Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supporting Information

Insights into the local structure evolution and thermophysical properties of NaCl-KCl-$MgCl_2$-$LaCl_3$ melt driven by Machine Learning

S1. Experimental and Materials

NaCl (Aladdin, >99.5%), KCl (Aladdin, >99.5%), $MgCl_2$ (Collins, >99%), and $LaCl_3$ (KAIWEI, >99%) were purchased from Shanghai Aladdin Biochemical Technology Co., Ltd., Shanghai Gaoxin Chemical Glass Instrument Co., Ltd., and Shanghai Kewei Chemical Technology Co., Ltd., respectively. With a homemade apparatus, the densities of NKML molten salts were determined for each sample from 923 K to 1073 K in 50 K intervals, according to Archimedes' principle. For the experiments, a given volume of titanium sphere was immersed in 110 g molten salt under the pull of a platinum wire (d = 0.3 mm), and the change in mass, which can deduce the density shown as Equation (S-1), was monitored by an electronic balance (Sartorius BSA224S-CW).

$$\rho = \frac{\Delta m}{V_0[1+\alpha(T-298)]^3} \tag{S-1}$$

Where ρ is the density of the melt, Δm is the difference between the balance before and after complete submersion of the titanium sphere, V_0 is the corrected volume of the titanium sphere and α is the coefficient of linear expansion of the platinum.

During the measurement, the traction of the platinum wire also entered the melt, so the titanium sphere volume needs to be corrected at room temperature using deionized water, as shown in Equation (S-2).

$$V_0 = \frac{\Delta m'}{\rho_w} + \pi D \sigma_w / \rho_w g \qquad \text{(S-2)}$$

Where $\Delta m'$ is the difference between the indicated balance values of the platinum wire and the titanium ball before and after immersion in deionized water. σ_ω, ρ_ω, D, and g are the surface tension, density, and platinum wire diameter of the deionized water, and the acceleration of gravity, respectively.

This work measured the shear viscosity of NKML melts supported by a rotational rheometer (EC twist 502 rheometer, Anton Paar) based on the concentric cylinders principle, with a hollow cylinder as the outer column, and a rotor as the inner column. During the measurement, the gap between the cylinder and rotor filled with molten salt, and the rotor rotated at a constant rate driving the melt flowing, while the cylinder was stationary. In the radial direction, the velocity gradient of the melt yielded torque on the rotor. Combining the geometric parameters of the equipment and Newton's law of viscosity, the shear viscosity of the molten salt is determined as Equation (S-3).

$$\eta = \frac{M}{4\pi(h+c)\omega}\left(\frac{1}{R_i^2} - \frac{1}{R_a^2}\right) \qquad \text{(S-3)}$$

Where M is the torque on the rotor, h is the depth of the rotor immersion in the melt, c is extra depth resulting from rotor cross-section, ω is the angular rate of rotation, R_i is the outer diameter of the rotor, and R_a is the cylinder inner diameter. During the measurement, 15 g of molten salt was analyzed in an argon (99.99%) atmosphere from

973 K at 2 K/min to 1023 K with a shear rate of 50 s^{-1}.

Table S1 coordination number comparison by AIMD and DPMD simulation for S1 at 973 K.

Coordination numbers	AIMD	DPMD
Na-Cl	5.34	5.76
K-Cl	6.84	7.48
Mg-Cl	4.74	4.76
La-Cl	6.70	7.31

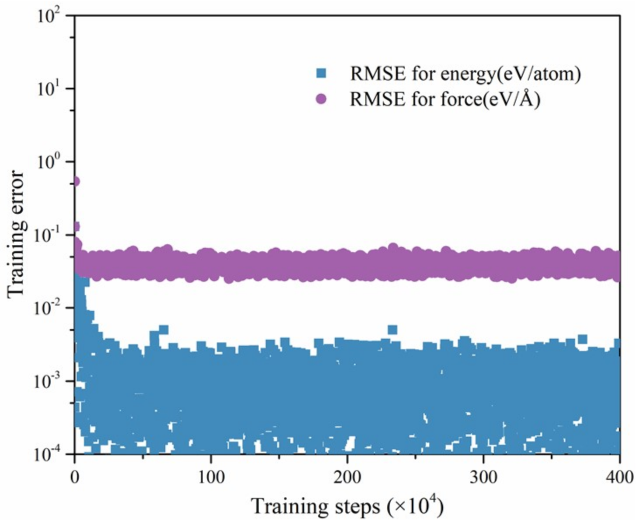

Figure S1 Evolution of root-mean-square errors of energy and force during training for NaCl-KCl- $MgCl_2$-$LaCl_3$ molten salt.

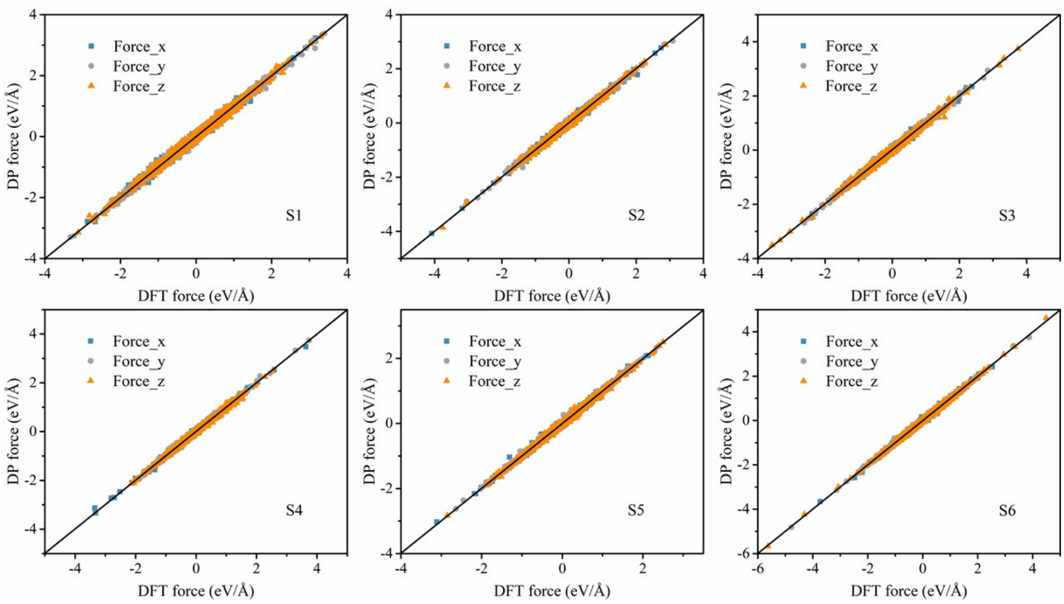

Figure S2 Force comparison between DFT calculation and DP prediction for NaCl-KCl-$MgCl_2$-$LaCl_3$ melts with different compositions.

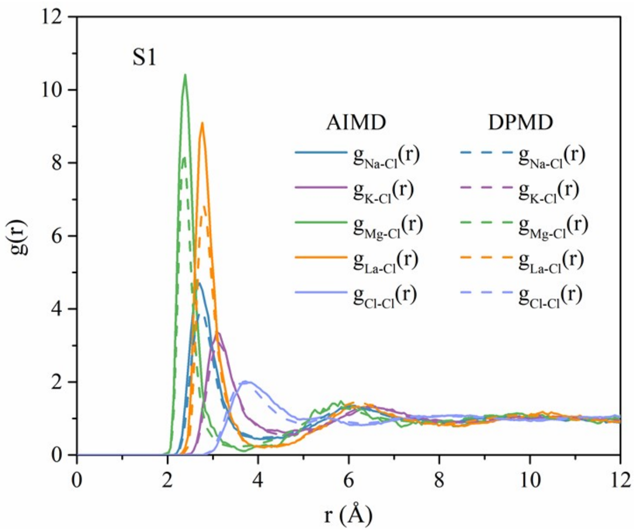

Figure S3 Comparison of PRDFs predicted using DPMD and AIMD at 973 K for S1.

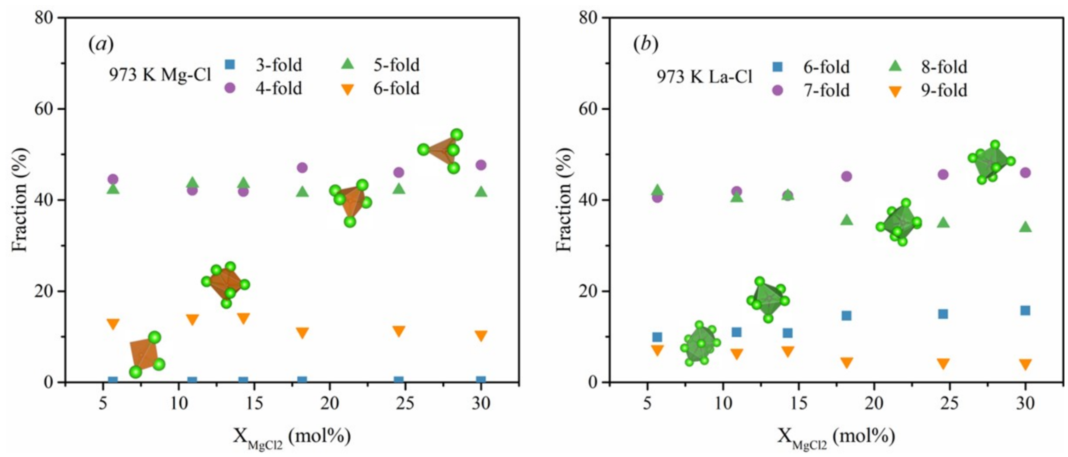

Figure S4 Evolution of Mg-Cl coordination number of Mg-Cl and La-Cl for different $MgCl_2$ concentrations at 973 K.

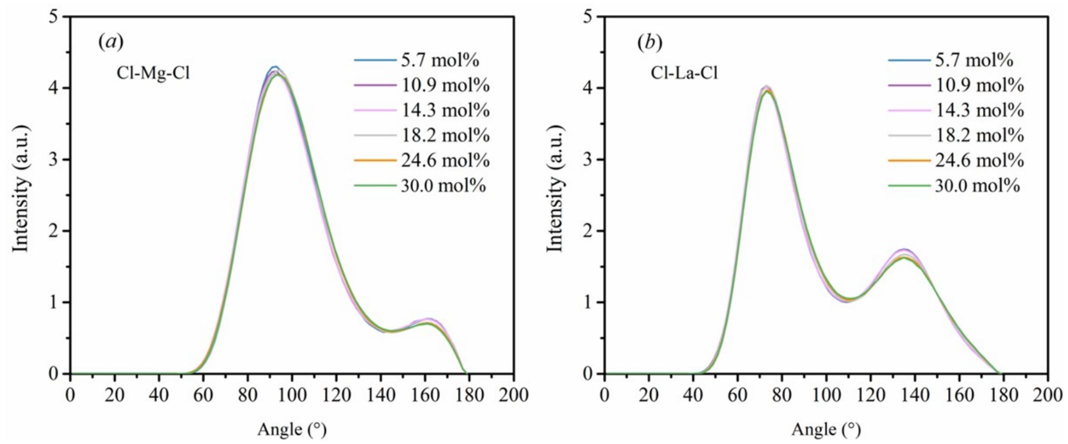

Figure S5 Evolution of ADFs of Cl-Mg-Cl, Cl-La-Cl with $MgCl_2$ concentration. (*a*) is for Cl-Mg-Cl, (*b*) is for Cl-La-Cl.

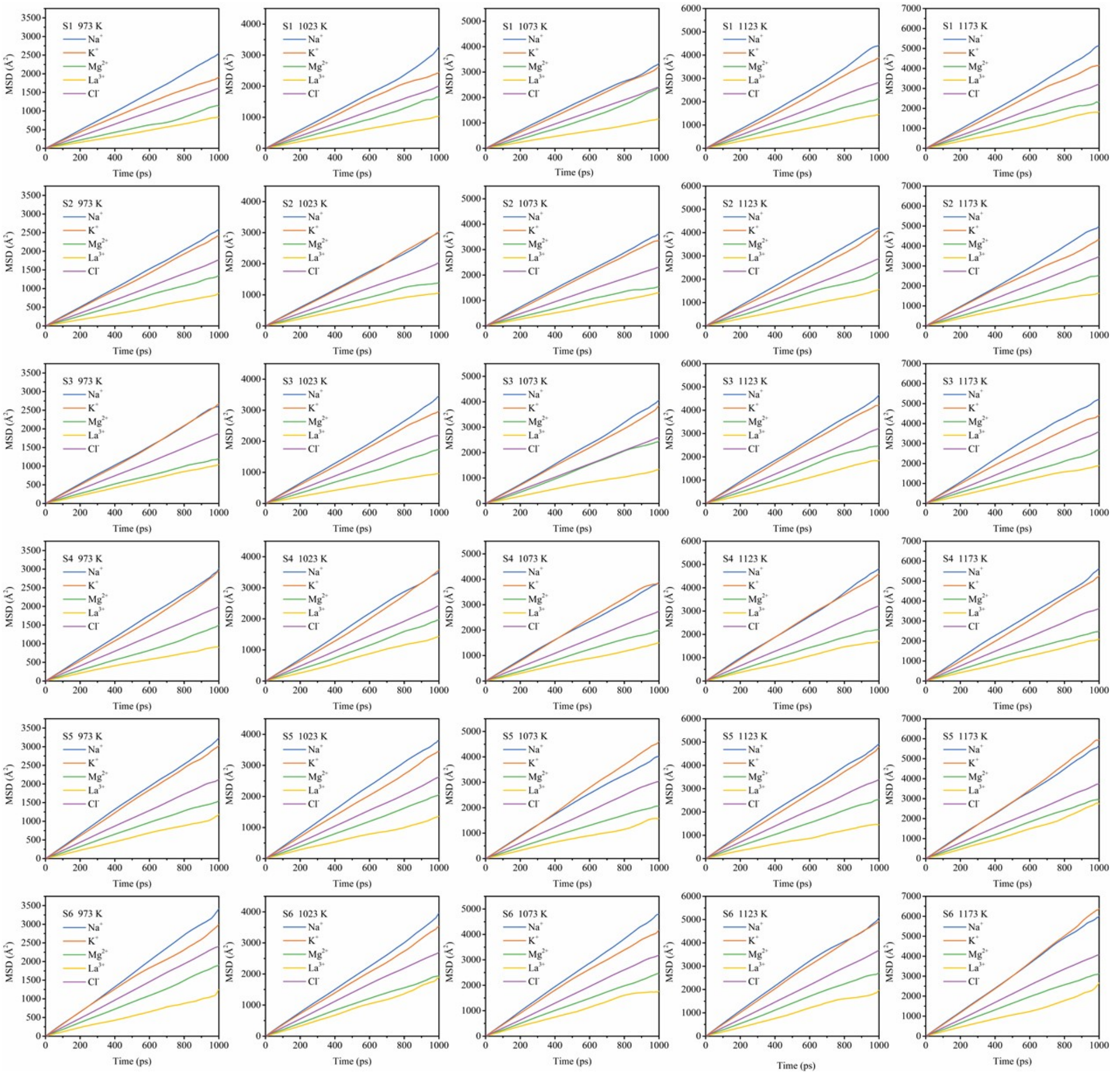

Figure S6 Mean square displacement of all ions in the NaCl-KCl-$MgCl_2$-$LaCl_3$ molten salt for different components from 973 K to 1173 K.

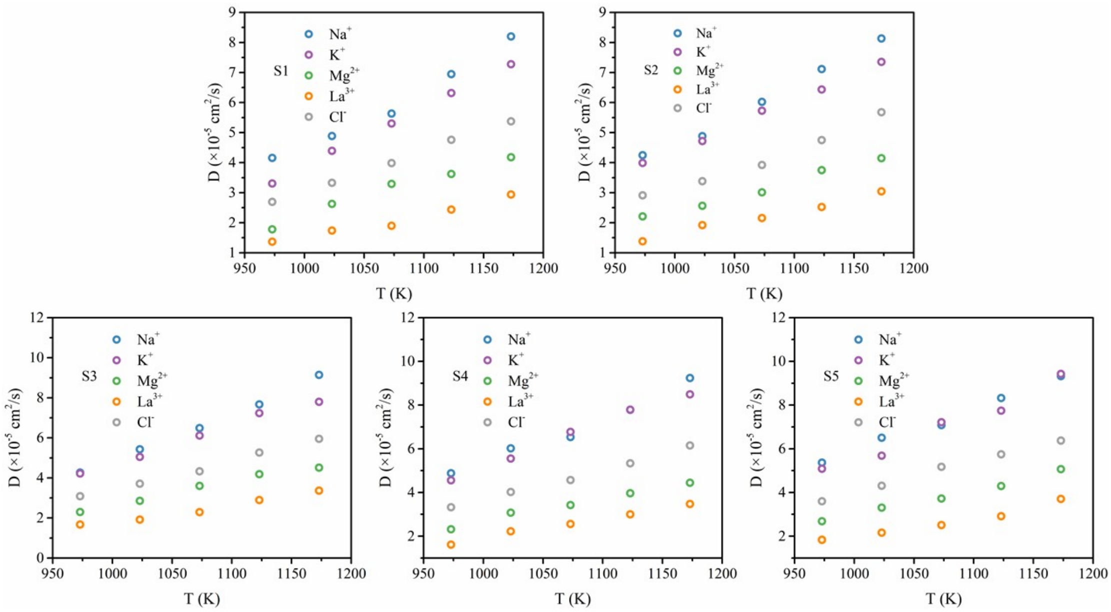

Figure S7 Self-diffusion coefficients of all ions in the NaCl-KCl-$MgCl_2$-$LaCl_3$ system at different temperatures as a function of temperature for S1 to S5.