Measurements of Ionic Liquids Thermal Conductivity and Thermal Diffusivity

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Abstract

Thermal conductivity and thermal diffusivity of ionic liquids (ILs) are investigated in this work. A hot disk method for ILs thermal conductivity and thermal diffusivity measurement is utilized. Firstly, the thermal conductivity of water is measured to check the reliability of the hot disk method. In addition, the thermal conductivity of pure ILs, including BmimBF4, BmimPF6, OmimCl, BmimFeCl4, and OmimFeCl4, is measured. By comparison with thermal conductivity values of water, BmimBF4, and BmimPF6 in the literatures, it is found that the thermal conductivity values of ILs using hot disk method has high reliability. Therefore, the hot disk method is utilized for thermal conductivity measurement of ILs in this work. The experimental results also show that all the average thermal conductivity values of the 5 pure ILs are no more than 0.1898 Wm⁻¹K⁻¹, which is much lower than the average measured thermal conductivity of water, namely 0.6033 Wm⁻¹K⁻¹. Effect of nanoparticles (NPs) on thermal conductivity of ILs is also investigated. It is shown that the thermal conductivity of BmimBF4 does not change significantly in the presence of Fe₂O₃ NPs. However, the thermal conductivity of BmimPF6 decreases somewhat in the presence of R711 NPs. In addition, the thermal diffusivity of pure ILs, including BmimBF4, BmimPF6, BmimFeCl4, OmimCl and OmimFeCl4, is measured. All the average thermal diffusivity values of the ILs of BmimBF4, BmimPF6, BmimFeCl4, OmimCl and OmimFeCl4, are no more than 0.1185 mm²s⁻¹. The thermal

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diffusivity of water is 0.143 mm²s⁻¹ in the literature. It illustrates that the ILs also have a better thermal property than water for energy storage. ILs may be utilized as novel materials for energy storage. Effect of NPs on thermal diffusivity of ILs is also investigated. The results are similar to how NPs influence thermal conductivity of BmimBF₄ and BmimPF₆. In the presence of Fe₂O₃ NPs, thermal diffusivity of BmimBF₄ does not change significantly. However, in the presence of R711 NPs, thermal diffusivity of BmimPF₆ decreases somewhat.

Keywords: Ionic liquid, nanoparticle, thermal conductivity, thermal diffusivity, thermal property, hot disk.

Introduction

Ionic liquids (ILs) are environmentally friendly materials. ILs have many excellent properties, which currently are hot research topics. Due to the excellent properties, ILs have been utilized as novel and promising materials in many research fields. For example, ILs can be utilized in energy conversion and energy storage, including solar cells [1-7], batteries [8-16], and supercapacitors [17-28].

Thermal properties of ILs are important.[29-31] Thermal conductivity is the ability of a material to conduct heat, which is an important parameter for utilization of ILs [32-34]. Thermal conductivity and thermal diffusivity are crucial for application of ILs in real energy conversion and energy storage processes. However, there is scant literature that can be found for thermal conductivity measurement of ILs. Frez et al. utilized the transient grating method for the thermal conductivity measurement of 7 ILs [35]. In the work of Ge *et al.*, the transient hot-wire method was utilized to measure the thermal conductivity of 11 pure ILs [36]. Fröba *et al.* measured the thermal conductivity of ILs using parallel-plate method [37]. The thermal conductivity data of ILs are limited in the current literatures. Therefore, the thermal conductivity of ILs is measured using a hot disk method in this work. In addition, the effect of nanoparticles (NPs) on the thermal conductivity of ILs is investigated.

Thermal diffusivity is the ability of a material to conduct thermal energy relative to its ability to store thermal energy [38]. The definition of thermal diffusivity α is as follows:

$$\alpha = \frac{\kappa}{\rho c_p} \tag{1}$$

where α , κ , ρ , and c_p are thermal diffusivity, thermal conductivity, mass density, and specific heat capacity, respectively. Thermal diffusivity is also an important parameter for utilization of ILs. However, the thermal diffusivity measurement of ILs is extremely scant. Frez *et al.* [35] are the only researchers who experimentally measured the thermal diffusivity of pure ILs according to our own knowledge. In the

work of Frez *et al.*, transient grating technique was utilized for measurement of pure ILs thermal diffusivity [35]. We found the thermal diffusivity data of ILs are rarely known in the current literatures. Therefore, in this work the thermal diffusivity of ILs is measured using hot disk method. In addition, the effect of NPs on thermal diffusivity of ILs is investigated.

Theory

The theory of hot disk method is introduced in this part based on the Introduction Manual of Hot Disk Thermal Constants Analyser [39]. The hot disk is electrically heated for the thermal conductivity and thermal diffusivity measurement. The resistance increase can be described as a function of time as follows:

$$R(t) = R_0 \{ 1 + \beta [\Delta T_i + \Delta T_a(\tau)] \}$$
 (2)

where R(t) is disk resistance at time t, R_0 is disk resistance at time t = 0, β is temperature coefficient of resistivity, ΔT_i is the constant temperature difference, and $\Delta T_a(\tau)$ is time dependent temperature increase [39]. The time dependent temperature increase can be obtained by the theory as follows:

$$\Delta T_a(\tau) = \frac{P_0}{\pi^{3/2} \cdot a \cdot \kappa} \cdot D(\tau) \tag{3}$$

where P_0 is the total power output of the sensor, α is the overall radius of the disk, κ is the testing thermal conductivity of the sample, and $D(\tau)$ is a dimensionless time dependent function, [39]

where τ is given by the following:

$$\tau = \sqrt{\frac{t}{\Theta}} \tag{4}$$

where t is the time measured from the start of the transient recording, and Θ is the characteristic time which can be defined as follows: [39]

$$\Theta = \frac{\alpha^2}{\kappa} \tag{5}$$

During the experiment process, the experimental curve of temperature increase versus elapsed time can be obtained. Subsequently, the curve of temperature increase versus $D(\tau)$ can be obtained by calculation. Then the curve of temperature increase versus $D(\tau)$ can be fitted using a linear line. The slope of the linear fitting line can be obtained. As is shown in Equation 3, the slope of the linear fitting line is $\frac{P_0}{\pi^{3/2} \cdot a \cdot \kappa}$. As known, in the experiments of this work, the total power P_0 is 0.02 W, and the overall radius

of the disk a is 3.189 mm. Therefore, the thermal conductivity κ can be calculated [39]. In addition, after the value of thermal conductivity, the value of thermal diffusivity can be calculated based on Equations 3-5.

Experimental Section

Materials. ILs of 1-butyl-3-methylimidazolium tetrafluoroborate (CAS R.N. 174501-65-6, BmimBF4), 1-butyl-3-methylimidazolium hexafluorophosphate (CAS R.N. 174501-64-5, BmimPF6), and 1-methyl-3-octylimidazolium chloride (CAS R.N. 64697-40-1, OmimCl), are obtained from Sigma-Aldrich. The mass fractions purity of the 3 ILs are \geq 97 %. Fe₂O₃ nanoparticles (NPs) (Average size: < 50 nm) are also purchased from Sigma-Aldrich and utilized in this work. R711 NPs are received from Evonik Industries AG, Germany. ILs of BmimFeCl₄ and OmimFeCl₄ are synthesized in our laboratory and utilized in this work. The synthesis procedure of BmimFeCl₄ can be found from our previous work [40]. The chemical structures of the ILs utilized in this work are shown in Scheme 1. Samples of BmimPF₆ + R711 NPs and BmimBF₄ + Fe₂O₃ NPs are also prepared in this work. The two samples are treated using ultrasound instrument at 30 °C for at least 30 minutes before the measurement. The ultrasound can enhance the NPs dispersed in the ILs. It is found the dispersed ability of R711 NPs in BmimPF₆ is excellent. However, the dispersed ability of Fe₂O₃ NPs is not as good as the former one. The Fe₂O₃ NPs can be precipitated from the bulk BmimBF₄ after a long time quiescent process.

Thermal Conductivity Measurements. Thermal conductivity of ILs is measured using hot disk method. The experimental procedure is given as follows. The hot disk instrument should be turned on at least 1 hour before the thermal conductivity measurement experiments are performed. A sample is added into a beaker. Subsequently, the sensor is put into the sample. There should be sample material at least 25 mm around the senor. The total power and measurement time for the given experiments are 0.02 W and 5 s, respectively. The experimental data of the temperature increase versus elapsed time are recorded during the process. Finally, the thermal conductivity of the sample can be calculated based on Equations 3-5.

Thermal Diffusivity Measurements. Thermal diffusivity of ILs is also measured using hot disk method. The thermal diffusivity measurement procedure is the same as the experimental procedure for thermal conductivity measurement. The thermal diffusivity can be calculated based on the experimental data and Equations 3-5. Hot disk method is a new method for ILs thermal diffusivity measurement.

Results and Discussions

Thermal Conductivity and Thermal Diffusivity Measurements of ILs using Hot Disk Method

The thermal conductivity and thermal diffusivity measurement process using the hot disk method is shown as follows. In order to demonstrate the procedure, the method on determination of the thermal conductivity and thermal diffusivity of BmimBF₆ is shown as an example. The experiments for thermal conductivity and thermal diffusivity measurement of BmimBF₆ are repeated for three times. As shown in Fig. 1a, the experimental curve of temperature increase versus elapsed time can be obtained directly during the process. Subsequently, the curve of temperature increase versus $D(\tau)$ can be calculated and obtained, which is shown in Fig. 1b. In addition, the curve of temperature increase versus $D(\tau)$ can be fitted using a linear line. The linear line fitting results are also shown in Fig. 1b. The slope of the linear fitting line can be obtained. Based on Equations 3-5, the thermal conductivity and thermal diffusivity of BmimPF₆ can be calculated. Moreover, the experimental data of difference temperature versus square root of time is obtained and shown in Fig. 1c. Furthermore, the experimental data of temperature drift versus time can be obtained during the process and is shown for BmimPF₆ in Fig. 1d. This curve can be utilized to check the temperature drift during the process, which is important as to whether the experimental data is reliable or not.

Thermal Conductivity Measurements

Reliability of Hot Disk Method. The thermal conductivity of water is measured using the hot disk method at 21 °C. The experimental results are shown in Table 1. The average measured thermal conductivity value of water is 0.6033 Wm⁻¹K⁻¹ at 21 °C in this work. The reference thermal conductivity value of water at 20 °C is 0.6 Wm⁻¹K⁻¹ [34]. In addition, the measured thermal conductivity values of pure ILs BmimBF₄ and BmimPF₆ are 0.1898 Wm⁻¹K⁻¹ and 0.1733 Wm⁻¹K⁻¹ at 21 °C, respectively. The reference values of thermal conductivity of BmimBF₄ and BmimPF₆ are 0.186 Wm⁻¹K⁻¹ and 0.145 Wm⁻¹K⁻¹ at 25 °C, respectively [41]. The measurement results are in good agreements with the reference values. Therefore, the hot disk method are utilized to measure the thermal conductivity of ILs based materials in this work.

Pure ILs Thermal Conductivity Measurements. Thermal conductivity of pure ILs, including BmimBF₄, BmimPF₆, OmimCl, BmimFeCl₄, and OmimFeCl₄, is measured at 21 °C. The curve of temperature increase versus elapsed time of BmimPF₆ is shown in Fig. 1a. The curves of temperature increase versus time of the other 4 ILs are shown in Fig. 2. Based on the temperature increase versus time curves, the temperature increase versus D(τ) curves of the ILs can be obtained, which are shown in Fig. 1b and Fig. 3. Subsequently, based on Equations 3-5, the thermal conductivity of ILs can be calculated. The results are shown in Table 2. It is found that the measurement thermal conductivity values of the 5 pure ILs are no more than 0.1898 Wm⁻¹K⁻¹. However, the measurement thermal conductivity value of

water is 0.6033 Wm⁻¹K⁻¹. The thermal conductivity of the 5 pure ILs is thus much lower than that of water.

Effect of NPs on Thermal Conductivity of ILs. The effect of NPs on thermal conductivity of ILs is investigated. The two samples of BmimBF₄ + Fe₂O₃ NPs and BmimPF₆ + R711 NPs are utilized. The curves of temperature increase versus elapsed time are obtained and shown in Fig. 4. The curves of temperature increase versus D(τ) are obtained and shown in Fig. 5. Subsequently, the thermal conductivity values of the samples can be calculated. The results are shown in Table 3. As shown in Table 2, the thermal conductivity of BmimBF₄ is 0.1898 Wm⁻¹K⁻¹ at 21 °C. As shown in Table 3, the thermal conductivity of the BmimBF₄ + Fe₂O₃ NPs sample becomes 0.1895 Wm⁻¹K⁻¹ at 21 °C. Hence, it is shown that the thermal conductivity of BmimBF₄ does not change significantly in the presence of Fe₂O₃ NPs.

However, as shown in Table 2, the thermal conductivity of BmimPF₆ is 0.1733 Wm⁻¹K⁻¹ at 21 °C. As shown in Table 3, the thermal conductivity of the BmimPF₆ + R711 NPs sample becomes 0.1670 Wm⁻¹K⁻¹ at 21 °C. Hence, it illustrates that the thermal conductivity of BmimPF₆ decreases about 3.6 % in the presence of R711 NPs.

Thermal Diffusivity Measurements

Pure ILs Thermal Diffusivity Measurements. Thermal diffusivity of pure ILs, including BmimBF₄, BmimPF₆, BmimFeCl₄, OmimCl and OmimFeCl₄, is measured. The results are shown in Table 4. The thermal diffusivity values of the ILs are no more than 0.1185 mm²s⁻¹. This value is less than the thermal diffusivity of water 0.143 mm²s⁻¹ [42]. It illustrates that the ILs also have a better property than water for energy storage. Therefore, ILs may be considered as a type of novel materials for energy storage.

Effect of NPs on Thermal Diffusivity of ILs. The effect of NPs on thermal diffusivity of ILs is investigated. The results are shown in Table 5. Similarly to the effect of NPs on thermal conductivity, the samples of BmimBF₄ + Fe₂O₃ NPs and BmimPF₆ + R711 NPs are utilized. As shown in Table 4, the thermal diffusivity of BmimBF₄ is $0.1140 \text{ mm}^2\text{s}^{-1}$ at 21 °C. As shown in Table 5, the thermal diffusivity of the BmimBF₄ + Fe₂O₃ NPs sample becomes $0.1116 \text{ mm}^2\text{s}^{-1}$ at 21 °C. Hence, it is shown that the thermal diffusivity of BmimBF₄ does not change significantly in the presence of Fe₂O₃ NPs.

For BmimPF₆, as shown in Table 4, the thermal diffusivity is $0.1082 \text{ mm}^2\text{s}^{-1}$ at 21 °C. As shown in Table 5, the thermal conductivity of the BmimPF₆ + R711 NPs sample becomes $0.09673 \text{ mm}^2\text{s}^{-1}$ at 21 °C. Hence, the thermal diffusivity of BmimPF₆ decreases about 10.6 % in the presence of R711 NPs.

Conclusions

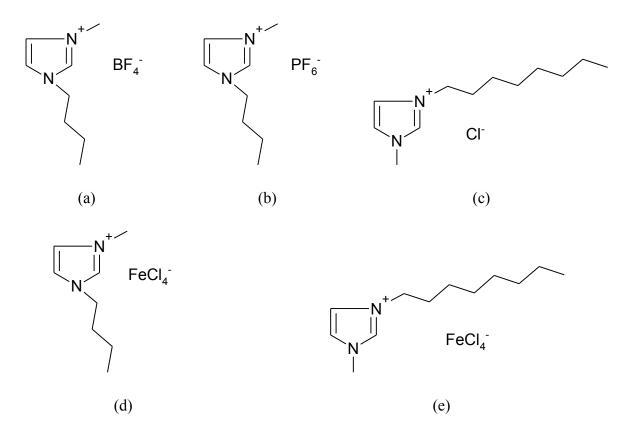
ILs represent a hot research topic and have thus been utilized in many scientific research fields. For example, ILs may be utilized in energy devices, including solar cells, batteries, and supercapacitors. Thermal conductivity and thermal diffusivity of ILs are crucial for real energy conversion and energy storage processes. However, the thermal conductivity and thermal diffusivity of ILs are often unknown. Therefore, in this work the thermal conductivity and thermal diffusivity of ILs are investigated. A thermal conductivity and thermal diffusivity measurement method for ILs is developed.

Thermal conductivity of pure ILs BmimBF₄, BmimPF₆, OmimCl, BmimFeCl₄ and OmimFeCl₄, is measured. It is found that the thermal conductivity of the five pure ILs is much lower than the thermal conductivity of water. The effect of NPs on thermal conductivity of ILs is also investigated. It is shown that the thermal conductivity of BmimBF₄ does not change significantly in the presence of Fe₂O₃ NPs. However, the thermal conductivity of BmimPF₆ decreases somewhat in the presence of R711 NPs.

Furthermore, thermal diffusivity of ILs is measured in this work. Thermal diffusivity of 5 pure ILs, including BmimBF₄, BmimPF₆, BmimFeCl₄, OmimCl and OmimFeCl₄, is measured. The thermal diffusivity of the ILs is no more than 0.1185 mm²s⁻¹ at 21 °C. The thermal diffusivity of water is 0.143 mm²s⁻¹ at 25 °C, which means ILs have a better property than water for energy storage. Therefore, ILs may be considered as a type of novel materials for energy storage. The effect of NPs on the thermal diffusivity of ILs is also investigated. It is shown that the thermal diffusivity of BmimBF₄ does not change significantly in the presence of Fe₂O₃ NPs. However, the thermal diffusivity of BmimPF₆ decreases about 10.6 % in the presence of R711 NPs.

Acknowledgements

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Scheme 1. Structure of BmimBF₄ (a), BmimPF₆ (b), OmimCl (c), BmimFeCl₄ (d), and OmimFeCl₄ (e).

Table 1. Thermal conductivity of water at 21 $^{\circ}$ C.

Water	Experiment number	Test value/Wm ⁻¹ K ⁻¹	Average value/Wm ⁻¹ K ⁻¹
	1	0.6080	
	2	0.6022	
	3	0.6147	
	4	0.6010	
	5	0.5906	
			0.6033

Table 2. Thermal conductivity of BmimBF₄, BmimPF₆, OmimCl, BmimFeCl₄ and OmimFeCl₄ at 21 °C.

Sample	Experiment number	Test value/Wm ⁻¹ K ⁻¹	Average value/Wm ⁻¹ K ⁻¹
BmimBF ₄	1	0.1900	
	2	0.1891	
	3	0.1890	
	4	0.1909	
			0.1898
BmimPF ₆	1	0.1738	
	2	0.1709	
	3	0.1753	
			0.1733
OmimCl	1	0.1881	
	2	0.1862	
	3	0.1918	
	4	0.1829	
			0.1873
BmimFeCl ₄	1	0.1671	
	2	0.1692	
	3	0.1703	
	4	0.1655	
			0.1680
OmimFeCl ₄	1	0.1692	
OmimFeCl ₄	1	0.1692	

2	0.1692	
3	0.1642	
4	0.1596	
		0.1656

Table 3. Thermal conductivity of samples of BmimBF $_4$ + Fe $_2$ O $_3$ NPs and BmimPF $_6$ + R711 NPs at 21 $^{\circ}$ C.

Sample	Experiment number	Test value/Wm ⁻¹ K ⁻¹	Average value/Wm ⁻¹ K ⁻¹
BmimBF ₄ + Fe ₂ O ₃ NPs	1	0.1892	
	2	0.1890	
	3	0.1864	
	4	0.1934	
			0.1895
BmimPF ₆ + R711 NPs	1	0.1685	
	2	0.1670	
	3	0.1645	
	4	0.1678	
			0.1670

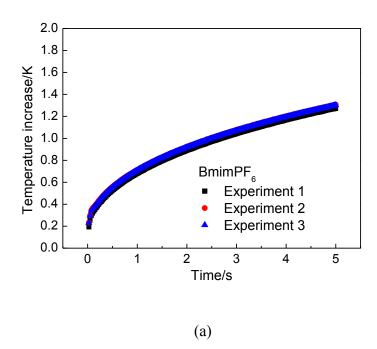
Table 4. Thermal diffusivity of BmimBF₄, BmimPF₆, OmimCl, BmimFeCl₄ and OmimFeCl₄ at 21 °C.

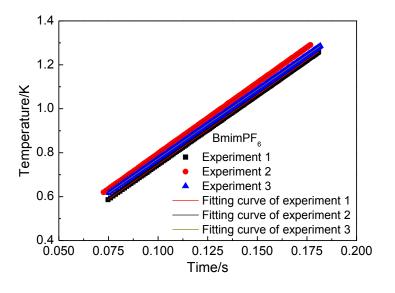
Sample	Experiment number	Test value/mm ² s ⁻¹	Average value/mm ² s ⁻¹
BmimBF ₄	1	0.1142	
	2	0.1141	
	3	0.1126	
	4	0.1151	
			0.1140
BmimPF ₆	1	0.1096	
	2	0.1041	
	3	0.1109	
			0.1082
OmimCl	1	0.1105	
	2	0.1055	
	3	0.1149	
	4	0.1000	
			0.1077
BmimFeCl ₄	1	0.1174	
	2	0.1208	
	3	0.1229	
	4	0.1129	
			0.1185
OmimFeCl ₄	1	0.1140	

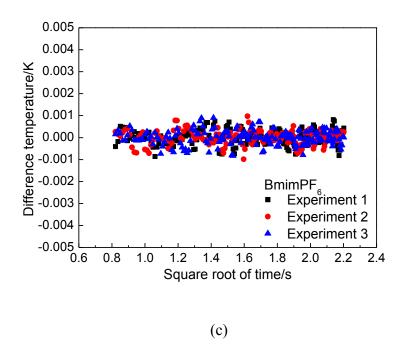
2	0.1177	
3	0.1085	
4	0.1017	
		0.1105

Table 5. Thermal diffusivity of samples of BmimBF $_4$ + Fe $_2$ O $_3$ NPs and BmimPF $_6$ + R711 NPs at 21 °C.

Sample	Experiment number	Test value/mm ² s ⁻¹	Average value/mm ² s ⁻¹
BmimBF ₄ + Fe ₂ O ₃ NPs	1	0.1111	
	2	0.1106	
	3	0.1062	
	4	0.1183	
			0.1116
BmimPF ₆ + R711 NPs	1	0.09929	
	2	0.09635	
	3	0.09298	
	4	0.09829	
			0.09673







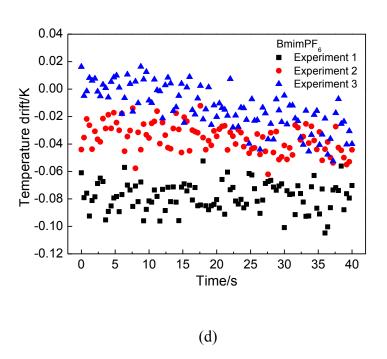
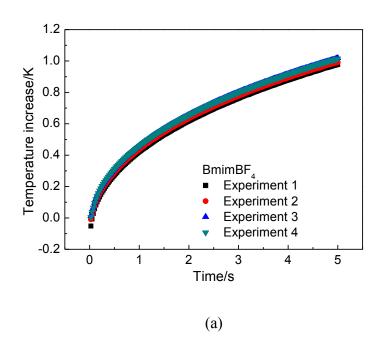


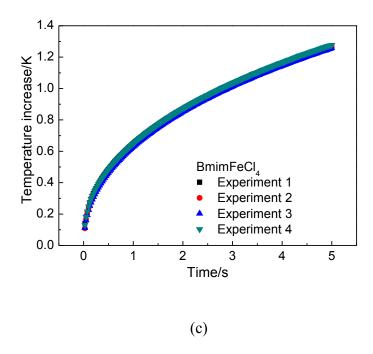
Fig. 1. Thermal conductivity and thermal diffusivity measurement of BmimPF₆. (a) Temperature increase versus $D(\tau)$. (The slopes of the fitting curves of experiments 1, 2 and 3 are 6.3337, 6.4415, 6.2822, respectively. The R² of all the fitting curves of experiments 1, 2 and 3 are 1). (c) Difference temperature versus square root of time; (d) Temperature drift versus time.



1.4 1.2 Temperature increase/K 1.0 8.0 0.6 OmimCl
Experiment 1
Experiment 2 0.4 0.2 Experiment 3
Experiment 4 0.0 0 2 5 1 3

(b)

Time/s



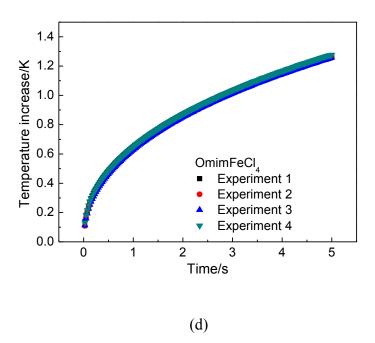
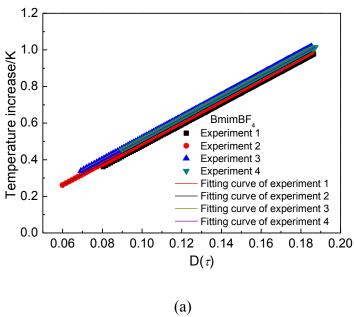
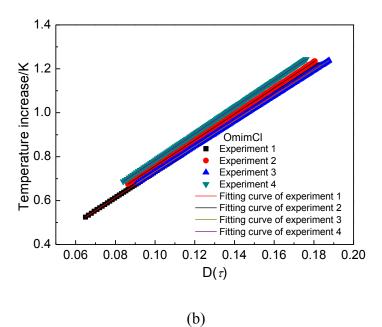
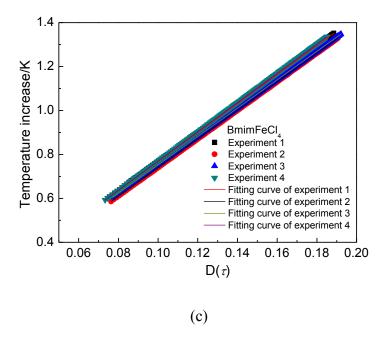


Fig. 2. Temperature increase versus time during the measurement process in BmimBF₄, OmimCl, BmimFeCl₄ and OmimFeCl₄.







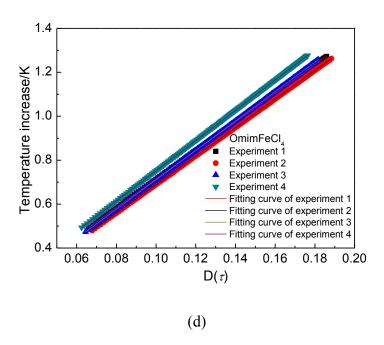
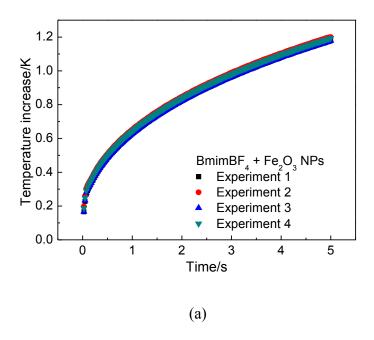


Fig. 3. Temperature increase versus $D(\tau)$ curves and the linear fitting lines of temperature increase versus $D(\tau)$ in BmimBF₄ (a, The slopes of the fitting curves of experiments 1, 2, 3 and 4 are 5.8127, 5.8035, 5.8376, 5.7915, respectively. The R^2 of all the fitting curves of experiments 1, 2, 3 and 4 are 1). OmimCl (b, The slopes of the fitting curves of experiments 1, 2, 3 and 4 are 5.8625, 5.9399, 5.7662, 6.0480, respectively. The R^2 of all the fitting curves of experiments 1, 2, 3 and 4 are 1), BmimFeCl₄ (c, The slopes of the fitting curves of experiments 1, 2, 3 and 4 are 6.5846, 6.5063, 6.4654, 6.6489, respectively. The R^2 of all the fitting curves of experiments 1, 2, 3, and 4 are 1) and OmimFeCl₄ (d, The slopes of the fitting curves of experiments 1, 2, 3 and 4 are 6.4980, 6.4942, 6.6907, 6.8855, respectively. The R^2 of all the fitting curves of experiments 1, 2, 3, and 4 are 1).



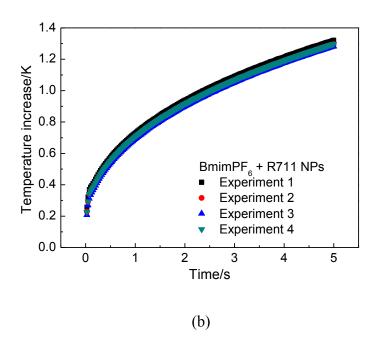
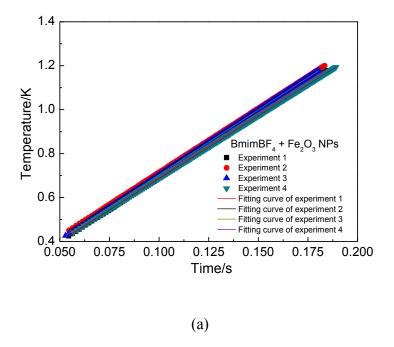


Fig. 4. Temperature increase versus time during the measurement process in $BmimBF_4 + Fe_2O_3$ NPs and $BmimPF_6 + R711$ NPs samples.



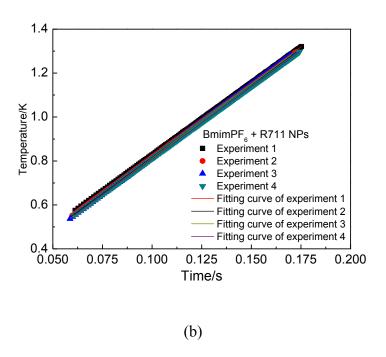


Fig. 5. Temperature increase versus $D(\tau)$ curves and the linear fitting lines of temperature increase versus $D(\tau)$ in the sample of BmimBF₄ + Fe₂O₃ NPs (a, The slopes of the fitting curves of experiments 1, 2, 3 and 4 are 5.8176, 5.8230, 5.9036, 5.6990, respectively. The R² of all the fitting curves of experiments 1, 2, 3, and 4 are 1) and BmimPF₆ + R711 NPs (b, The slopes of the fitting curves of experiments 1, 2, 3 and 4 are 6.5338, 6.5939, 6.6943, 6.5609, respectively. The R² of all the fitting curves of experiments 1, 2, 3, and 4 are 1).

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