass fraction).

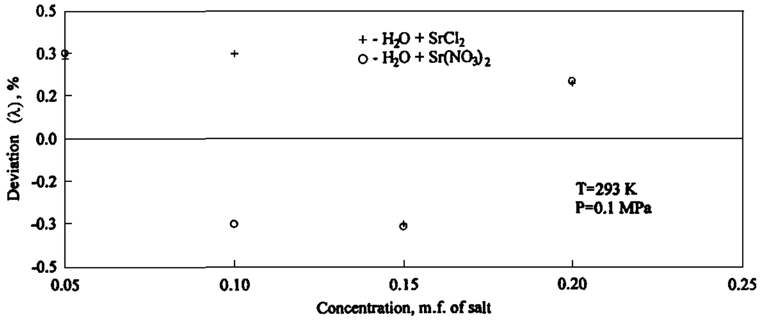

Fig. 7. Percentage deviations, $\delta\lambda = 100(\lambda_{exp} - \lambda_{this})/\lambda_{this}$, for thermal conductivity literature values [1] compared to present results at $P = 0.1$ MPa and $T = 293$ K for $H_2O + SrCl_2$ and $H_2O + Sr(NO_3)_2$ solutions (m.f. = mass fraction).

from Ref. 1 show deviations from this study of less than ±0.3%. Systematic errors were not observed, and the average deviation was ±0.25%.

4. CONCLUSION

By means of the guarded parallel-plate apparatus with a cylindrical thermal-conductivity cell, the $\lambda - P - T - x$ relationships for $H_2O + SrCl_2$ and $H_2O + Sr(NO_3)_2$ solutions have been measured in the range of temperatures from 293 to 473 K, at pressures up to 100 MPa, with concentrations between 0.025 and 0.20 mass fraction of salt [$SrCl_2$ and $Sr(NO_3)_2$]. The uncertainty was estimated to be ±1.6%.

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