International Journal of Thermophysics, Vol. 14, No. 4, 1993

Corresponding States Correlation for the Thermal Conductivity of Molten Alkali Halides[1]

Y. Nagasaka[2] and A. Nagashima[2]

Received December 31, 1992

The principle of corresponding states has been applied to the thermal-conductivity data for molten alkali halides which have been obtained by recent forced Rayleigh scattering measurements. The theory, which was developed by Harada et al. for the transport properties of uni-univalent molten salts, is based on the fluctuation-dissipation theorem with the pair interaction between ions composed of core repulsive and Coulombic potentials. Four characteristic parameters specific to each salt have been used to reduce the thermal conductivity and temperature. It has been found that the thermal conductivity of molten alkali halides is adequately correlated by the corresponding-states correlation ($\lambda^* \propto 1/T^*$) within experimental accuracy. By employing the correlation, the thermal conductivity of molten alkali fluorides, which could not be measured by the forced Rayleigh scattering method, is predicted.

KEY WORDS: alkali halides; correlation; corresponding states; molten salts; thermal conductivity; thermal diffusivity.

1. INTRODUCTION

In recent publications [1–3], we reported the results of new thermal diffusivity and thermal-conductivity determinations on 13 kinds of molten alkali halides in the temperature range from 937 to 1441 K at atmospheric pressure. The measurements have been carried out in the forced Rayleigh scattering instrument described in earlier publications [4–9]. Using this new optical technique, we have found that most of the previous experimental data for the thermal conductivity of molten alkali halides obtained by conventional steady-state methods contain significant systematic errors due

---

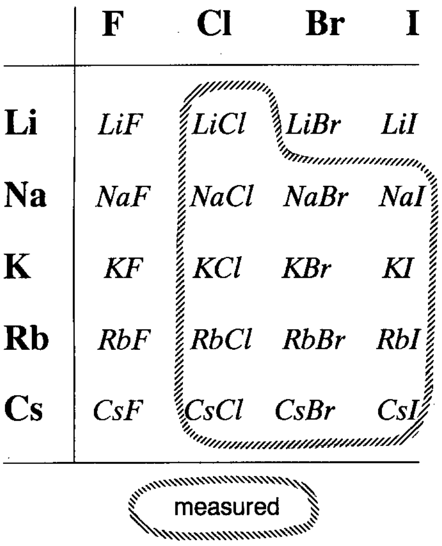
[1] Paper dedicated to Professor Joseph Kestin.

[2] Department of Mechanical Engineering, Keio University, 3-14-1 Hiyoshi, Yokohama 223, Japan.

0195-928X/93/0700-0923$07.00/0

to radiative and convective heat transfer in the sample salts which are inevitable at high temperatures. We have made the same observations in the case of molten $NaNO_3$ and KNO_3 [10]: the maximum systematic error is about 30% at 720 K. In contrast to molten alkali halides above 1000 K, these systematic errors grow up to 400 to 500% at the maximum.

Even though our new results have revealed correct behavior of the thermal conductivity of molten alkali halides, we have not succeeded in applying the forced Rayleigh scattering method to molten alkali metal fluorides such as LiF and KF, because of their strong corrosiveness against quartz and sapphire cells. Even in the case of LiCl near its melting point [1], the quartz glass cell turned slightly opaque (devitrification phenomena) during the course of measurements, which significantly increased the scattered light of the probing laser in the detected signals. Such an increase in scattered-light signals deteriorate the reproducibility. Moreover, fluoride salts completely corrode a sapphire glass. This problem may be experimentally solved by controlling the gas atmosphere around the high-temperature samples, or by using a diamond cell window, or by applying the levitation technique. Those solutions, however, seem to be difficult to apply for our present experimental setup. It should be mentioned that these fluoride salts are the most promising phase-change materials for



	F	Cl	Br	I
Li	*LiF*	*LiCl*	*LiBr*	*LiI*
Na	*NaF*	*NaCl*	*NaBr*	*NaI*
K	*KF*	*KCl*	*KBr*	*KI*
Rb	*RbF*	*RbCl*	*RbBr*	*RbI*
Cs	*CsF*	*CsCl*	*CsBr*	*CsI*

Fig. 1. Thirteen kinds of measured molten alkali halides by the forced Rayleigh scattering method of the 20 salts in total.

the solar receiver/thermal energy storage system in space due to their high latent heat of fusion per unit mass [11]. Therefore, reliable thermophysical-properties data, especially thermal conductivity, of fluoride salts are of practical importance.

If we remamber that we have systematically determined the thermal conductivity of 13 molten alkali halides of 20 salts in total as shown in Fig. 1, and these salts have the simplest molecular structure among other molten salts, which simplifies the theoretical treatment, it may be possible to correlate the measured data by means of the law of corresponding states [12]. Thus we shall be able to predict the thermal conductivity of fluorides and other salts for which no reliable experimental data are available.

2. LAW OF CORRESPONDING STATES FOR MOLTEN SALTS

2.1. Introduction

It is well-known that the principle of corresponding states is a useful tool for estimating the thermodynamic and transport properties of gases and liquids [13]. The principle was originated by van der Waals and is based on the reduction of the variables with the critical constants. The theoretical basis of the existence of corresponding states relationship for thermodynamic properties can be derived in case of the gases whose molecules are spherical and nonpolar. In other words, the intermolecular pair potential $\phi(r)$ for these molecules can be represented by some universal function f with two scaling parameters ε and σ in the form

$$\phi(r) = \varepsilon f(r/\sigma) \qquad (1)$$

For the transport properties of dilute gases, Kestin and his collaborators [14] proved that the collision integrals for all potentials of Eq. (1) are universal functions of the reduced temperature $(T^* = kT/\varepsilon)$ through the experimental data for monatomic gases. Moreover, Kestin's group has been able to show that this universality can be extended for the interactions between several quite complicated nonpolar molecules and some weakly polar molecules and for the interactions of the molecules with monatomic species [15, 16]. By using this extended law of corresponding-states correlation, it is possible to calculate the viscosity, diffusion coefficient, and thermal conductivity of gases and gas mixtures at low density with good accuracy [17].

With respect to the transport properties of polyatomic liquids, we no longer have the principle of corresponding states in the same sense of rigor as for filute gases. However, if we confine ourselves to the same group of substances such as normal alkanes and tolerate some loss of accuracy, it has been proved that a unified correlation scheme based on consideration of the exact hard-sphere theory is valid for the thermal conductivity, viscosity, and self-diffusion coefficient [18, 19].

Considering the above-mentioned facts, the question may arise as to whether or not a law of corresponding states can be applied for more complicated systems such as molten salts. In view of the predictive power of the law of corresponding states for the practical applications, we recognize that the law should be extended to much more complicated substances and extreme physical conditions even with sacrificing some degree of exactness. The first attempt of this extension was developed by Reiss et al. [20] for the vapor pressure, surface tension, and melting point for molten alkali halides. White and David [21] showed a corresponding-states correlation for the thermal conductivity of molten alkali nitrates. Unfortunately, there were almost no reliable experimental data for the thermal conductivity at that time. Young and O'Connell [22] proposed an empirical corresponding-states correlation for the density, viscosity, thermal conductivity, ionic diffusivity, and specific conductivity of uni-univalent molten salts. There was, however, no theoretical background for their characteristic parameters. On the other hand, Abe and Nagashima [23] applied the principle of corresponding states for the viscosity of alkali halides and their binary mixtures. They further confirmed that their universal equation is applicable to alkali nitrates, liquid metals, hydrocarbons, and rare gases.

2.2. Harada's Corresponding States Correlation for Thermal Conductivity of Uni-Univalent Molten Salts

In this section, we briefly summarize the corresponding-state theory proposed by Harada and his co-workers [24–27]. Only the outline of the theory will be introduced here: for more details the reader should consult the original papers. According to the fluctuation-dissipation theorem [28, 29], the thermal conductivity λ of isotropic liquid is expressed as

$$\lambda = \frac{1}{VkT^2} \int_0^\infty \langle \dot{A}(0)\, \dot{A}(t) \rangle \, dt \tag{2}$$

Here V is the volume of a system, k the Boltzmann's constant, T the absolute temperature, and t the time. The dynamical quantity A can be represented by the following equation.

$$A(t) = \sum_i r_{ix} \left[\frac{p_i^2}{2m_i} + \sum_{j>i} \phi_{ij} \right] \tag{3}$$

and

$$\dot{A} = \frac{dA}{dt} \tag{4}$$

where r_{ix}, p_i, and m_i indicate the x component of the position, the momentum, and the mass of the ith ion, respectively, while ϕ_{ij} is the pair interaction potential between ion i and ion j. The advantages of the above time-correlation function formulas are that (1) they are quite general in the sense that they do not depend on the details of any particular density region and (2) they give us exact expression for the thermal conductivity that can be used as a starting point for approximations [29]. Therefore, we can employ Eq. (2) as the basic formula for the evaluation of the thermal conductivity of molten salts. Since no direct methods exist for the evaluation of the time-correlation functions for dense fluids, they are usually calculated by molecular dynamics simulations [17]. It is alternatively possible to derive the corresponding states correlation by expanding the time-correlation function of the dynamical quantities for the thermal conductivity with the mass difference between the anion and the cation.

In the case of molten salts, the pair interaction between i and j ions of the separation distance r_{ij} is expressed by the sum of the core-repulsive and the Coulombic potentials as

$$\phi_{ij}(r_{ij}) = \psi_{ij} \exp(-r_{ij}/\rho_{ij}) + z_i z_j e^2 \xi / r_{ij} \tag{5}$$

where ψ_{ij} and ρ_{ij} denote the potential parameters of repulsion between i and j ions. The second term in Eq. (5) represents the Coulombic potential between i and j ions with the valences z_i and z_j, and e is the electronic charge; ξ is a parameter specific to the salt species, which incorporates the effects of the weak long-range potential and the dielectric constant.

In the case of molten alkali halides the interaction potential between anion and cation dominates the ion configuration; accordingly the values of ψ and ρ for unlike ion pair can be used for any ion pairs. The pair potential can be reduced as

$$\phi_{ij}^*(r_{ij}^*) = \phi_{ij}(r_{ij}^*)/A = \exp[-d(r_{ij}^* - 1)/\rho] + z_i z_j e^2 \xi / (A d r_{ij}^*) \tag{6}$$

$$A = \psi \exp(-d/\rho) \tag{7}$$

$$r_{ij}^* = r_{ij}/d \tag{8}$$

where d denotes a characteristic separation distance between the unlike ion pair. The momentum of the ith ion is also reduced to the following form:

$$p_i^* = p_i/(A\tilde{m}_i)^{1/2} \tag{9}$$

where $\tilde{m}_i$ is an arbitrary mass. By employing the above reduced parameters, Eqs. (6) to (9), the Hamiltonian for molten alkali halides and the Liouville operator can be also reduced. Since molten alkali halides are composed of anions and cations with different masses m_A and m_C, an unique mass which characterizes the salt is needed for obtaining the reduced form of the thermal conductivity. If we choose this characteristic mass properly, the perturbed Hamiltonian due to the mass difference disappears. The selected characteristic mass m_S can be expressed as follows:

$$m_S = [2(m_A m_C)^{1/2}/(m_A^{1/2} + m_C^{1/2})]^2 \tag{10}$$

By using this characteristic mass as a reference, the thermal conductivity of molten salts can be expressed as the sum of the thermal conductivity for the reference system of the anion and cation with a unique mass and of perturbation terms with respect to the mass difference. The final expression is written in the reduced form:

$$\lambda^*(T^*) = \lambda d^2/[k(\Lambda/m_S)^{1/2}] = \lambda_0^*(T^*) + \sum_{n=1}^{\infty} S_{2n}^*(T^*)\mu_s^{2n} \tag{11}$$

$$\mu_S = (m_A^{1/2} - m_C^{1/2})/(m_A^{1/2} + m_C^{1/2}) \tag{12}$$

Here, the reduced temperature T^* is given by the following equation:

$$T^* = kTd/(e^2\xi) \tag{13}$$

In Eq. (11), λ_0^* is the reduced thermal conductivity of the reference mass system and the second term indicates that the effect of mass difference between unlike ion pairs on the thermal conductivity is described in terms of a series of the mass difference of μ_S. S_n^* denotes the reduced coefficient of the nth term in perturbation expansion of thermal conductivity. It was found that when we select the characteristic separation distance between the unlike ion pair d appropriately, the thermodynamic properties (molar volume, vapor pressure, and surface tension [25]) and the transport properties (electrical conductivity, viscosity, and self-diffusion coefficient [26]) can be well described by the law of corresponding states. Therefore, we expect that the reduced from of the thermal conductivity of molten alkali halides along the saturation line would mainly depend on the reduced temperature T^*. The values of the parameters ψ and ρ for alkali halides are taken from Tosi and Fumi [30], and the d is calculated from the following equation [26]:

$$d/\rho = \zeta[0.4069 + 0.9075\ln(\psi/kT) + 6.042 \times 10^{-7}(\psi/kT)] \tag{14}$$

Table I. Interionic Potential Parameters ψ, ρ, ξ, and ζ for Alkali Halides [26]

	ψ (10^{-17} J)	ρ (10^{-11} m)	ξ	ζ
LiF	2.67	2.99	0.827	0.970
NaF	4.17	3.30	0.867	0.929
KF	8.39	3.38	0.893	0.931
RbF	15.5	3.28	0.905	0.947
CsF	98.5	2.82	0.867	0.960
LiCl	3.78	3.42	0.801	0.988
NaCl	20.1	3.17	0.947	0.974
KCl	28.6	3.37	1.010	0.963
RbCl	72.6	3.18	0.982	0.959
LiBr	4.41	3.53	0.845	1.005
NaBr	16.4	3.40	0.948	0.972
KBr	44.7	3.35	1.014	0.967
RbBr	64.7	3.35	0.989	0.956
LiI	1.90	4.30	0.858	0.982
NaI	9.79	3.86	0.959	0.967
KI	44.8	3.55	1.024	0.968
RbI	108.0	3.37	1.038	0.967

where the values of ξ and ζ are presented in Ref. 26. Table I lists all these potential parameters for alkali halides.

The advantages of this corresponding states correlation for uni-univalent molten salts are summarized as follows: (i) the theoretical foundation of the correlation scheme is clear; (ii) the correlation can be applied for both thermodynamic and transport properties with a consistent manner; and (iii) the characteristic parameters involved have theoretical basis and are not adjustable valuables depending on the properties. For the thermal conductivity, the validity of this corresponding states correlation has not yet been confirmed owing to lack of the reliable experimental data.

3. CORRESPONDING-STATES CORRELATION FOR THERMAL CONDUCTIVITY OF MOLTEN ALKALI HALIDES

Table II summarizes the temperature range, the accuracy, and the optimum values of coefficients in Eq. (16) for molten alkali halides obtained by our forced Rayleigh scattering experiments. In order to prove that these systematically measured thermal conductivities can be correlated with the aid of the corresponding-states law described in the previous section, Fig. 2 shows the relation between λ^* and $1/T^*$. There are 322 data

Table II. The Temperature Range, Accuracy, and Optimum Values of the Coefficients in Eq. (16) of Molten Alkali Halides Measured by the Forced Rayleigh Scattering Method

	Temperature Range (K)	Number of data points	Accuracy (%)		Optimum value of the coefficients in Eq. (16)		
			a	λ^a	λ_m $(\mathrm{W \cdot m^{-1} \cdot K^{-1}})$	b $(10^{-4}\ \mathrm{W \cdot m^{-1} \cdot K^{-2}})$	T_m (K)
LiCl	967–1321	21	±11	±20	0.626	−2.9	883
NaCl	1170–1441	34	±4	±8	0.519	−1.8	1074
KCl	1056–1335	45	±5	±8	0.389	−1.7	1043
RbCl	1046–1441	58	±6	±15	0.249	−1.1	990
CsCl	960–1360	33	±9	±18	0.209	−1.2	918
NaBr	1050–1267	44	±7	±15	0.320	−0.8	1020
KBr	1035–1245	24	±3	±7	0.218	−0.4	1007
RbBr	1031–1326	25	±7	±15	0.203	−1.1	953
CsBr	948–1314	25	±11	±19	0.149	−0.2	909
NaI	961–1099	12	±7	±11	0.206	−0.3	935
KI	965–1234	28	±5	±9	0.150	−1.0	958
RbI	963–1226	31	±6	±15	0.136	−0.7	913
CsI	937–1227	12	±10	±20	0.119	−0.8	894

[a] Accuracy of the converted thermal conductivity ($\lambda = a\rho C_p$), including estimated accuracy of ρ and C_p.

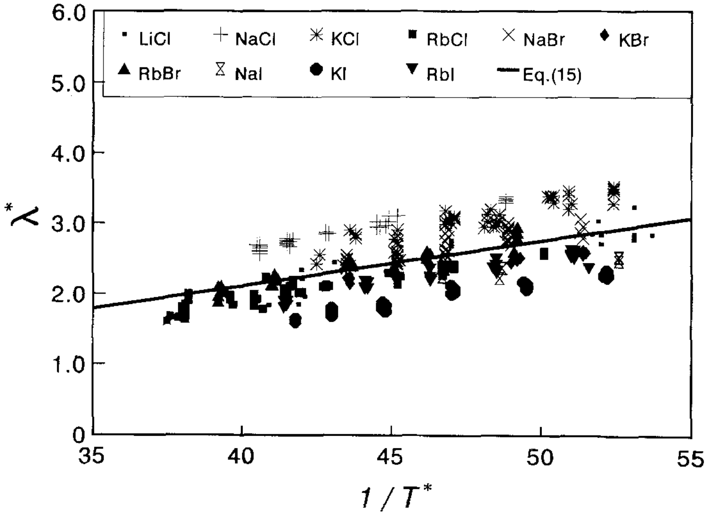


Fig. 2. Corresponding-states correlation for the thermal conductivity of molten alkali halides measured by the forced Rayleigh scattering method.

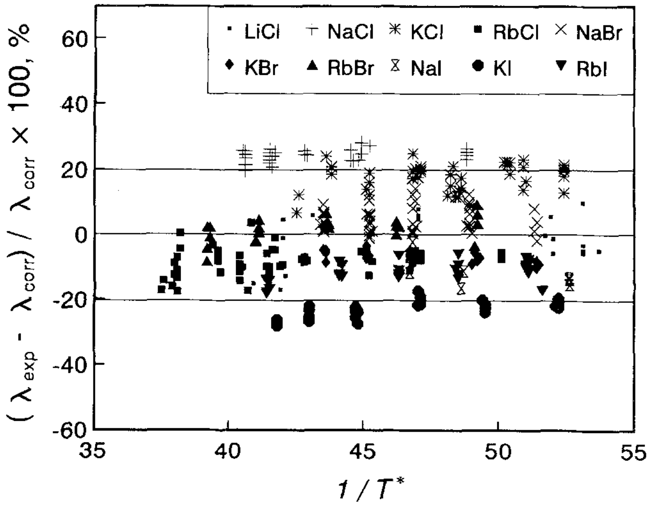


Fig. 3. Deviations of the experimental thermal conductivity data of molten alkali halides from Eq. (15).

points of 10 kinds of molten alkali halides. Since CsCl, CsBr, and CsI have different crystal structures (CsCl type) from the other alkali halides (NaCl type) so that the potential parameters are not obtained, these cesium halides are omitted in the present correlation. The relation between λ^* and $1/T^*$ can be correlated by the following equation:

$$\lambda^* = -0.473 + 6.457 \times 10^{-2}(1/T^*) \tag{15}$$

Equation (15) reproduces the entire body of experimental data within a standard deviation of 14% as depicted in Fig. 3. By taking into account both the estimated accuracies for the experimentally determined thermal conductivity and this standard deviation, it can be concluded that the thermal conductivity of molten alkali halides is adequately correlated by the corresponding-states correlation within experimental accuracy. In addition, it may be worth mentioning that recent molecular-dynamic calculations for the thermal conductivity of molten NaCl and KCl [31] reasonably agree with the present corresponding-states correlation.

4. PREDICTION OF THERMAL CONDUCTIVITY OF MOLTEN ALKALI METAL FLUORIDES AND IODIDES

It seems reasonable from the following reasons that we can predict the thermal conductivity of molten alkali fluorides and iodides, which we have not been able to determine by the forced Rayleigh scattering experiment, with the aid of the universal relationship given in Eq. (15). (i) This corresponding-states correlation for molten alkali halides has been proved to be valid for both thermodynamic and transport properties except for thermal conductivity. (ii) Equation (15) has been correlated on the basis of satisfactory number of reliable data points. (iii) The standard deviation of the experimental data from Eq. (15) is consistent with the estimated accuracy. Figure 4 shows the thermal conductivity of molten alkali fluorides calculated from Eq. (15) in the temperature range about 200 K above their melting points [32]. The predicted temperature dependencies of the thermal conductivity exhibit all weak negativities. The predicted results are also represented by the simple linear equation:

$$\lambda = \lambda_m + b(T - T_m) \tag{16}$$

The coefficients in Eq. (16) predicted by the corresponding-states correlation are listed in Table III, in which LiBr and LiI are also included. The accuracy of the predicted results is estimated to ±15 to ±20%.

Since the thermal conductivity of molten LiF is of practical importance, Fig. 5 displays the comparison of the present predicted values with

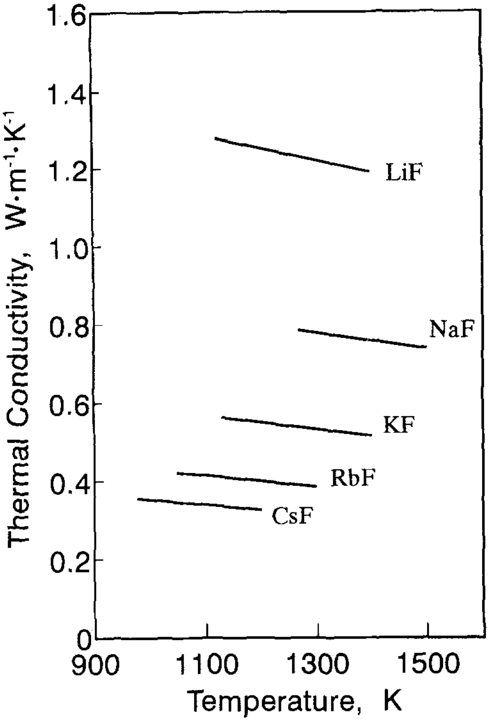


Fig. 4. Thermal conductivity of molten alkali metal fluorides estimated from the corresponding-states correlation of Eq. (15).

Table III. Coefficients in Eq. (16) Predicted by the Law of Corresponding States for Molten Alkali Halides

	λ_m $(W \cdot m^{-1} \cdot K^{-1})$	b $(10^{-4} W \cdot m^{-1} \cdot K^{-2})$	T_m (K) [32]
LiF	1.276	−3.08	1121
NaF	0.785	−2.03	1268
KF	0.563	−1.73	1131
RbF	0.419	−1.43	1068
CsF	0.356	−1.38	986
LiBr	0.458	−1.49	823
LiI	0.384	−1.28	723

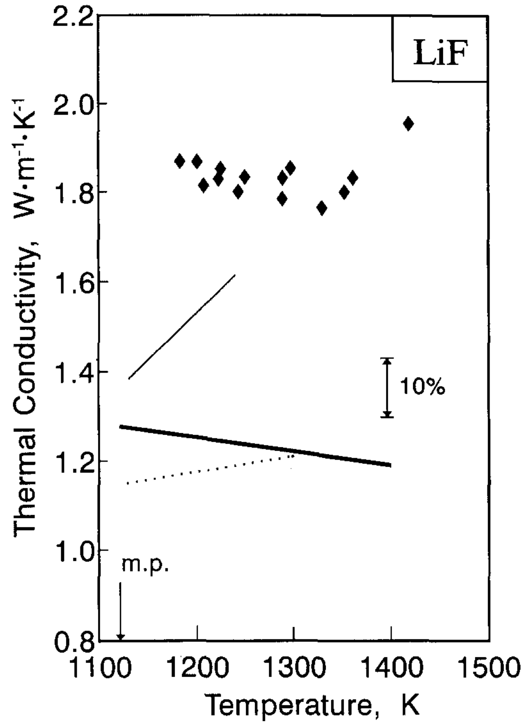


Fig. 5. Comparison of the present estimation with other experimentally determined thermal conductivity data for molten LiF: ◆, Sreenivasan [33]; ——, Smirnov et al. [34]; ----, Golyshev et al. [35]; ——, present prediction.

other previous experimentally determined data for LiF. The data of Sreenivasan [33] obtained by the quasi–steady-state concentric cylinder method are about 60% larger than our prediction. This difference may be mainly due to radiative and convective effects because of a large temperature difference ($\Delta T = 2.4$ to 13.6 K) and a large gap between cylinders (1.6 to 3.2 mm). Smirnov et al. [34] also measured the thermal conductivity of molten alkali halides systematically by the steady-state concentric cylinder method and their values agree with our prediction near the melting point, but they show a strong positive temperature dependence. The data of Golyshev et al. [35] obtained by the steady-state concentric cylinder method (molybdenum wall; gap, 2.5 mm) with a correction of about 10 to 20% for radiation agree with the present correlation within the claimed accuracy, but again, the temperature coefficient is positive.

We conclude that the thermal conductivity of molten alkali halides can be adequately correlated by the principle of corresponding states proposed by Harada. The prediction scheme for the thermophysical properties described in the present paper, which is a combination of systematically conducted experiment with correlation procedure based on a rigorouos theory, will become more important under extreme conditions. Further studies will be needed for mixtures and molten salts other than uni-univalent.

ACKNOWLEDGMENT

The authors are grateful to Professor M. Harada of Institute of Atomic Energy, Kyoto University, for his contributions to this work in many discussions.

REFERENCES

1. Y. Nagasaka, N. Nakazawa, and A. Nagashima, *Int. J. Thermophys.* **13**:555 (1992).
2. N. Nakazawa, Y. Nagasaka, and Nagashima, *Int. J. Thermophys.* **13**:753 (1992).
3. N. Nakazawa, Y. Nagasaka, and A. Nagashima, *Int. J. Thermophys.* **13**:763 (1992).
4. T. Hatakeyama, K. Kadoya, M. Okuda, Y. Nagasaka, and A. Nagashima, *Trans. JSME* **B53**:1590 (1987).
5. Y. Nagasaka, T. Hatakeyama, and A. Nagashim, *Trans. JSME* **B53**:2545 (1987).
6. Y. Nagasaka, T. Hatakeyama, M. Okuda, and A. Nagashima, *Rev. Sci. Instrum.* **59**:1156 (1988).
7. T. Hatakeyama, Y. Miyahashi, M. Okuda, Y. Nagasaka, and A. Nagashima, *Trans. JSME* **B54**:1131 (1988).
8. Y. Nagasaka and A. Nagashima, *Int. J. Thermophys.* **9**:923 (1988).
9. N. Nakazawa, M. Akabori, Y. Nagasaka, and A. Nagashima, *Trans. JSME* **B56**:1467 (1990).
10. Y. Nagasaka and A. Nagashima, *Int. J. Thermophys.* **12**:769 (1991).
11. J. B. Kesseli and D. E. Lacy, *Proc. 22nd IECEC* (1987), p. 162.
12. Y. Nagasaka and A. Nagashima, *Proc. 11th Jap. Symp. Thermophys. Prop.* (1990), p. 275.
13. J. O. Hirschfelder, C. F. Curtiss, and R. B. Bird, *Molecular Theory of Gases and Liquids* (Wiley, New York, 1964).
14. J. Kestin, S. T. Ro, and W. A. Wakeham, *Physica* **58**:165 (1972).
15. J. Kestin, S. T. Ro, and W. A. Wakeham, *J. Chem. Phys.* **56**:5837 (1972).
16. Y. Abe, J. Kestin, H. E. Khalifa, and W. A. Wakeham, *Ber. Bunsenges. Phys. Chem.* **83**:271 (1979).
17. G. C. Maitland, M. Rigby, E. B. Smith, and W. A. Wakeham, *Intermolecular Forces; Their Origin and Determination* (Clarendon Press, Oxford, 1981), pp. 564–567.
18. M. J. Assael, J. H. Dymond, M. Papadaki, and P. M. Patterson, *Int. J. Thermophys.* **13**:269 (1992).
19. S. F. Y. Li, R. D. Trengove, W. A. Wakeham, and M. Zalaf, *Int. J. Thermophys.* **7**:273 (1986).
20. H. Reiss, S. W. Mayer, and J. L. Katz, *J. Chem. Phys.* **35**:820 (1961).
21. L. R. White and H. T. Davis, *J. Chem. Phys.* **47**:5433 (1967).
22. R. E. Young and J. P. O'Connell, *Ind. Eng. Chem.* **10**:410 (1971).

23. Y. Abe and A. Nagashima, *J. Chem. Phys.* **75**:3977 (1981).
24. M. Harada, A. Yamanaka, M. Tanigaki, and Y. Tada, *J. Chem. Phys.* **76**:1550 (1982).
25. M. Harada, M. Tanigaki, and Y. Tada, *Ind. Eng. Chem. Fundam.* **22**:116 (1983).
26. Y. Tada, S. Hiraoka, T. Uemura, and M. Harada, *Ind. Eng. Chem. Res.* **27**:1042 (1988).
27. M. Harada, A. Shioi, and T. Miura, *Proc. 11th Jap. Symp. Thermophys. Prop.* (1990), p. 279.
28. J. P. Hansen and I. R. McDonald, *Theory of Simple Liquids*, 2nd ed. (Academic Press, New York, 1986).
29. D. A. McQuarrie, *Statistical Mechanics* (Harper & Row, New York, 1976), pp. 467–591.
30. M. P. Tosi and F. G. Fumi, *J. Phys. Chem. Solids* **25**:45 (1964).
31. P. Sindzingre and M. J. Gillan, *J. Phys. Condens. Matter* **2**:7033 (1990).
32. Japan Society of Thermophysical Properties (eds.), *Thermophysical Properties Handbook* (Yokendo, Tokyo, 1990).
33. K. Sreenivasan, Ph.D. thesis (University of Pennsylvania, 1967).
34. M. V. Smirnov, V. A. Khokhlov, and E. S. Filatov, *Electrochim. Acta* **32**:1019 (1987).
35. V. D. Golyshev, M. A. Gonik, V. A. Petrov, and Yu. M. Putilin, *Teplofiz. Vys. Temp.* **21**:899 (1983).