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Thermal Conductivity of Ionic Liquids

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Abstract

Ionic liquids (ILs) have attracted great attention as green solvents, heat carriers, and electrolytes. They can be obtained with specific thermophysical properties and functions by changing the kind of species of cations and anions. Knowledge of the fundamental thermophysical properties of ILs, such as their densities, viscosities, and thermal conductivities, is needed to design ILs with desirable thermophysical properties. In this chapter, we will review the various measurement results for the thermal conductivities of the pure components of ILs and methods for predicting the thermal conductivity of an IL, which are based on its structure and physical properties, by conducting correlations between these parameters. In the recent years, the thermal conductivities of IoNano fluids, which comprise of nanoparticles dispersed in an IL, have attracted great attention. Therefore, we will review the unique thermal conductivities of IoNano fluids.

Keywords: thermal conductivity, ILs, nanofluids, correlation, prediction

1. Introduction

ILs are salts that exist in the liquid at ambient temperature, and their characteristics include nonvolatility, flame retardancy, high ionic conductivity, and exhibiting a liquid state at a wide temperature range, among others. Since Wilkes et al. discovered a water-stable IL system using BF_4^- , a nonchloroaluminate anion, in 1992 [1], the study of ILs has drastically increased, and ILs have drawn great attention for a variety of applications, such as an alternative reaction solvent to replace organic solvents, as thermal medium, and as electrolyte in batteries. **Figure 1** shows the cations and anions that are typically used in ILs. There are infinite potential combinations of cations and anions and, additionally, various chemical modifications can be made to the organic moieties of the ions. Thus, there is a possibility of creating ILs tailored for specific purposes; for this reason, ILs are also called “designer solvents.”

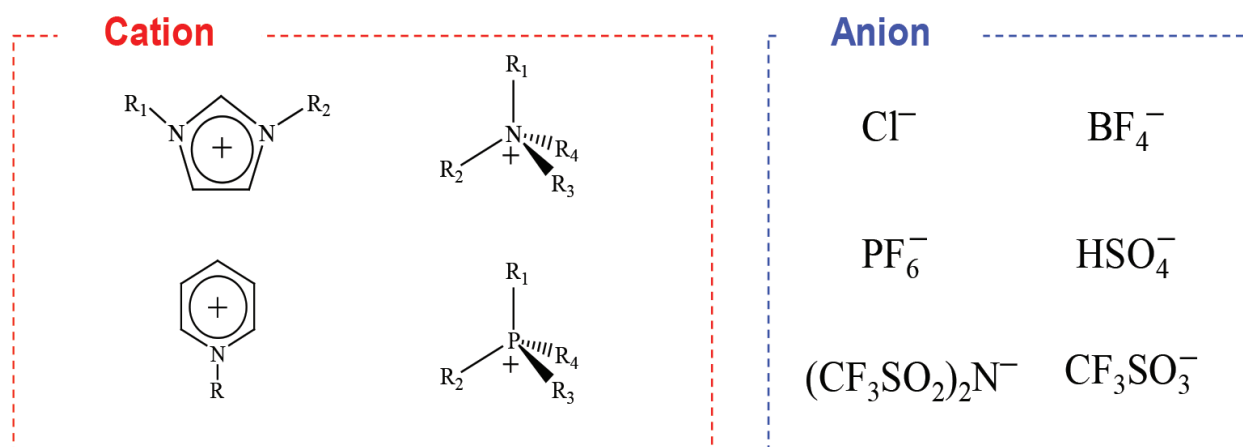


Figure 1. Cations and anions typically used in ILs.

A variety of ILs have been synthesized, many of which have melting points much greater than room temperature. For this reason, ILs are broadly defined as the ones having melting point below 100°C. Numerous ILs have been created; however, the prediction of their thermophysical properties such as the thermal conductivity on the basis of the molecular structure would be useful to develop novel ILs. In this chapter, we review the various measurement results for the thermal conductivities of the pure components of IL, the progress made toward the development of effective methods of correlating and predicting the thermal conductivities, and the thermal conductivities of IoNano fluids, which, in the recent years, have attracted great attention as high-temperature heat mediums.

2. Thermal conductivities of the pure components of ILs

2.1. Investigating the thermal conductivities of ILs

The thermal conductivity of an IL was first reported by Valkenburg et al. [2]. The known thermophysical properties of ILs, including their thermal conductivities, are summarized in ILs database of NIST [3]. During the early stages of IL research, since it was necessary to synthesize an IL to measure its thermophysical properties, there was less number of researches. However, once ILs became commercially available, the amount of information reported about the thermophysical properties of ILs increased drastically. However, the number of the reported thermal conductivity values is fewer than the other thermophysical properties, and it has been reported that two or three component systems have almost no thermal conductivity. Only four research groups have reported the thermal conductivities of binary mixtures [4–7], and only two groups have reported on ternary mixtures containing ILs [6, 8]. The reported thermal conductivity values for various ILs range from 0.106 [4] to 0.238 W/(m K) [7] at 298.15 K (**Figure 2**) and many reported values are around 0.15 W/(m K) [3, 9] (**Figure 3**). The thermal conductivities of ILs were found to be about the same as those of organic solvents, such as methanol and toluene.

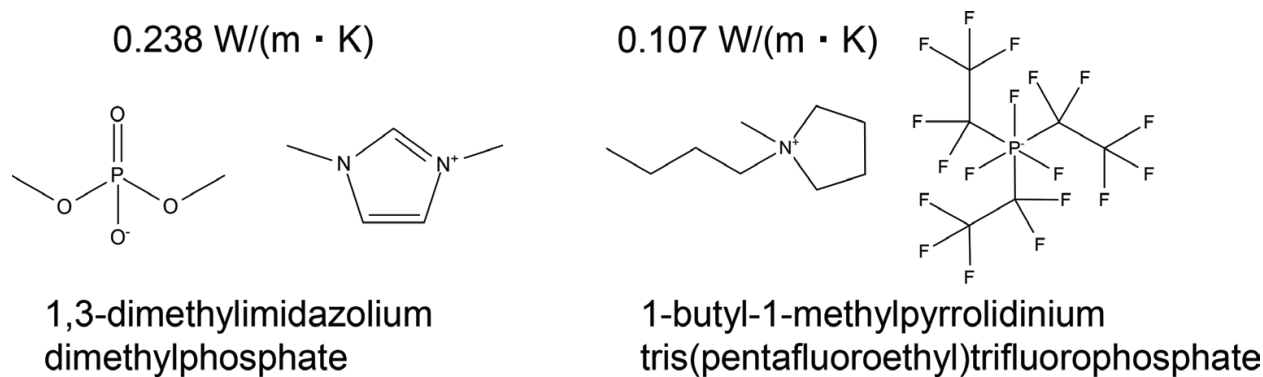


Figure 2. The ILs with the highest thermal conductivity (left) [7] and the lowest thermal conductivity (right) [4] at 298.15 K that have been reported to date.

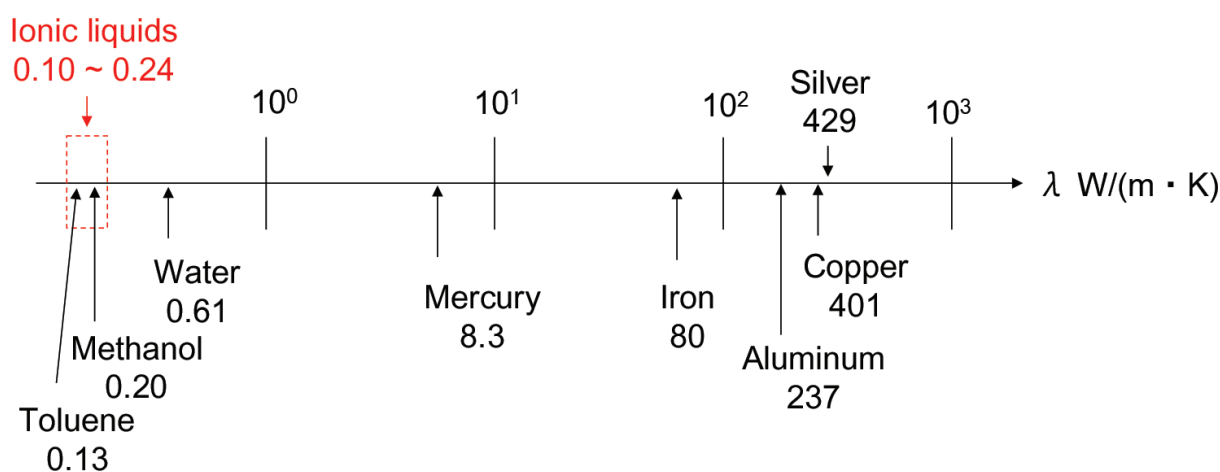


Figure 3. Thermal conductivities of various substances at 298.15 K [3, 9].

It is well known that the thermophysical properties of ILs are influenced by impurities [10]. For example, it has been reported that the viscosity of an IL decreases by about 1% with the addition of only 100 ppm of water [11]. The influence of trace water and chloride on the thermal conductivity of an IL has been reported to be relatively small compared to that on the other thermophysical properties, such as viscosity and density, as reported by Rooney et al. [4] (**Figure 4**). Our group has also investigated the effect of chloride ions on the thermal conductivities of IL [5] by measuring and comparing the thermal conductivity of 1-butyl-3-methylimidazolium tetrafluoroborate with different chloride concentrations of 421 and 4580 ppm; the difference between the thermal conductivities of these ILs was 1.2%.

We additionally investigated the influence of the alkyl chain length on thermal conductivity of ILs [12–16]. **Figure 5** shows the relation between alkyl chain length and thermal conductivity at 293 K. The thermal conductivity of *n*-alkanes was calculated by REFPROP 9.0 [17]. The results indicated that the alkyl chain length does not significantly affect the thermal conductivity. This observation differed from the findings of other studies regarding the thermal conductivities of *n*-alkanes [18] and the influence of the viscosity of the IL [19].

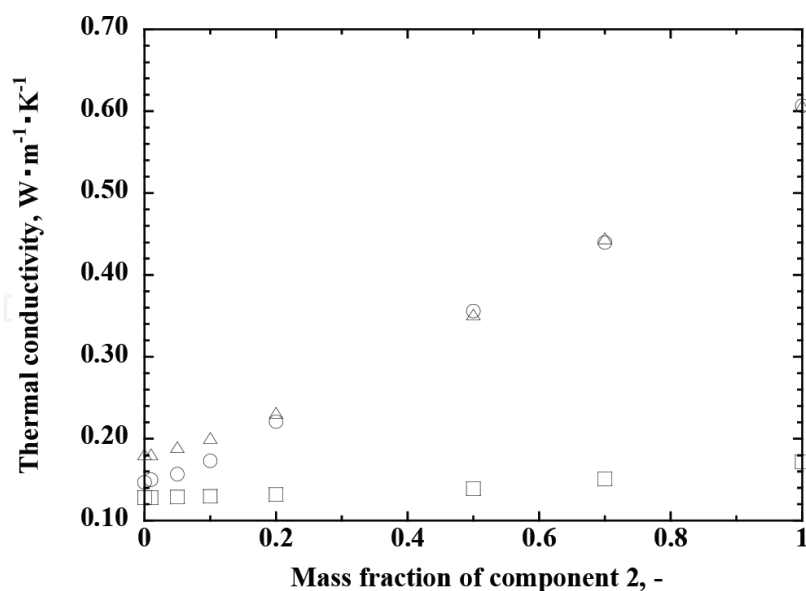


Figure 4. Effects of water and chloride on the thermal conductivities of ILs [4]: °, 1-butyl-3-methylimidazolium trifluoromethanesulfonate (1) + water (2); Δ, 1-ethyl-3-methylimidazolium ethylsulfate (1) + water (2); □, 1-hexyl-3-methylimidazolium bis-(trifluoromethylsulfonyl)imides (1) + 1-hexyl-3-methylimidazolium chloride (2).

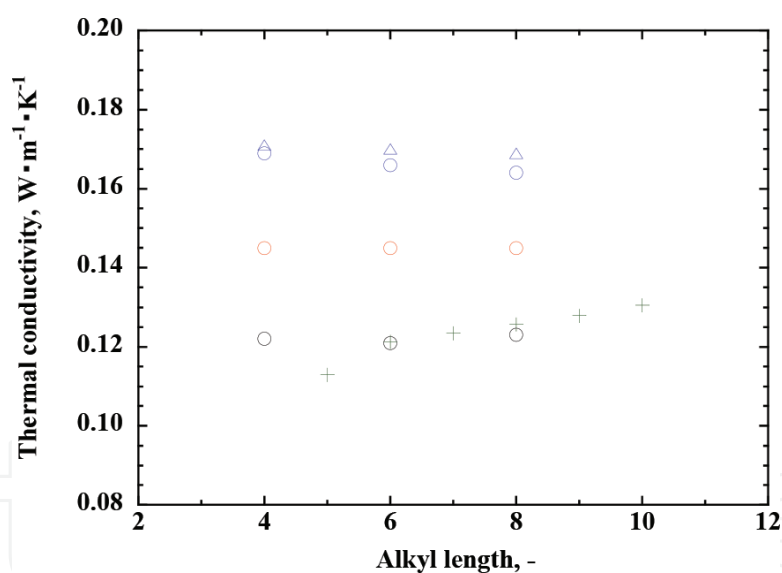


Figure 5. Relationship between the alkyl chain length and the thermal conductivities of various ILs at 293 K: 1-alkyl-3-methylimidazolium hexafluorophosphates; 1-alkyl-3-methylimidazolium tetrafluoroborates; 1-alkyl-3-methylimidazolium bis[(trifluoromethyl)sulfonyl] amides; *N*-alkylpyridinium tetrafluoroborates; and *n*-alkanes [12–17].

The temperature and pressure dependence of the thermal conductivity of ILs has been investigated [12, 14]. **Figure 6** shows a comparison of the temperature dependence of thermal conductivities of various ILs and organic solvents. The results showed that the temperature dependence of the thermal conductivity of each IL is very small in comparison with those of toluene and benzene. This is because the temperature dependence of the density of an IL is small. **Figure 7** shows a comparison of the thermal conductivities of various ILs and organic

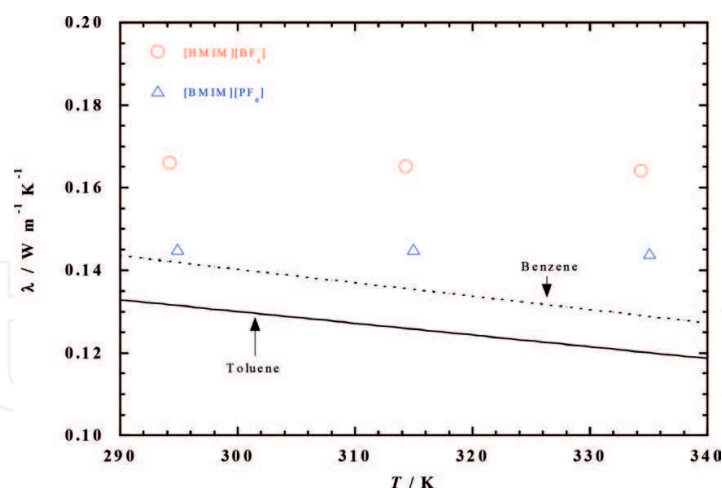


Figure 6. Temperature dependences of the thermal conductivities of various ILs and organic solvents.

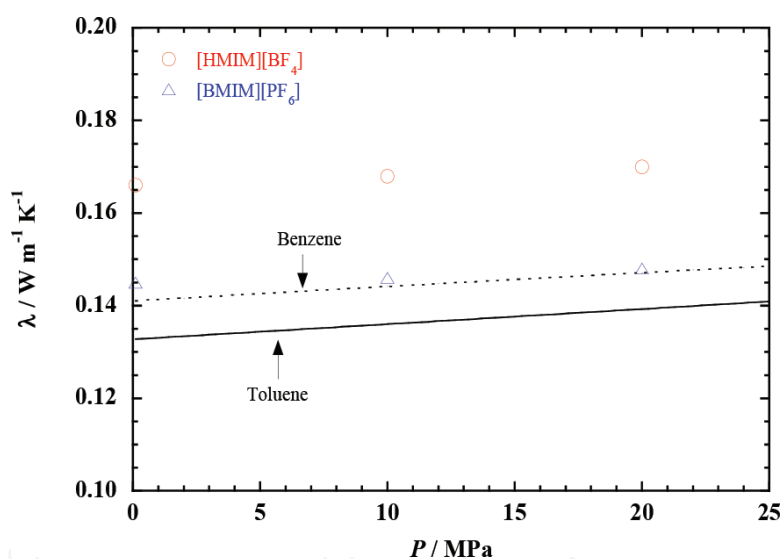


Figure 7. Pressure dependences of the thermal conductivities of various ILs and organic solvents.

solvents as functions of the pressure. It was found that the pressure dependence of the thermal conductivity of each IL is very small (within 20 MPa) in comparison with those of toluene and benzene. This is because the pressure dependence of the density of an IL is small.

2.2. Prediction and correlation of the thermal conductivity of an IL with its physical properties

A variety of ILs has been created as “designer solvents.” Therefore, the prediction of the thermal conductivity of an IL based on its structure and other physical properties would be very useful for designing novel ILs. So far, an empirical-prediction method [13, 20, 21], group-contribution method [22–24], quantitative structure-property relationship method [25], prediction

method using a neural network [26, 27], and many other methods [28–35] have been proposed for the prediction of the thermal conductivities of ILs. In this section, the empirical-prediction method based on other physical properties and prediction method using group-contribution method are introduced.

We proposed a correlation equation based on the Mohanty equation [36] to describe the relation between the thermal conductivity and viscosity of an IL [13]:

$$\log (M \lambda / \eta) = 1.9596 - 0.004499 M, \quad (1)$$

where M is the molar mass, λ is the thermal conductivity, and η is the viscosity of the IL. However, Eq. (1) was obtained by assuming that the molar mass of the IL is two times larger than the actual value. Fröba et al. proposed another correlation method and described the relation between the thermal conductivity and density of an IL as follows [20]:

$$\lambda M \rho = 0.1244M + 18.84, \quad (2)$$

where M is the molar mass, λ is the thermal conductivity, and ρ is the density of the IL. From 45 data points for 36 ILs, the standard deviation and mean absolute deviation of the experimental data relative to the predictions were 7.8 and 6.5%, respectively at 293.15 K and atmospheric pressure. Koller et al. extended this model to calculate the thermal conductivity at any temperature as follows [21]:

$$\lambda(T) = \left(0.0960 + \frac{21.43}{M \rho(T_{\text{ref}})} \right) \left(\frac{\rho(T)}{\rho(T_{\text{ref}})} \right)^{0.826} \quad (3)$$

From 469 data points for 53 different ILs, the mean absolute deviation and root-mean-square deviation of the experimental data relative to the values predicted using Eq. (3) were 4.81 and 6.32%, respectively.

As mentioned above, novel ILs are synthesized and many new substances will be synthesized in the near future. Therefore, it would be very useful to predict the thermophysical properties from the structure of an IL.

Gardas and Coutinho proposed predicting the thermal conductivity of an IL by the group-contribution method [22]. When this method was proposed, few reports were available regarding the thermal conductivities of ILs from which a prediction equation could be derived. However, the following equation was derived to correlate the thermal conductivity to the specific cation and anion and the length of the alkyl chain of the cation:

$$\lambda = A_{\lambda} - B_{\lambda} T, \quad (4)$$

where T is the temperature in K and A_{λ} and B_{λ} are the fitting parameters that can be obtained from the group-contribution approach. These parameters can be derived as follows:

$$A_{\lambda} = \sum_{i=1}^k n_i a_{i,\lambda} \quad B_{\lambda} = \sum_{i=1}^k n_i b_{i,\lambda} , \quad (5)$$

where n_i is the number of groups of type i , k is the total number of different groups in the molecule, and the parameters $a_{i,\lambda}$ and $b_{i,\lambda}$ estimated for the ILs studied are given in **Table 1**.

Wu et al. proposed a prediction formula in which the group is divided finely so that more ILs can be predicted based on their structures [24]:

$$\lambda = \sum_{i=0}^2 a_i \left(\sum_{j=1}^k n_j \Delta \lambda_{0,j} \right)^i [1 + k_0(1 - T_r)^{2/3}] , \quad (6)$$

where $T_r = T/T_c$ is the reduced temperature, T_c is the critical temperature in K (which is easy to obtain for many ILs by applying the group-contribution method proposed by Valderrama group [37]), k_0 is the temperature-independent constant, n_j is the number of groups of type j , k is the total number of different groups in the molecule, and the parameters a_i and $\Delta \lambda_{0,j}$ are estimated using **Table 2**. The calculated values and those reported in the literature were in agreement for 286 data points for 36 ILs with an average absolute deviation of 1.66%.

Species	$a_{i,\lambda}$	$b_{i,\lambda} \text{ (K}^{-1}\text{)}$
<i>Cations</i>		
1,3-dimethylimidazolium (+)	0.1356	1.564×10^{-5}
1,1-dimethylpyrrolidinium (+)	0.1325	1.668×10^{-5}
Tetramethylphosphonium (+)	0.1503	3.230×10^{-5}
<i>Anions</i>		
PF_6^-	0.0173	9.088×10^{-6}
BF_4^-	0.0874	8.828×10^{-5}
Tf_2N^-	0.0039	2.325×10^{-5}
CF_3SO_3^-	0.0305	5.284×10^{-5}
EtSO_4^-	0.0700	6.552×10^{-5}
Cl^-	0.0166	1.000×10^{-5}
<i>Groups</i>		
CH_2	0.0010	2.586×10^{-6}
CH_3	0.0042	7.768×10^{-6}

Table 1. Group-contribution parameters, a_{ij} and b_{ij} for Eq. (5) at 293–390 K [22].

Without rings		With rings		Coefficients	Values
Group	Values of $\Delta\lambda_{0j}$	Group	Values of $\Delta\lambda_{0j}$		
-CH ₃	-2.66388	>C=	-0.90423	k_0	4.05711
-CH ₂ - (with ammonium-)	0.60575	=CH-	-1.32204	a_0	0.03647
-CH ₂ - (with phosphonium-)	0.71023	-CH ₂ -	-0.49677	a_1	-0.00097
-CH ₂ - (with others)	0.68295	>CH-	2.20488	a_2	0.00003
>C<	-0.36265	-NH-	2.53635		
>CH	5.29795	-N=	0.83850		
-CN	1.51636	>N- / >N<†	-3.22547		
-COO-	1.51468				
-COOH	-10.35869				
>N- / >N<† / -N-	-1.24392				
Without rings					
Group	Values of $\Delta\lambda_{0j}$				
-NH ₂	-3.20298				
-SO ₂	6.88560				
-O- / [-O] ⁻	0.00273				
-OH	-1.33002				
-F	1.69009				
-Cl	-6.92175				
-B	-3.97751				
-P	-2.50897				

Table 2. The coefficient values obtained by the group-contribution method using Eq. (6) [24].

Lazzús proposed a prediction equation to calculate the thermal conductivity of an IL as a function of the temperature and pressure [23]. The experimentally measured thermal conductivities of 41 ILs (including 400 experimental data points) in the range of 0.1–0.22 W/(m K) were used to design the proposed method for the temperature range of 273–390 K and the pressure range of 0.1–20 MPa. The results showed that the proposed group-contribution method can be used to accurately predict the thermal conductivity of an IL as a function of the temperature and pressure present with lower deviations between the predicted and actual values, including an average absolute relative deviation (AARD) of less than 1.90% and an R^2 of 0.9879 for the correlation dataset and AARD of less than 2.33% and R^2 of 0.9754 for the prediction dataset.

3. Thermal conductivities of IoNano fluids

Nanofluids are dispersions of nanoparticles and are expected to be applied as heat transfer media and used for inkjet printing. They were first reported by Choi [38] in 1995. Since then, it has been reported that the thermal conductivity of ethylene glycol is improved by 40% when 0.3 vol% of 10-nm Cu nanoparticles are dispersed in ethylene glycol [39]. A dispersion of nanoparticles in an IL is called an “IoNano fluid,” as first dubbed by Castro et al. in 2010 in a

study of the thermal conductivities of nanofluids with carbon nanotubes dispersed in ILs [40, 41]. As described above, the thermal conductivities of ILs have been reported to be about the same as those of ethanol and methanol, which is not high. For this reason, it is expected that the thermal conductivity of an IL can be enhanced by dispersing nanoparticles in it.

It is known that the thermal conductivity of a nanofluid agrees well with the Hamilton-Crosser model [42]:

$$\frac{k_{eff}}{k_0} = \frac{k_p + (n-1)k_0 + (n-1)(k_p - k_0)\phi_p}{k_p + (n-1)k_0 - (k_p - k_0)\phi_p} \quad (7)$$

where k_0 , k_{eff} and k_p are the thermal conductivities of the dispersion medium, nanofluid, and dispersoid, respectively, in W/(m K), n is the particle shape parameter [–], and ϕ_p is the volume fraction of the dispersoid (also unitless).

Figure 8 shows the calculated enhancement of the thermal conductivity rate based on the Hamilton-Crosser model when silver, alumina, and barium titanate spherical nanoparticles are used as the dispersoid in 1-butyl-3-methylimidazolium tetrafluoroborate ([BMIM][BF₄]) as a representative IL. **Table 3** shows the thermal conductivity of each dispersoid tested. The results show that even with dispersoids with significantly different thermal conductivities, the resulting enhancement in the thermal conductivity does not vary significantly when the volume fraction is about 15% ($n = 3$). This suggests that, for the case of dispersing spherical nanoparticles in an IL, the thermal conductivity of the dispersoid does not significantly affect the thermal conductivity of the obtained nanofluid. Thus, to further increase the thermal conductivity of an IL using spherical nanoparticles, the volume fraction of the dispersoid must be increased; however, this introduces a problem of flowability. Therefore, while considering the use of a nanofluid as a heating medium, it is necessary to increase the thermal conductivity using the smallest possible volume fraction of nanoparticles. To remarkably increase the thermal conductivity enhancement rate with a low nanoparticle concentration, it is necessary

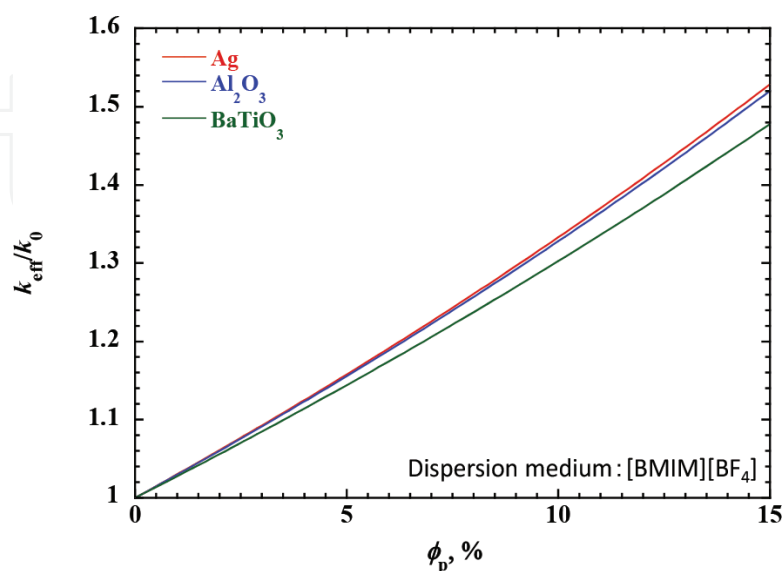


Figure 8. Relationship between the enhancement of the thermal conductivity and the volume fraction of nanoparticles based on the Hamilton-Crosser model at 298 K.

	k_p , W/(m K)
Ag	428
Al ₂ O ₃	36
BaTiO ₃	6

Table 3. Thermal conductivities of dispersoids.

to increase the particle diameter, n . Increasing n increases the slope; this is possible by using a material with a large aspect ratio.

Wang et al. reported that the thermal conductivity of an IoNano fluid of carbon nanotubes dispersed in 1-hexyl-3-methylimidazolium tetrafluoroborate (**Figure 9**) [43] was improved by more than 10% by adding only 0.03 wt% of graphene. In addition, the results showed that the thermal conductivities of the pure IL component (the dispersion medium) and the IoNano fluid increase with an increase in temperature. **Figure 6** shows the temperature dependence of the thermal conductivities of benzene, toluene, and 1-hexyl-3-methylimidazolium tetrafluoroborate. Usually, the thermal conductivity of a liquid decreases with increasing temperature; it is known that this holds true for ILs and that the temperature dependence of an IL is smaller than those of organic solvents including benzene and toluene. The thermal conductivity values of [HMIM][BF₄] reported by other researchers [40] indicate that the thermal conductivity gradually decreases as the temperature increases. Thus, it is conceivable that the thermal conductivity value reported by Wang et al. was influenced by convection. Assael et al. studied the reported thermal conductivity values of the nanofluid in detail and pointed out that there are many reported cases that are affected by convection [44, 45]. Based on this finding, it is necessary to carefully consider the previously reported thermal conductivity values for nanofluids because they may include the influence of convection.

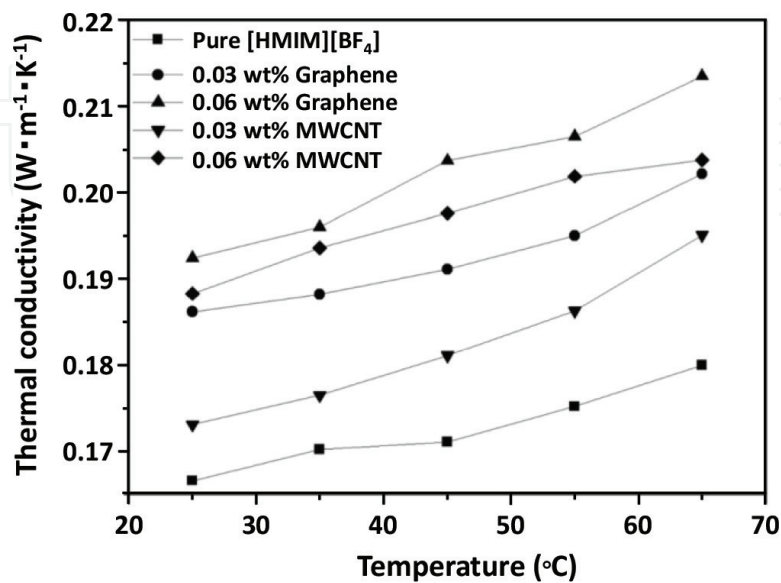


Figure 9. Thermal conductivities of IoNano fluids and [HMIM][BF₄] as functions of temperature [43].

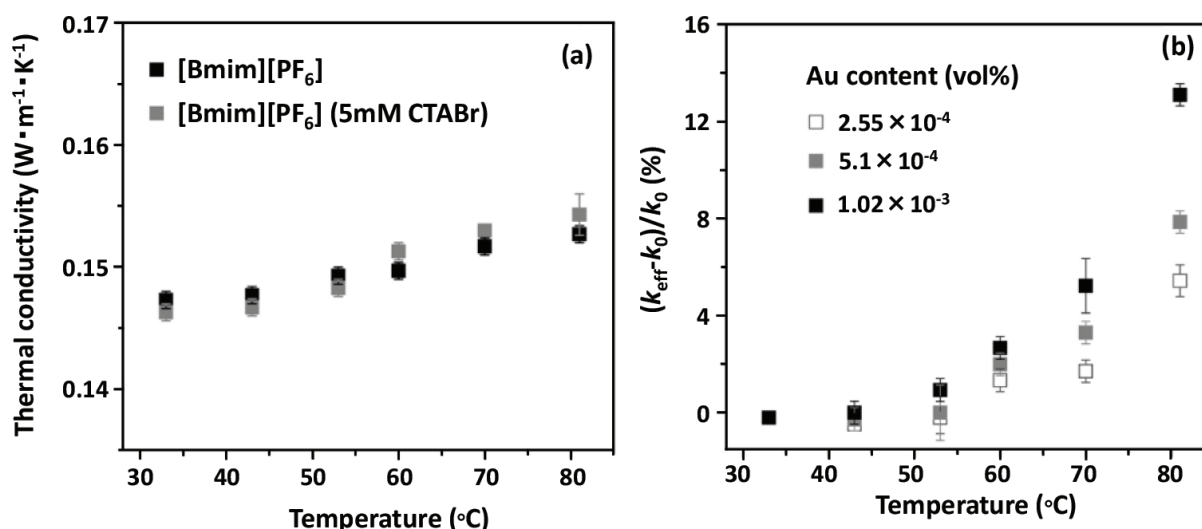


Figure 10. Thermal conductivity of [BMIM][PF₆] over a range of temperatures (a) with and without 5 mmol CTABr and (b) the thermal conductivity enhancement in Au/[BMIM][PF₆] nanofluids due to various Au concentrations [46].

Wang et al. measured the thermal conductivity of Au/[BMIM][PF₆] nanofluids of Au nanoparticles dispersed in a typical IL 1-butyl-3-methylimidazolium hexafluorophosphate medium ([BMIM][PF₆]) [46]. The thermal conductivities of the Au/[Bmim][PF₆] nanofluids stabilized by cetyltrimethylammonium bromide (CTABr) increases with an increase in the temperature and the rate of the increase also increases with an increase in the temperature (Figure 10). However, it was also seen that the thermal conductivity of [BMIM][PF₆] increases with an increase in the temperature (Figure 10(a)). In the thermal conductivity values of [BMIM][PF₆] reported by other researchers [13, 40], the thermal conductivity gradually decreases as the temperature increases; thus, the thermal conductivity measured by Wang et al. likely contains the influence of convection.

Adriana et al. reported the thermal conductivities of IoNano fluids, such as Al₂O₃/[C₄ mpyrr][NTf₂] and MWCNT/[C₄ mim][(CF₃SO₂)₂N]. The results showed that the enhancement rate of the thermal conductivity is almost the same as that predicted based on the Hamilton-Crosser equation; no significant enhancement in thermal conductivity has been confirmed in these systems [47].

França et al. reported that the thermal conductivity improved from 4 to 26% between MWCNT/[C₄mim][CF₃SO₂]₂N and MWCNT/[C₂mim][EtSO₄] [48]. Although the measurements were obtained by commercial equipment, KD2 Pro, which introduces a large measurement uncertainty, the thermal conductivity was found to gradually decrease with an increase in the temperature in the pure IL component and IoNano fluid; this indicates that the measured values were almost not affected by convection.

4. Summary

In this study, the thermal conductivities of ILs and IoNano fluids were reviewed. The thermal conductivity is one of the physical properties of ILs for which few studies have been reported. The thermal conductivity of an IL is difficult to measure and the observed data are often

influenced by convection. Although the prediction and the correlation method have been proposed and examined, the results are not useful unless they are based on precisely measured values. Therefore, it is necessary to precisely measure and publish more data regarding the thermal conductivities of ILs and IoNano fluids.

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