

Machine learning based analysis of collective diffusion in inorganic solid-state electrolytes

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STATE OF PURPOSE

Despite playing a central role in the design of high performance solid-state electrolytes (SSE), little is known about the processes governing ionic diffusion in these materials and the spatio-temporal correlations acting on migrating particles. Computer simulations can reproduce the trajectories of individual diffusing ions in real time with extraordinary accuracy, thus providing incredibly valuable atomistic data that in practice cannot be resolved by experiments.

However, the identification of hopping events in computer simulations typically relies on active supervision and definition of arbitrary material-dependent geometrical parameters, thus frustrating high throughput screenings of diffusing paths and mechanisms across simulation databases and the assessment of many-diffusing-ion correlations.

RESULTS

Here, we introduce a novel approach for analysing ion hopping events in molecular dynamics (MD) simulations in a facile and totally unsupervised manner, what would allow the extraction of completely new descriptors related to these diffusions. Our approach relies on the k-means clustering algorithm and allows to identify with precision which and when particles diffuse in a simulation and the exact migrating paths that they follow as well, as it is exemplified in Figure 1.

We apply such a powerful analysis tool to a comprehensive database of density functional theory *ab initio* MD (DFT-AIMD) simulations, analyzed in

a previous work [1], comprising several families of SSE and tens of millions of atomic configurations. By doing this, we are able to (1) quantify correlations between many diffusing ions, (2) identify predominant collective migrating mechanisms and (3) determine how specific and novel migration descriptors such as the length and duration of individual ionic hops correlate with the overall ion diffusion coefficients or structural properties of the material, all under realistic finite-temperature conditions.

For instance, the distribution of lengths of diffusions can be easily extracted for each family and the database in general, presenting slightly different trends, as shown in Figure 2. Gaining such insights points towards new, more efficient SSE, provided the close relation between this magnitudes and their diffusion performance.

Concretely, we show that the probability of finding N-correlated particles in a simulation decreases exponentially with N, proving that these correlations do not depend on temperature and are mostly pairwise for SSE, as shown in Figure 3. The fundamental mechanisms involved in such processes are also depicted and analysed.

DATA AND CODE AVAILABILITY

The analysis scripts resulting from this study have been made publicly available as a Python package, that is user-friendly and easily adaptable [2] to any desired MD code output, as well as the DFT-AIMD database [3].

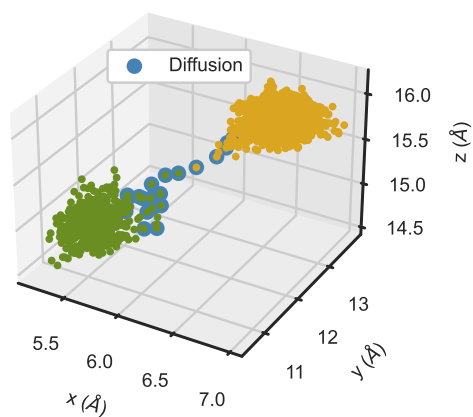


Fig. 1. Example of the performance of our unsupervised algorithm at extracting the diffusive path for one random particle of a DFT-AIMD simulation of $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ at a temperature of 400K.

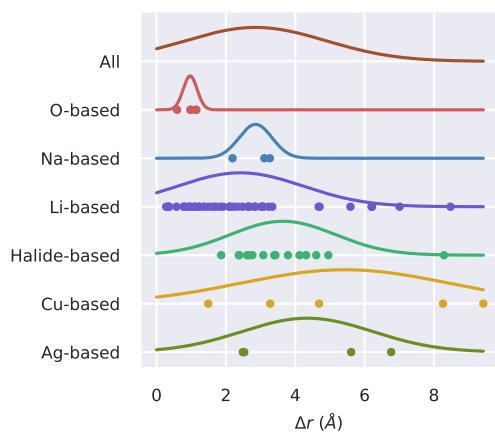


Fig. 2. Distribution of spatial lengths of diffusion for each family and the general DFT-AIMD database studied in the present work (labels from the left, accounting 'All' for the distribution of the set of all spatial lengths). This figure shows different tendencies for each family, as well as it exemplifies one of the analysis that can be easily extracted with the present approach.

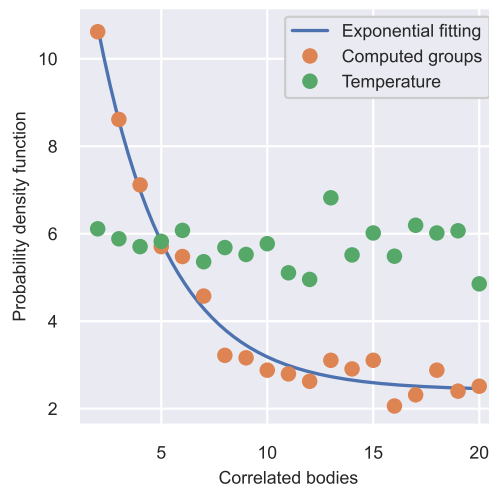


Fig. 3. The probability density function of finding N -correlated bodies for a MD simulation in SSE decreases exponentially with the number of involved bodies, independently of the temperature, with a fair predominance of pairwise correlations.

REFERENCES

- [1] C. López, A. Emperador, E. Saucedo, *et al.*, *Universal ion-transport descriptors and classes of inorganic solid-state electrolytes*, *Materials Horizons*, 2023, doi: 10.1039/D2MH01516A
- [2] C. López, R. Rurali, C. Cazorla, *Repository with all the developed codes*, 2023, url: <https://github.com/IonRepo/IonDiff>
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