

SUPPORTING INFORMATION

Ionic Dynamics of Charge Carrier in Layered Solid Materials for Mg Rechargeable Batteries

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Text S1: Moment tensor potential (MTP)¹ training and validation details

The MTP models were parameterized on both DFT-calculated energies and DFT-calculated forces, with the energies given 100 times the weight of the forces as suggested by previous benchmark tests.² The maximum level of basis functions, lev_{max} ,³ which is related to the maximum polynomial degree, was chosen to be 12. To account for long-range electrostatic interactions in the layered structures, MTP cutoff radius R_{cut} that defines individual atomic neighborhoods was set at 7 Å.

During the dynamic evolution of the structure during MTP-MD, MTP re-training was conditionally activated by the occurrence of an “unlabeled” configuration as determined by the D-optimality criterion,⁴ corresponding to the *extrapolation grade*⁵ parameter coded in MLIP. The MTP-MD was terminated once the extrapolation grade exceeds the selection threshold, which we set to 10, consistent with recent publications.⁶ Subsequently, structures that has extrapolation grade above 1.5 were selected from the molecular dynamics trajectory to be added to the DFT training set. The MTP was re-trained on the updated training set and MTP-MD run was restarted with the new potential.

After iteration of active learning, the fitted MTP potential model can carry out nanosecond long MD production without extrapolation. We validate the fitted MTP with DFT on 100 snapshots evenly selected from 2-ns LOTF-MD at 1000 K. The fitting and validation error of three systems in this study are summarized in Table S1.

Text S2: Statistical error analysis and weighted Arrhenius fitting

He, X. F. *et al.*, defined the relative standard deviation as the ratio between the standard deviation of the diffusivity and the diffusivity itself.⁷ Through a series of benchmarks they fit the following equation for the relative standard deviation (RSD) of ionic diffusivities:

$$RSD = \frac{\sigma_D}{D_{true}} = \frac{3.43}{\sqrt{N_{eff}}} + 0.04 \quad (1)$$

where

$$N_{eff} = \frac{\max[\text{TMSD}(\Delta t)]}{a^2} \quad (2)$$

where $\text{TMSD}(\Delta t)$ is the total mean squared displacement (the sum of the mean squared displacements of the lithium atoms) after time Δt and a is the length of a site-to-site ion hop.

To estimate the uncertainty in the calculated activation energies we start from the Arrhenius equation

$$D(T) = D_0 e^{-\frac{E_a}{k_B T}} \quad (3)$$

where $D(T)$ is the diffusivity at temperature T , D_0 is the diffusivity in the limit of infinite temperature, and E_a is the activation energy for diffusion. Taking the natural log of both sides gives an equation that is linear in E_a :

$$\ln(D(T)) = \ln(D_0) - \frac{E_a}{k_B T} \quad (4)$$

The variance of $\ln(D(T))$, $\sigma_{\ln(D(T))}^2$, is approximately related to the variance in $D(T)$, $\sigma_{D_T}^2$, through the following equation:⁸

$$\sigma_{\ln(D(T))}^2 \approx \frac{\sigma_{D_T}^2}{D_T^2} \quad (5)$$

where we estimate $\sigma_{D_T}^2$ using the equation of He, X. F., et al.⁷ (Equation Δt). We estimate E_a by performing a weighted least squares regression⁹ of $\ln(D(T))$ with respect to $\frac{1}{k_B T}$, where the weight of each sample is the inverse of $\sigma_{\ln(D(T))}^2$ for that sample. E_a is a coefficient of the linear

estimator, and the variance of E_a is taken as the square of the standard error of the coefficient. The weighted least squares regression and standard error of the coefficient were calculated using the SciPy package.¹⁰

Text S3: Estimation of diffusivity upper limit at 1000 K

If no diffusive hops have been observed in 20 *ns*, we estimate the upper limit of diffusivity by assuming that Mg-ion undergoes a site-to-site hopping exactly every 20 nanoseconds. The materials in this study have a typical hopping distance between adjacent Mg favorable sites of about 3 Å. Using equation (1) in the main text we estimate the upper limit on Mg diffusivity at 1000K:

$$D = \frac{1}{6} \times \frac{(3\text{\AA})^2}{200 \text{ ns}} = 7.5 \times 10^{-9} \text{ cm}^2 / \text{s}$$

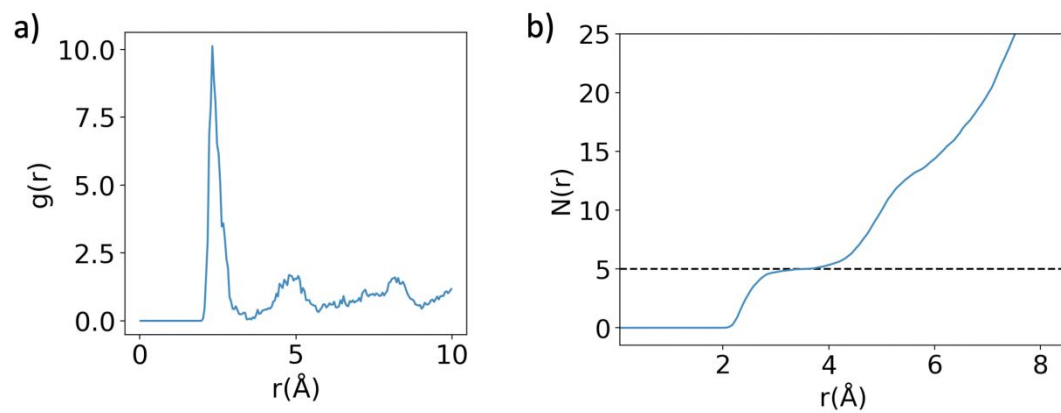


Figure S1. a) Radial distribution function $g(r)$ and b) Coordination number analysis $N(r)$ of Mg-Cl pair in the $\text{MgCl}^+/\text{BiOCl}$ system.

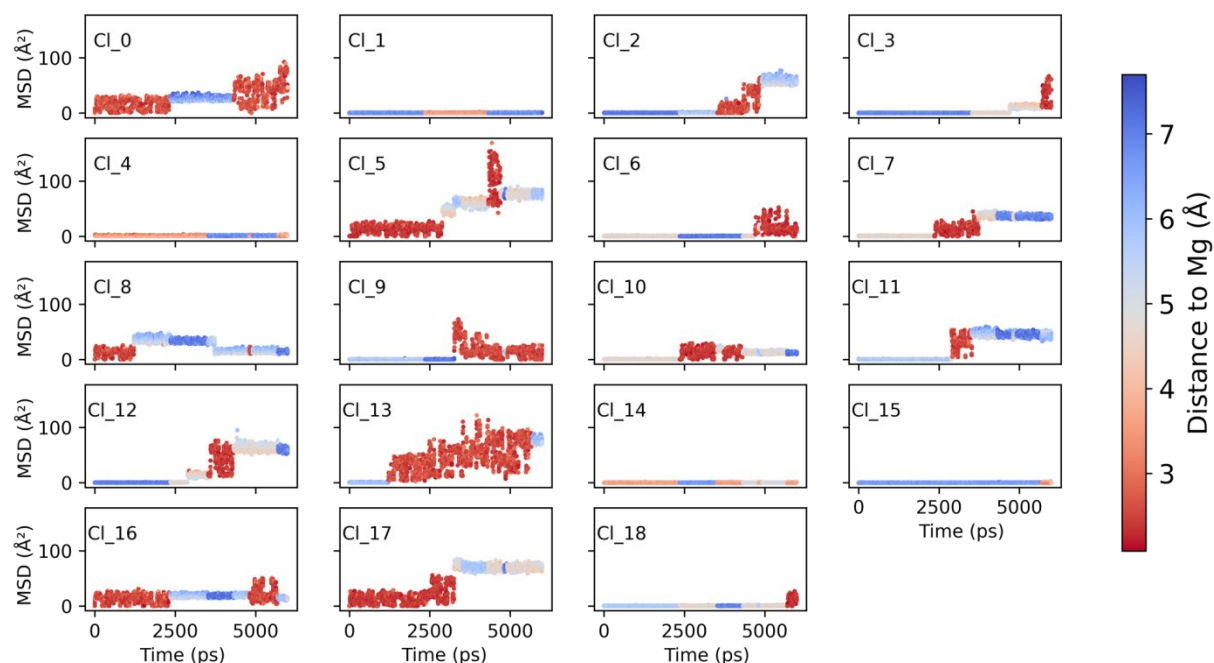


Figure S2. Mean square displacement (MSD) as a function of time for Cl ions during the 6-ns trajectory. Within each subplot, the x axis represents time and the y axis the magnitude of MSD in \AA^2 with reference to its initial location. The warmer color represents Cl ion closer to the Mg ion while the cooler the farther to the Mg ion. Diffusional hopping of several Cl ions occurred during 6-ns MD production.

Table S3. Mean absolute errors (MAE) for predictions of energy and forces by MTP with reference to DFT.

System	Dataset	Number of configurations	Energy MAE (meV/atom)	Force MAE (meV/Å)
BiOCl/MgCl ⁺	Training	8144	1.86	79.2
	Validation	100	3.06	81.6
ScOCl/MgCl ⁺	Training	4615	0.90	65.5
	Validation	100	2.73	74.5
BiOBr/MgBr ⁺	Training	6744	1.76	52.3
	Validation	100	4.03	54.2

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