

Ionic Conductivities of Molten Alkali Metal Chloride Binary Systems by Equilibrium Molecular Dynamics Simulation: Composition and Temperature Dependence

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Conductivity of molten alkali metal chlorides is the foundation of molten-salt electrolysis process. In the study, equilibrium molecular dynamics was performed to investigate the ionic conductivities of the molten alkali metal chloride binary systems. The calculated values were in good agreement with the experimental results. Simple databases for conductivity have been built based on the verified algorithm. Mathematic expressions of conductivity vs. temperature & composition have also been obtained according to calculated data. Ionic conductivities of these melts exhibited positive dependences on temperature. Adding a smaller alkali metal cation into molten alkali chlorides could improve the conductive performance. With the gradual addition of LiCl, ionic conductivities of molten LiCl-NaCl, LiCl-KCl, LiCl-RbCl, and LiCl-CsCl all increased monotonously. The conductivities of molten NaCl-KCl, NaCl-RbCl, and NaCl-CsCl also increased with the continuous addition of NaCl. The improvement effects of LiCl on ionic conductivities of molten KCl, RbCl, and CsCl are more significant than those of NaCl.

Keywords: ionic conductivity, equilibrium molecular dynamics simulation, molten alkali metal chlorides.

1. INTRODUCTION

Molten alkali halides have been widely used in many industrial applications due to the excellent properties under high temperature [1]. Accurately and integrally revealing the micro-structures and transport properties of molten alkali halides had long been the subjects of many scientists. Restricted by the high temperature, corrosive and volatility characteristics of molten alkali halides, it is difficult to systematically determine the local structures and dynamics properties through experiments. It is also not surprising that there are substantial deficiencies and even disparities among the reported experimental results for molten salts. Molecular dynamics simulation which had successfully calculated the properties of many melts [2–4] has been an effective means to investigate inorganic molten salts at different temperatures and varying compositions.

Since Tosi and Fumi [5, 6] developed the potential parameters for molten alkali halides, the system has been researched intensively through molecular dynamics, and the ionic conductivity of molten alkali chlorides also has been studied dispersedly in succession. Hansen [7] calculated the ionic conductivities of molten NaCl at 1267 K by molecular dynamics with the time autocorrelation function, and the results were in good agreements with the experimental data. Ciccotti [8] simulated the velocity autocorrelation functions and calculated the ionic conductivities of molten NaCl and RbCl. Trullas [9] calculated the diffusive transport properties and ionic conductivities of molten NaCl and KCl by molecular dynamics method. Koishi [10] obtained the

ionic conductivities of molten NaCl by non-equilibrium molecular dynamics method. Galamba [11, 12] calculated the ionic conductivities of molten NaCl and KCl through equilibrium and non-equilibrium molecular dynamics simulations. Wang [13] studied the temperature dependence of the conductive property for pure molten alkali chlorides through equilibrium molecular dynamics simulation method. In addition to pure molten salt, the conductive performance of molten alkali halide mixtures have also been studied through MD simulations. Merlet [14] and Morgan [15] calculated the ionic conductivities of molten LiF-KF and LiCl-KCl systems through molecular dynamics calculations. Wang [16, 17] have also investigated the composition dependences of ionic conductivity for molten alkali chloride binary mixtures at 1100 K. Sun [18] investigated the ionic conductivities of (Li, Na, K, Cs)Cl and (Li, K, Rb, Cs)Cl systems through molecular dynamics calculations.

Ionic conductivity is a very important index of electrolyte that increasing the ionic conductivity of electrolyte can effectively improve the current efficiency and reduce the energy consumption in electrolysis process [19]. It is important to improve the conductive performance of electrolyte through composition or temperature regulation. Alkali chlorides have been the main additives of molten electrolyte for metal production. However, the existing data for the ionic conductivities of molten alkali chlorides is discrete and far from consolidated. Due to the harsh experimental conditions and tedious computational

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process, there is no systematical research for the ionic conductivities of molten alkali chlorides.

It is significant and urgent to accurately predict the ionic conductivities for molten alkali chlorides over a wide range of conditions. Based on previous work, ionic conductivities of some more widely used molten alkali chloride binary systems at different temperatures and varying compositions have been calculated through equilibrium molecular dynamics method to satisfy the requirements of practical engineering and to consummate the basic research.

2. METHODOLOGY

2.1. Simulation details

Equilibrium molecular dynamics (EMD) simulations were performed to calculate the ionic conductivities of molten alkali metal chloride binary mixtures at different temperatures and components, i.e. LiCl-NaCl, LiCl-KCl, LiCl-RbCl, LiCl-CsCl, NaCl-KCl, NaCl-RbCl, and NaCl-CsCl.

Following Born-Mayer-Huggins potential which had been successfully used in the simulations of molten alkali halides [20], was applied to describe the interactions between ions:

$$U_{ij}(r) = \frac{q_i q_j}{r} + B_{ij} \exp\left(\frac{\sigma_{ij} - r}{\rho}\right) - \frac{C_{ij}}{r^6} - \frac{D_{ij}}{r^8}. \quad (1)$$

In Eq. 1, the first term describes the electrostatic interactions between ions, q is the ionic charge, and formal charge (+1 for alkali ions and -1 for chloride) was used. The second term represents Born-Huggins exponential short-range repulsion due to the overlap of electron clouds. $B_{ij} = A_{ij} \cdot b$, in which b is a constant, being equal to 0.338×10^{-19} J, A_{ij} is Pauling factor [21, 22], being equal to $1 + \frac{Z_i}{n_i} + \frac{Z_j}{n_j}$, where Z denotes charge of ion and n denotes the number of outer electrons, σ is the crystal ionic "radii", and ρ is the hardness parameter. The last two terms describe the dipole-dipole and dipole-quadrupole dispersion interactions, and the dispersion constants C_{ij} and D_{ij} for alkali chloride systems had been determined by Mayer [23]. Detailed description of potential parameter processing for molten alkali metal chloride binary mixtures and the corresponding parameters had been given in previous work [24]. The LAMMPS code [25] was used to perform EMD simulations. All the simulations were performed with a cubic box consisting of 1024 ions (512 cations and 512 anions respectively, and the numbers of each cation were determined according to its mole fractions). Initial positions of ions were in random distribution. Initial velocities of ions were in random allocation and obeyed Gaussian distribution. Periodic Boundary Condition (PBC) was used to keep particle numbers constant and to eliminate boundary effect. Long-range interactions were handled with Ewald [26] summation method to eliminate truncation errors. For Ewald summation, the precise which determined the relative root mean square (RMS) error in per-atom forces calculated by the method was set to be 1.0×10^{-6} , meaning that the RMS error would be a factor of 1000000 smaller than the reference force that two unit point charges exert on each other at a distance of 1 Å. The cutoff radius r_c for the short-range potential and the real space part of the Ewald

summation was set to be $L/2$, where L is the side length of the simulation cell. Verlet algorithm [27] was used to solve Newton's equations of motion.

For all the simulations, the systems were first equilibrated in isothermal-isobaric (NPT) ensemble with pressure fixed at 0 GPa around the desired temperature using the Nose-Hoover thermostat and barostat [28, 29]. Subsequently, production runs were performed in canonical (NVT) ensemble with Nose-Hoover thermostat method [30, 31] at the equilibrated cell volume. Damping parameters that determined relaxation rates of temperature and pressure were set to be 0.1 ps. To control initial oscillations, Nose-Hoover chains were used for the thermostat and barostat, and the chain numbers were set to be 3 in this work [32]. For each calculation, the relaxation time was 500 ps and the production time was 10 ns, with a time step of 1 fs.

2.2. Evaluated properties

Based on the Green-Kubo formula [33], the ionic conductivity λ can be calculated from the time integral of the charge flux auto-correlation function through EMD method:

$$\lambda = \frac{1}{3VkT} \int_0^\infty \langle J_Z(0) \cdot J_Z(t) \rangle dt, \quad (2)$$

where k is the Boltzmann constant, T is temperature, and V is the volume of the simulation cell. The charge flux vector $J_Z(t)$ is defined as:

$$J_Z(t) = \sum_{i=1}^n z_i e v_i(t), \quad (3)$$

where $z_i e$ and v_i are the charge and velocity of ion i , respectively. Each of the three independent components (i.e. J_{xZ} , J_{yZ} , J_{zZ}) of the charge flux vector provides an independent estimate of ionic conductivity, and the averaged value will be taken as the final ionic conductivity value.

3. ALGORITHM VALIDATION

Ionic conductivities of molten LiCl-NaCl, LiCl-KCl, LiCl-RbCl, LiCl-CsCl, NaCl-KCl, NaCl-RbCl, and NaCl-CsCl with different components and temperatures have been simulated from the time integral of charge flux autocorrelation function through equilibrium molecular dynamics simulation.

3.1. Decay and integral behaviors of charge flux autocorrelation function

Before ionic conductivity calculations, decay behavior and integral stability of the charge flux auto-correlation function should be tested first, and the time to calculate charge flux auto-correlation function needs to be determined, which must be long enough to capture the whole decay process and ensure the integral reach a stationary value. The components calculated in this study distributed uniformly from 0 to 100 in the measure of mole percentage, and the pure salts at both ends are selected to complete the test. Here, the charge flux auto-correlation functions and corresponding running integrals of molten LiCl and CsCl have been picked out and analyzed due to their respective cation size characteristics.

Fig. 1 displayed the normalized charge flux auto-correlation functions and corresponding running integrals of molten LiCl and CsCl. Both charge flux auto-correlation functions decayed to zero quickly and the corresponding integrals reached the plateau after about 0.8 ps. To ensure the data accuracy, 1.0 ps has been adopted to calculate charge flux auto-correlation functions in this paper. For all the ionic conductivity calculation, the charge flux auto-correlation functions were averaged for 10 million steps to ensure good statistics that repeated runs gave the same plateau value.

3.2. Model verification

To verify the accuracy of the simulation model, the calculated ionic conductivities should be tested through experimental results. Being enslaved to the demanding and costly experimental conditions, systematic and extensive ionic conductivity measurement for high-temperature molten alkali chlorides at various temperatures and compositions is unrealistic under the current circumstances, so the experimental data for ionic conductivities of molten chlorides are scarce and dispersed now. The comparisons between some simulated results with available experimental values [34] have been presented in Fig. 2, with the experimental (calculated) results at 1100 K with the unit of S/cm were 1.75 (1.74) for molten RbCl, 2.35 (2.19) for molten 50 % LiCl-RbCl, 1.89 (1.88) for molten

25 % NaCl-RbCl, 2.15 (2.12) for molten 50 % NaCl-RbCl, 2.62 (2.61) for molten 75 % NaCl-RbCl, an so on.

The calculated results agreed well with the experimental data, which confirmed the accuracy of the simulated method. With this prerequisite, massive calculations for the ionic conductivities of molten LiCl-NaCl, LiCl-KCl, LiCl-RbCl, LiCl-CsCl, NaCl-KCl, NaCl-RbCl, and NaCl-CsCl have been launched.

4. RESULTS AND DISCUSSION

For all the molten alkali metal chloride binary systems, the calculated compositions were evenly distributed from 0 to 100 % with an interval of 6.25 %, in the measure of mole fraction of one component. Considering the computation cost and the industrial application that molten alkali chlorides are mainly used below 800 °C, the upper limit of computed temperature was set to be 1200 K. Then the calculated temperatures were set to be within near melting point to 1200 K with an interval of 25 K, with the lower limits determined through the equilibrium phase diagram of the corresponding system.

The calculated ionic conductivities of molten LiCl-NaCl, LiCl-KCl, LiCl-RbCl, LiCl-CsCl, NaCl-KCl, NaCl-RbCl, and NaCl-CsCl in the temperature range of near melting point – 1200 K across the whole concentration scope have been recorded in Table 1 – Table 7.

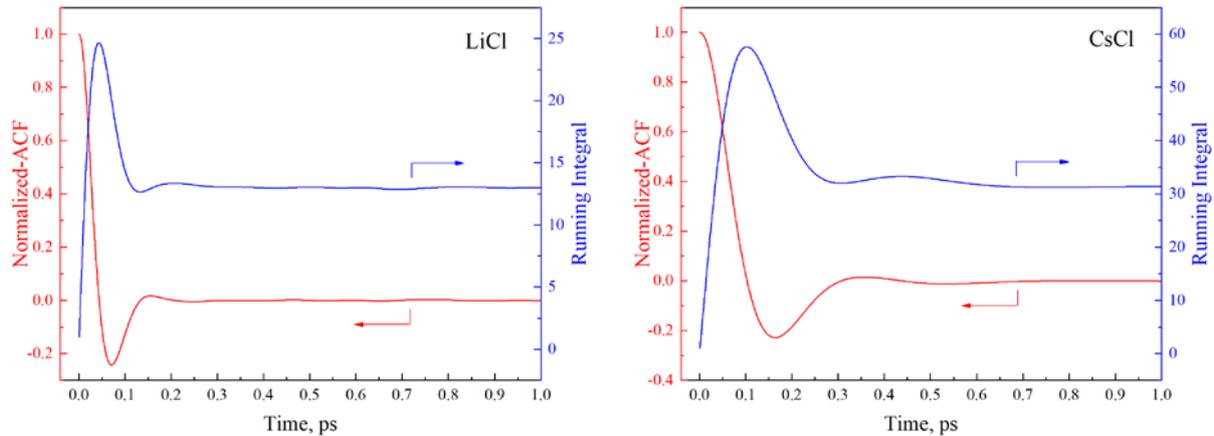


Fig. 1. Time evolutions of normalized charge flux auto-correlation functions and corresponding running integrals for molten LiCl and CsCl

Table 1. Ionic conductivities of molten LiCl-NaCl at different temperatures (units: S/cm)

LiCl, % Temp, K	0.00	6.25	12.50	18.75	25.00	31.25	37.50	43.75	50.00	56.25	62.50	68.75	75.00	81.25	87.50	93.75	100
850	-	-	-	-	-	-	-	-	-	-	-	4.20	4.15	-	-	-	-
875	-	-	-	-	-	-	-	-	-	-	-	4.24	4.43	4.29	4.62	-	-
900	-	-	-	-	-	-	-	-	-	3.94	4.08	4.33	4.40	4.77	4.72	4.92	5.30
925	-	-	-	-	-	-	-	-	3.76	4.08	4.12	4.64	4.79	4.64	5.09	5.43	5.91
950	-	-	-	-	-	-	-	3.84	3.90	4.11	4.30	4.14	4.58	4.71	5.15	5.37	5.34
975	-	-	-	-	-	-	3.77	3.92	4.16	4.28	4.24	4.40	4.67	4.92	5.23	5.49	5.75
1000	-	-	-	-	-	3.76	3.94	3.92	4.22	4.46	4.68	4.73	4.69	5.27	5.24	5.27	5.97
1025	-	-	-	-	3.72	3.83	3.94	4.17	4.21	4.33	4.56	4.62	5.11	5.06	5.45	5.61	6.05
1050	-	-	-	3.64	3.79	3.91	4.02	4.18	4.37	4.19	4.55	4.77	5.20	5.17	5.50	5.70	5.89
1075	-	3.51	3.66	3.80	3.86	3.95	4.15	4.09	4.29	4.41	4.57	4.81	4.99	4.99	5.51	5.82	6.29
1100	3.55	3.57	3.56	3.84	3.71	4.04	3.91	4.15	4.53	4.41	4.70	4.74	5.14	5.19	5.58	5.65	6.41
1125	3.48	3.66	3.65	3.77	3.88	4.01	4.12	4.34	4.51	4.50	4.51	5.13	5.33	5.17	5.15	5.99	6.31
1150	3.63	3.53	3.88	3.72	3.95	4.14	4.01	4.35	4.30	4.68	4.91	4.91	5.26	5.38	5.47	6.06	6.27
1175	3.56	3.75	3.75	3.93	3.93	4.05	4.16	4.48	4.60	4.73	4.93	5.08	4.96	5.41	5.93	5.89	5.95
1200	3.63	3.72	3.72	3.74	4.05	4.24	4.12	4.55	4.41	4.69	4.88	5.34	5.31	5.42	6.03	6.13	6.36

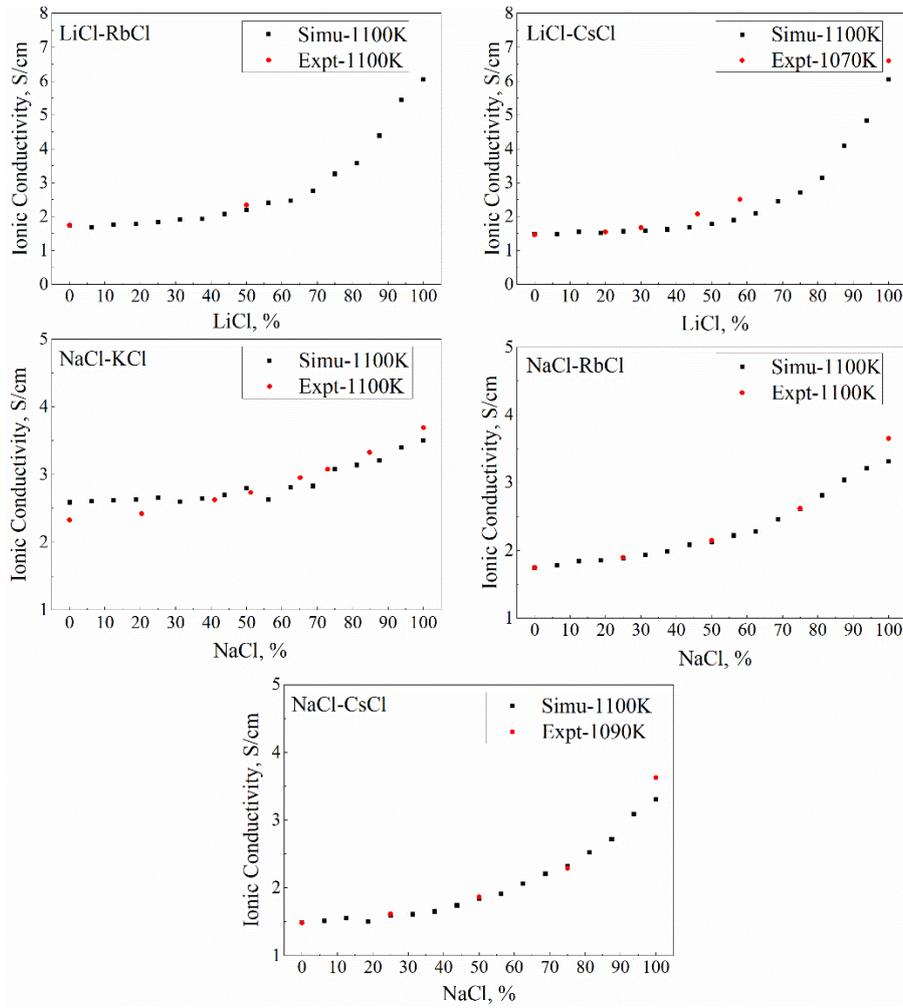


Fig. 2. Comparisons between computed values with available experimental results for ionic conductivities

In addition, the expressions of ionic conductivity on temperature and component have been fitted according to the calculated results through the software of 1stOpt with Levenberg-Marquardt (LM)-Universal Global Optimization (UGO) algorithms and convergence criteria of 1.0×10^{-10} .

4.1. Molten LiCl-NaCl

Ionic conductivities of molten LiCl-NaCl have been calculated extensively and listed in Table 1. For molten LiCl-NaCl, the ionic conductivity presented positive dependences on both temperature and LiCl content.

As revealed previously, with the gradual addition of LiCl, the shear viscosities of molten LiCl-NaCl decreased first and then presented slight oscillation tendency under the effects of both local structure and interatomic force. Being different from shear viscosity, the ionic conductivities of molten LiCl-NaCl increased monotonously with LiCl content at a certain temperature. As analyzed from the radial distribution functions, the addition of Li^+ into molten NaCl weakened interactions between counter-ions, making the ions more diffusive. The overall charge transport of the system was influenced greatly by the cation-anion association, so the conductive performance could be enhanced by LiCl. With the gradual addition of LiCl, the interactions between counter-ions got weakened continuously, and the migration ability of ions was enhanced little by little, resulting in the gradual increase of

ionic conductivity.

The expression of ionic conductivity for molten LiCl-NaCl system at the temperature range of 850–1200 K has been fitted through the data in Table 1, as follows:

$$\lambda_{(\text{LiCl-NaCl})} = -5.5810 + 1.6958 \times 10^{-2}x + 1.4294 \times 10^{-2}T - 2.7581 \times 10^{-5}x^2 - 5.5304 \times 10^{-6}T^2 + 1.2461 \times 10^{-6}x^3. \quad (4)$$

In Eq. 4 x denotes the mole fraction of LiCl, T denotes temperature.

4.2. Molten LiCl-KCl

Ionic conductivities of molten LiCl-KCl at different temperatures and components have been recorded in Table 2. The ionic conductivity of molten LiCl-KCl system increased with temperature as well as LiCl content. Along with the addition of LiCl, the interactions of Li-Cl and K-Cl pairs got weakened, and the charge transport of the system gained enhanced, resulting in the increase of the ionic conductivity.

Based on Table 2, the expression of ionic conductivity for molten LiCl-KCl system within full composition range and temperature range of 700–1200 K has been fitted as follows:

$$\lambda_{(\text{LiCl-KCl})} = -4.7857 + 1.9633 \times 10^{-2}x + 1.1345 \times 10^{-2}T - 5.5203 \times 10^{-4}x^2 - 4.3429 \times 10^{-6}T^2 + 7.1226 \times 10^{-6}x^3. \quad (5)$$

Table 2. Ionic conductivities of molten LiCl-KCl at different temperatures (units: S/cm)

LiCl, % Temp, K	0.00	6.25	12.50	18.75	25.00	31.25	37.50	43.75	50.00	56.25	62.50	68.75	75.00	81.25	87.50	93.75	100.00
700	-	-	-	-	-	-	-	-	-	1.67	1.81	-	-	-	-	-	-
725	-	-	-	-	-	-	-	-	-	1.86	2.02	2.15	-	-	-	-	-
750	-	-	-	-	-	-	-	-	-	2.01	2.07	2.30	-	-	-	-	-
775	-	-	-	-	-	-	-	-	2.03	2.09	2.25	2.45	2.64	-	-	-	-
800	-	-	-	-	-	-	-	-	2.09	2.17	2.37	2.54	2.78	-	-	-	-
825	-	-	-	-	-	-	-	2.07	2.16	2.31	2.41	2.59	2.82	3.23	-	-	-
850	-	-	-	-	-	-	-	2.18	2.30	2.41	2.52	2.78	3.00	3.36	4.02	-	-
875	-	-	-	-	-	-	2.24	2.23	2.43	2.54	2.71	2.83	3.14	3.50	3.94	-	-
900	-	-	-	-	-	-	2.26	2.35	2.49	2.60	2.74	2.93	3.30	3.66	4.21	4.55	5.30
925	-	-	-	-	-	2.30	2.29	2.41	2.59	2.63	2.88	3.08	3.24	3.69	4.22	4.73	5.91
950	-	-	-	-	-	2.45	2.41	2.52	2.57	2.74	2.95	3.17	3.51	3.84	4.08	4.56	5.34
975	-	-	-	-	2.41	2.38	2.53	2.53	2.73	2.83	2.94	3.20	3.63	4.12	4.35	4.88	5.75
1000	-	-	-	2.45	2.47	2.53	2.50	2.64	2.64	2.82	3.00	3.28	3.51	3.77	4.47	4.84	5.97
1025	-	-	2.44	2.48	2.43	2.52	2.60	2.64	2.88	3.01	3.01	3.32	3.55	4.17	4.47	4.92	6.05
1050	-	2.44	2.44	2.59	2.50	2.63	2.65	2.68	2.88	3.00	3.17	3.48	3.80	4.15	4.75	4.87	5.89
1075	2.54	2.46	2.54	2.55	2.56	2.63	2.73	2.86	2.87	3.03	3.10	3.55	3.75	3.99	4.88	5.58	6.29
1100	2.54	2.49	2.61	2.60	2.68	2.70	2.73	2.87	3.01	3.06	3.29	3.59	3.83	4.31	4.55	5.28	6.41
1125	2.59	2.51	2.56	2.68	2.74	2.68	2.81	2.80	3.04	3.25	3.37	3.63	3.63	4.18	4.69	5.36	6.31
1150	2.58	2.68	2.61	2.62	2.79	2.69	2.91	2.83	3.05	3.13	3.41	3.59	4.03	4.32	4.88	5.49	6.27
1175	2.66	2.62	2.62	2.67	2.62	2.78	2.79	2.91	3.13	3.12	3.38	3.92	3.80	4.25	4.70	5.20	5.95
1200	2.73	2.68	2.77	2.79	2.72	2.84	2.81	3.12	3.08	3.04	3.33	3.67	3.81	4.29	4.73	5.50	6.36

4.3. Molten LiCl-RbCl

Table 3 listed the ionic conductivities of molten LiCl-RbCl at different temperatures and compositions in detail. With the increases of temperature and LiCl content, the ionic conductivity of molten LiCl-RbCl system increased monotonously, due to the interaction weakening phenomenon of counter-ion pairs. The following mathematical expression of temperature and composition dependences of ionic conductivity for molten LiCl-KCl system at the temperature range of 625 – 1200 K has been fitted based on Table 3:

$$\lambda_{(\text{LiCl-RbCl})} = -3.8023 + 3.0644 \times 10^{-2}x + 7.8421 \times 10^{-3}T - 8.9982 \times 10^{-4}x^2 - 2.6797 \times 10^{-6}T^2 + 1.0305 \times 10^{-5}x^3. \quad (6)$$

Table 3. Ionic conductivities of molten LiCl-RbCl at different temperatures (units: S/cm)

LiCl, % Temp, K	0.00	6.25	12.50	18.75	25.00	31.25	37.50	43.75	50.00	56.25	62.50	68.75	75.00	81.25	87.50	93.75	100.00
625	-	-	-	-	-	-	-	-	-	0.76	-	-	-	-	-	-	-
650	-	-	-	-	-	-	-	-	0.79	0.89	1.03	-	-	-	-	-	-
675	-	-	-	-	-	-	-	-	0.92	1.03	1.15	-	-	-	-	-	-
700	-	-	-	-	-	-	-	-	1.02	1.10	1.30	-	-	-	-	-	-
725	-	-	-	-	-	-	-	-	1.10	1.24	1.37	1.55	-	-	-	-	-
750	-	-	-	-	-	-	-	1.14	1.19	1.31	1.48	1.68	-	-	-	-	-
775	-	-	-	-	-	-	-	1.23	1.30	1.46	1.61	1.81	2.08	-	-	-	-
800	-	-	-	-	-	-	1.24	1.32	1.41	1.50	1.68	1.92	2.14	-	-	-	-
825	-	-	-	-	-	-	1.32	1.38	1.47	1.61	1.80	2.01	2.29	2.65	-	-	-
850	-	-	-	-	-	1.33	1.41	1.48	1.55	1.69	1.90	2.14	2.43	2.81	3.45	-	-
875	-	-	-	-	-	1.40	1.46	1.57	1.61	1.78	1.98	2.18	2.60	2.94	3.59	4.52	-
900	-	-	-	-	1.43	1.48	1.56	1.64	1.71	1.87	2.05	2.38	2.57	3.04	3.52	4.31	5.30
925	-	-	-	1.49	1.48	1.56	1.62	1.67	1.76	1.91	2.19	2.41	2.67	3.15	3.55	4.48	5.91
950	-	-	-	1.53	1.56	1.63	1.68	1.78	1.84	2.02	2.16	2.51	2.78	3.24	3.68	4.68	5.34
975	-	-	1.51	1.64	1.62	1.65	1.73	1.88	1.94	2.05	2.22	2.60	2.89	3.39	3.89	4.82	5.75
1000	-	1.58	1.63	1.60	1.69	1.70	1.81	1.92	2.00	2.18	2.28	2.56	2.98	3.31	4.07	4.53	5.97
1025	1.64	1.64	1.65	1.68	1.74	1.76	1.84	1.91	2.02	2.14	2.45	2.72	2.89	3.61	4.01	4.80	6.05
1050	1.64	1.67	1.72	1.72	1.73	1.82	1.92	1.98	2.06	2.27	2.44	2.65	3.12	3.51	3.89	4.84	5.89
1075	1.70	1.69	1.74	1.77	1.77	1.88	1.94	2.04	2.13	2.30	2.52	2.81	3.00	3.57	4.16	5.13	6.29
1100	1.70	1.69	1.76	1.82	1.81	1.88	1.97	2.06	2.20	2.37	2.55	2.88	3.39	3.60	4.17	5.04	6.41
1125	1.75	1.81	1.81	1.87	1.89	2.01	2.02	2.20	2.25	2.43	2.66	2.90	3.26	3.77	4.31	5.05	6.31
1150	1.80	1.78	1.82	1.87	1.92	1.96	2.08	2.12	2.24	2.41	2.74	2.85	3.17	3.78	4.08	4.99	6.27
1175	1.80	1.85	1.85	1.88	1.96	2.02	2.09	2.16	2.33	2.45	2.59	2.93	3.46	3.61	4.12	5.06	5.95
1200	1.84	1.89	1.88	1.96	2.00	2.03	2.08	2.29	2.40	2.43	2.84	2.89	3.32	3.81	4.77	5.37	6.36

4.4. Molten LiCl-CsCl

Ionic conductivities of molten LiCl-CsCl at different compositions and temperature range of 625 – 1200 K have been calculated and listed in Table 4. For molten LiCl-CsCl system, the ionic conductivities increased with increasing temperature for all the components. At a certain temperature, the ionic conductivities exhibited a gradually increasing trend as the content of LiCl increased. Ionic conductivity expression for molten LiCl-CsCl system at 625 – 1200 K within the full component range has been fitted from the data in Table 4, as follows:

$$\lambda_{(\text{LiCl-CsCl})} = -2.5773 + 3.5567 \times 10^{-2}x + 5.3306 \times 10^{-3}T - 1.2145 \times 10^{-3}x^2 - 1.6002 \times 10^{-6}T^2 + 1.3107 \times 10^{-5}x^3. \quad (7)$$

Table 4. Ionic conductivities of molten LiCl-CsCl at different temperatures (units: S/cm)

LiCl, % Temp, K	0.00	6.25	12.50	18.75	25.00	31.25	37.50	43.75	50.00	56.25	62.50	68.75	75.00	81.25	87.50	93.75	100.00
625	-	-	-	-	-	-	-	-	-	0.62	-	-	-	-	-	-	-
650	-	-	-	-	-	-	-	-	-	0.74	-	-	-	-	-	-	-
675	-	-	-	-	-	-	-	-	0.77	0.79	0.93	-	-	-	-	-	-
700	-	-	-	-	-	-	-	0.77	0.84	0.88	1.03	-	-	-	-	-	-
725	-	-	-	-	-	-	-	0.84	0.91	1.00	1.14	1.30	-	-	-	-	-
750	-	-	-	-	-	-	0.91	0.93	1.00	1.07	1.25	1.38	-	-	-	-	-
775	-	-	-	-	-	-	0.96	0.98	1.04	1.18	1.27	1.50	1.72	-	-	-	-
800	-	-	-	-	-	0.98	1.01	1.06	1.12	1.22	1.41	1.58	1.86	-	-	-	-
825	-	-	-	-	-	1.08	1.11	1.18	1.23	1.33	1.41	1.62	1.91	2.34	-	-	-
850	-	-	-	-	1.13	1.15	1.19	1.22	1.28	1.37	1.56	1.70	2.06	2.45	2.89	-	-
875	-	-	-	1.17	1.20	1.18	1.22	1.25	1.32	1.41	1.61	1.81	2.13	2.62	3.12	3.99	-
900	-	-	1.18	1.20	1.17	1.27	1.30	1.33	1.42	1.49	1.66	1.93	2.23	2.71	3.25	4.01	5.30
925	-	1.18	1.24	1.21	1.27	1.26	1.33	1.35	1.47	1.58	1.73	2.00	2.23	2.73	3.44	4.21	5.91
950	1.29	1.28	1.31	1.31	1.34	1.34	1.39	1.43	1.53	1.60	1.76	2.06	2.45	2.70	3.38	4.23	5.34
975	1.30	1.34	1.32	1.34	1.37	1.38	1.43	1.51	1.62	1.73	1.86	2.14	2.45	2.95	3.59	4.58	5.75
1000	1.35	1.36	1.38	1.40	1.39	1.42	1.45	1.53	1.64	1.75	1.95	2.11	2.55	3.05	3.61	4.68	5.97
1025	1.38	1.40	1.41	1.45	1.48	1.45	1.51	1.60	1.69	1.84	1.96	2.24	2.42	2.95	3.75	4.69	6.05
1050	1.41	1.46	1.43	1.45	1.49	1.56	1.57	1.60	1.71	1.87	2.10	2.28	2.51	2.98	3.62	4.85	5.89
1075	1.47	1.45	1.49	1.46	1.54	1.53	1.62	1.72	1.74	1.89	2.13	2.34	2.71	3.16	3.96	4.78	6.29
1100	1.51	1.51	1.54	1.54	1.60	1.56	1.64	1.76	1.85	1.92	2.06	2.37	2.65	3.17	4.01	4.91	6.41
1125	1.54	1.52	1.55	1.53	1.54	1.66	1.67	1.76	1.87	1.96	2.10	2.37	2.77	3.28	3.92	4.91	6.31
1150	1.57	1.58	1.57	1.59	1.66	1.63	1.71	1.78	1.91	2.04	2.17	2.45	2.78	3.14	3.77	5.11	6.27
1175	1.59	1.61	1.60	1.61	1.66	1.66	1.75	1.85	1.86	2.08	2.28	2.43	2.83	3.21	4.08	4.51	5.95
1200	1.62	1.64	1.64	1.65	1.67	1.72	1.73	1.89	1.91	2.06	2.33	2.60	2.98	3.24	4.18	4.99	6.36

4.5. Molten NaCl-KCl

Table 5 recorded the calculated ionic conductivities of molten NaCl-KCl at different temperatures and compositions. The ionic conductivities increased with temperature for all the components, and the addition of NaCl into molten KCl developed its conductivity.

Similarly, the interactions between counter-ions were weakened with the addition of NaCl into molten KCl, and correspondingly the conductive performance was improved by NaCl. For molten KCl, the weakening effect of NaCl on the interactions between counter-ions was smaller than that of LiCl, so is its conductivity enhancement effect.

The fitted mathematical expression for the ionic conductivities of molten NaCl-KCl system at 950–1200 K within the full component range was as follows:

$$\lambda_{(\text{NaCl-KCl})} = -4.1450 + 1.1492 \times 10^{-3}x + 1.0665 \times 10^{-2}T - 3.4204 \times 10^{-5}x^2 - 4.1621 \times 10^{-6}T^2 + 4.9834 \times 10^{-7}x^3. \quad (8)$$

Table 5. Ionic conductivities of molten NaCl-KCl at different temperatures (units: S/cm)

NaCl, % Temp, K	0.00	6.25	12.50	18.75	25.00	31.25	37.50	43.75	50.00	56.25	62.50	68.75	75.00	81.25	87.50	93.75	100.00
950	-	-	-	-	-	-	-	-	2.38	-	-	-	-	-	-	-	-
975	-	-	-	-	-	-	2.43	2.42	2.52	2.55	2.54	-	-	-	-	-	-
1000	-	-	-	-	2.41	2.39	2.50	2.56	2.64	2.67	2.66	2.75	-	-	-	-	-
1025	-	-	-	2.50	2.42	2.43	2.55	2.57	2.70	2.72	2.77	2.79	2.94	-	-	-	-
1050	-	2.49	2.53	2.51	2.51	2.54	2.59	2.67	2.72	2.78	2.85	2.82	3.01	3.09	-	-	-
1075	2.54	2.53	2.57	2.54	2.56	2.60	2.65	2.74	2.70	2.71	2.81	2.88	2.94	3.06	3.14	3.19	-
1100	2.54	2.56	2.52	2.50	2.69	2.61	2.67	2.67	2.76	2.83	2.88	2.90	2.95	3.11	3.24	3.32	3.55
1125	2.59	2.54	2.48	2.66	2.62	2.64	2.77	2.73	2.78	2.81	2.87	3.04	3.10	3.16	3.40	3.58	3.48
1150	2.58	2.60	2.56	2.70	2.71	2.64	2.75	2.76	2.90	2.85	2.88	3.03	3.21	3.23	3.17	3.37	3.63
1175	2.66	2.71	2.69	2.72	2.71	2.72	2.84	2.79	2.88	2.87	2.95	3.02	3.12	3.24	3.37	3.57	3.56
1200	2.73	2.66	2.65	2.64	2.75	2.70	2.75	2.76	2.82	2.96	2.92	3.09	3.24	3.18	3.37	3.47	3.63

4.6. Molten NaCl-RbCl

Table 6 listed the ionic conductivities of molten NaCl-RbCl at different temperatures and compositions. With the gradual addition of NaCl or increase of temperature, the ionic conductivities of molten NaCl-RbCl system increased gradually.

Based on Table 6, the ionic conductivity expression for molten NaCl-RbCl system at 850–1200 K within the full component range has been fitted, as follows:

$$\lambda_{(\text{NaCl-RbCl})} = -2.3609 + 6.0959 \times 10^{-3}x + 5.8005 \times 10^{-3}T - 3.5140 \times 10^{-5}x^2 - 1.9049 \times 10^{-6}T^2 + 1.5195 \times 10^{-6}x^3. \quad (9)$$

Comparing molten LiCl-NaCl and NaCl-RbCl, that the ionic conductivities of molten LiCl-NaCl increased gradually with LiCl content and those of molten NaCl-RbCl decreased gradually with RbCl content, it could be concluded that the ionic conductivities of molten alkali chlorides could only be improved by the addition of a smaller metal alkali cation, while introducing a larger cation would deteriorate the conductive performance.

Table 6. Ionic conductivities of molten NaCl-RbCl at different temperatures (units: S/cm)

NaCl, % Temp, K	0.00	6.25	12.50	18.75	25.00	31.25	37.50	43.75	50.00	56.25	62.50	68.75	75.00	81.25	87.50	93.75	100.00
850	-	-	-	-	-	-	-	1.51	-	-	-	-	-	-	-	-	-
875	-	-	-	-	-	-	1.52	1.57	1.63	-	-	-	-	-	-	-	-
900	-	-	-	-	-	1.56	1.59	1.65	1.67	-	-	-	-	-	-	-	-
925	-	-	-	-	1.56	1.59	1.66	1.73	1.76	1.88	-	-	-	-	-	-	-
950	-	-	-	1.55	1.62	1.62	1.71	1.74	1.81	1.92	2.00	-	-	-	-	-	-
975	-	-	1.53	1.63	1.58	1.68	1.75	1.80	1.89	2.01	2.09	2.22	-	-	-	-	-
1000	-	1.57	1.63	1.67	1.69	1.73	1.78	1.90	1.95	2.03	2.18	2.28	2.43	-	-	-	-
1025	1.64	1.59	1.71	1.71	1.75	1.80	1.87	1.95	1.97	2.07	2.19	2.36	2.45	2.60	-	-	-
1050	1.64	1.69	1.71	1.76	1.75	1.82	1.88	1.96	2.03	2.11	2.22	2.39	2.54	2.71	2.90	-	-
1075	1.70	1.73	1.72	1.79	1.74	1.87	1.94	2.01	2.10	2.19	2.31	2.43	2.63	2.68	2.99	3.22	-
1100	1.70	1.77	1.82	1.80	1.85	1.96	2.02	2.07	2.13	2.15	2.30	2.47	2.56	2.79	2.88	3.27	3.55
1125	1.75	1.78	1.82	1.84	1.84	1.94	2.02	2.14	2.13	2.21	2.34	2.47	2.61	2.89	2.99	3.29	3.48
1150	1.80	1.85	1.85	1.91	1.98	1.99	2.08	2.09	2.12	2.31	2.49	2.60	2.68	2.79	3.04	3.20	3.63
1175	1.80	1.84	1.86	1.90	2.02	2.07	2.04	2.19	2.25	2.38	2.46	2.61	2.74	2.87	3.23	3.43	3.56
1200	1.84	1.84	1.93	1.95	1.97	2.04	2.07	2.14	2.26	2.37	2.52	2.66	2.87	2.93	3.09	3.38	3.63

4.7. Molten NaCl-CsCl

Ionic conductivities for molten NaCl-CsCl at different temperatures and compositions have been calculated and recorded in Table 7.

The ionic conductivities of molten NaCl-CsCl presented positive dependences on both temperature and NaCl content due to the interaction weakening phenomenon of Na-Cl and Cs-Cl pairs. The following mathematical expression of ionic conductivity for molten NaCl-CsCl system at 800–1200 K within full composition range has been fitted based on Table 7:

$$\lambda_{(\text{NaCl-CsCl})} = -2.0551 + 4.9090 \times 10^{-3}x + 4.9782 \times 10^{-3}T - 1.0142 \times 10^{-4}x^2 - 1.5866 \times 10^{-6}T^2 + 2.5080 \times 10^{-6}x^3. \quad (10)$$

In Eq. 10 x denotes the mole fraction of NaCl, T denotes temperature.

5 CONCLUSIONS

To reveal the effects of temperature and composition on melt conductivity, ionic conductivities of molten LiCl-NaCl, LiCl-KCl, LiCl-RbCl, LiCl-CsCl, NaCl-KCl, NaCl-RbCl, and NaCl-CsCl in the temperature range of near melting point-1200 K within full composition range have been calculated with equilibrium molecular dynamics

simulation method.

1. The ionic conductivity of molten LiCl-NaCl, LiCl-KCl, LiCl-RbCl, LiCl-CsCl, NaCl-KCl, NaCl-RbCl, and NaCl-CsCl exhibited positive dependences on temperature, i.e., increased with the increasing of temperature.
2. LiCl can improve the conductive performances of other molten alkali chlorides. With the gradual increase of LiCl content, ionic conductivities of molten LiCl-NaCl, LiCl-KCl, LiCl-RbCl, and LiCl-CsCl increased monotonously. NaCl could also enhance the conductive performances of molten KCl, RbCl, and CsCl.
3. For molten alkali chlorides, adding a smaller alkali metal cation could effectively improve the ionic conductivity. The smaller the added cations are, the more significant the effects are. While introducing a larger alkali cation would deteriorate the conductive performance.
4. Simple databases for ionic conductivities of molten LiCl-NaCl, LiCl-KCl, LiCl-RbCl, LiCl-CsCl, NaCl-KCl, NaCl-RbCl, and NaCl-CsCl have been built based on the simulated results. Corresponding mathematical equations for ionic conductivities on temperature and composition dependences have also been fitted according to the calculated results.

Table 7. Ionic conductivities of molten NaCl-CsCl at different temperatures (units: S/cm)

NaCl, % Temp, K	0.00	6.25	12.50	18.75	25.00	31.25	37.50	43.75	50.00	56.25	62.50	68.75	75.00	81.25	87.50	93.75	100.00
800	-	-	-	-	-	1.05	1.07	-	-	-	-	-	-	-	-	-	-
825	-	-	-	-	-	1.06	1.20	-	-	-	-	-	-	-	-	-	-
850	-	-	-	-	1.16	1.17	1.21	1.20	-	-	-	-	-	-	-	-	-
875	-	-	-	1.22	1.20	1.21	1.25	1.26	1.38	-	-	-	-	-	-	-	-
900	-	-	1.20	1.18	1.24	1.27	1.29	1.29	1.45	1.59	-	-	-	-	-	-	-
925	-	1.20	1.29	1.22	1.30	1.35	1.39	1.40	1.54	1.59	1.66	-	-	-	-	-	-
950	1.29	1.33	1.33	1.32	1.34	1.40	1.43	1.43	1.56	1.63	1.79	-	-	-	-	-	-
975	1.30	1.30	1.39	1.37	1.41	1.45	1.51	1.46	1.63	1.70	1.81	1.92	-	-	-	-	-
1000	1.35	1.38	1.40	1.42	1.43	1.46	1.55	1.53	1.63	1.76	1.89	1.97	2.14	-	-	-	-
1025	1.38	1.42	1.44	1.44	1.49	1.54	1.60	1.52	1.68	1.82	1.86	2.02	2.26	2.43	-	-	-
1050	1.41	1.42	1.46	1.48	1.52	1.52	1.63	1.61	1.80	1.80	1.96	2.13	2.24	2.46	2.74	-	-
1075	1.47	1.48	1.52	1.53	1.52	1.63	1.64	1.63	1.76	1.91	2.02	2.13	2.29	2.47	2.74	3.03	-
1100	1.51	1.51	1.55	1.55	1.62	1.62	1.69	1.65	1.85	1.93	2.05	2.21	2.37	2.54	2.86	3.24	3.55
1125	1.54	1.58	1.57	1.59	1.64	1.66	1.69	1.74	1.89	1.96	2.14	2.26	2.36	2.69	2.77	3.12	3.48
1150	1.57	1.59	1.59	1.66	1.68	1.69	1.77	1.76	1.93	2.05	2.11	2.27	2.43	2.68	2.87	3.15	3.63
1175	1.59	1.62	1.60	1.64	1.71	1.72	1.80	1.80	1.94	2.05	2.17	2.26	2.42	2.63	2.88	3.35	3.56
1200	1.62	1.65	1.68	1.64	1.71	1.77	1.82	1.81	1.95	2.02	2.22	2.28	2.51	2.73	2.97	3.35	3.63

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