



Research Highlights

Unsupervised machine learning accelerates solid electrolyte discovery

Xu Zhang, Bin Tang, Zhen Zhou*

School of Materials Science and Engineering, Computational Centre for Molecular Science, Institute of New Energy Material Chemistry, Key Laboratory of Advanced Energy Materials Chemistry (Ministry of Education), Renewable Energy Conversion and Storage Center (ReCast), Nankai University, Tianjin 300350, China

Received 24 November 2019; revised 4 December 2019; accepted 5 December 2019

Traditional organic liquid electrolytes used in commercial Li-ion batteries would incur serious safety issues due to their flammability and volatility [1]. The exploration and design of solid electrolytes with high room-temperature Li-ion conductivities (σ_{RT}) are important to improve the safety and cycle life of Li-ion batteries [2]. Although previous investigations have proven that various physical factors correlate with Li-ion diffusion in solids, there is no unified theory to explain the similarity among distinctive crystal structures of solid-state Li-ion conductors (SSLCs). In addition, the exploration of a vast composition-structure space of thousands of materials is extremely difficult. The current investigations mainly rely on ‘trial and error’ and are limited to a few kinds of candidates, such as lithium thiophosphates, garnets, sodium super ionic conductors (NASICONs), perovskites and argyrodites [3]. Therefore, the discovery of novel SSLCs from wider materials on basis of available knowledge is significant.

Machine learning (ML) methods have the potential to greatly improve the efficiency and accuracy of materials exploration due to its capability to find the complex patterns hidden behind high-dimensional data. A good predictive ML model should be trained with a sufficient amount of data but the data size for SSLCs is quite small and limited to several widely investigated materials. Also, ML models are generally sensitive to the variances and errors of the training data, and the Li-ion conductivity obtained in experiments could be affected by various factors and then varies greatly. The insufficiency of high-quality data for SSLCs limits the application of supervised ML techniques to capture the structure–property relationship and accurately predict the ionic conductivity for thousands of compounds.

To address these issues, researchers from Toyota Research Institute of North America and University of Maryland, USA,

adopted an unsupervised learning scheme to screen and classify all known Li-containing compounds from Inorganic Crystal Structure Database (ICSD) [4]. The labels of training data for unsupervised learning can be unknown while for supervised learning, the training data should be well labeled. In their work, unsupervised ML models were used to cluster the materials into groups with high or low ionic conductivity. Then, they used high-throughput *ab initio* molecular dynamics (AIMD) simulations to investigate σ_{RT} of potential SSLCs with high ionic conductivity. The corresponding workflow is shown in Fig. 1a.

The researchers used digital diffraction patterns of crystal structures in which the representation for each crystal structure was built according to Bragg's law to describe the periodic crystal lattice with a set of X-ray diffraction (XRD) intensities. Note that only the anion lattice of the crystal structure was considered here. The modified XRD (mXRD) was used as features for unsupervised ML. The researchers proposed that the compounds in group V and VI (Fig. 1b) show the characteristics of moderately distorted anion lattices, which is closely related to the disordered Li sublattice and could be attributed to high Li-ion conductivity.

Then they systematically computed Li-ion conductivity for the compounds from these fast-conducting groups with AIMD simulations. Finally, sixteen novel compounds with σ_{RT} higher than 10^{-4} S cm⁻¹ were obtained. Especially, Li₈N₂Se, Li₆K-BiO₆ and Li₅P₂N₅ possess σ_{RT} as high as over 10^{-2} S cm⁻¹. These novel SSLCs possess greatly different structures and chemical compositions compared with well-known fast SSLCs and the results also reveal the structure–property relationship between Li-ion conduction and anion lattice.

Compared with previous results based on supervised ML models for the direct prediction of ionic conductivity in solids [5], the unsupervised model in this work was used to cluster the materials and required fewer features for learning and the model is less susceptible to variances and errors in the dataset,

* Corresponding author.

E-mail address: zhouzhen@nankai.edu.cn (Z. Zhou).

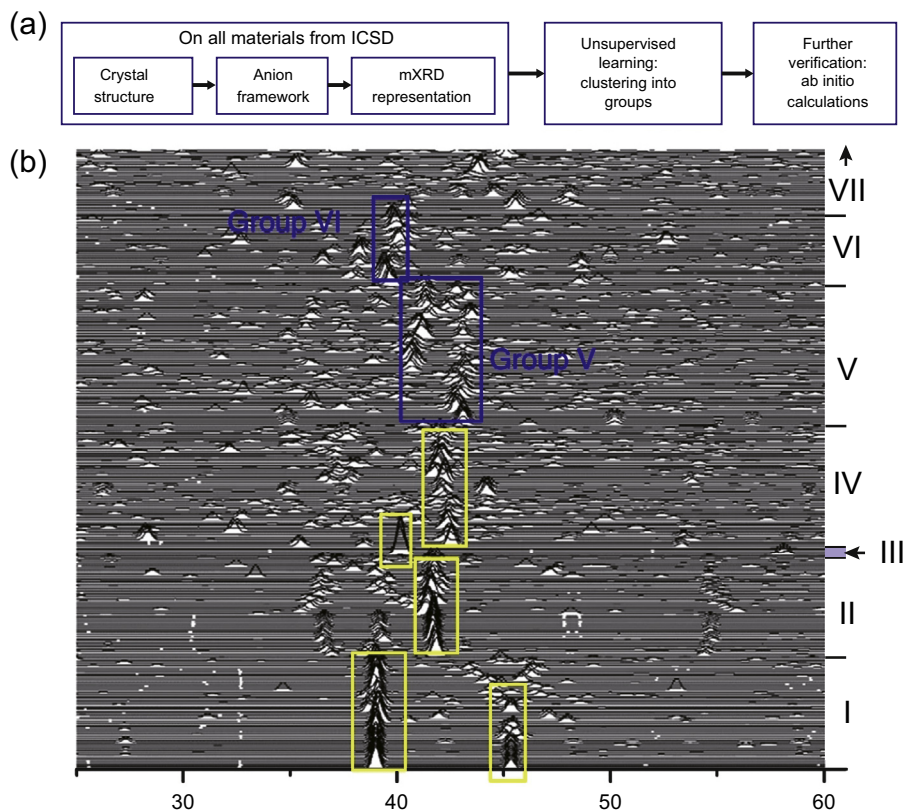


Fig. 1. (a) Workflow of the unsupervised ML for guiding discovery of SSLCs. (b) mXRD of materials in group I–VII. Reprinted from Ref. [4].

which is appropriate for the fields with scarce data. However, since only the anion lattice was considered, some compounds uncovered by this grouping did not exhibit fast Li-ion diffusion and further investigations including more features should be performed. At the current stage only the ionic conductivity was primarily checked computationally, and other performances should be investigated via computations and particularly experiments to evaluate whether the proposed novel SSLCs meet the desired attributes for commercial solid electrolytes. Recently, unsupervised learning has been more widely used in materials science such as novel materials discovery [6], feature extraction and phase mapping. This work not only provides a strong scheme for solid electrolyte discovery but also demonstrates a new research paradigm for functional materials innovation.

Conflict of interest

There are no conflicts of interest to declare.

Acknowledgements

This work was supported by NSFC (21933006) in China.

References

- [1] Z. Zhang, Y. Shao, B. Lotsch, Y.-S. Hu, H. Li, J. Janek, L.F. Nazar, C.-W. Nan, J. Maier, M. Armand, L. Chen, *Energy Environ. Sci.* 11 (2018) 1945–1976.
- [2] J.C. Bachman, S. Muy, A. Grimaud, H.H. Chang, N. Pour, S.F. Lux, O. Paschos, F. Maglia, S. Lupart, P. Lamp, L. Giordano, Y. Shao-Horn, *Chem. Rev.* 116 (2016) 140–162.
- [3] Y. Wang, W.D. Richards, S.P. Ong, L.J. Miara, J.C. Kim, Y. Mo, G. Ceder, *Nat. Mater.* 14 (2015) 1026–1031.
- [4] Y. Zhang, X. He, Z. Chen, Q. Bai, A.M. Nolan, C.A. Roberts, D. Banerjee, T. Matsunaga, Y. Mo, C. Ling, *Nat. Commun.* 10 (2019) 5260.
- [5] H. Wang, Y. Ji, Y. Li, *WIREs Comput. Mol. Sci.* (2019), e1421.
- [6] W. Sun, C.J. Bartel, E. Arca, S.R. Bauers, B. Matthews, B. Orvananos, B.R. Chen, M.F. Toney, L.T. Schelhas, W. Tumas, J. Tate, A. Zakutayev, S. Lany, A.M. Holder, G. Ceder, *Nat. Mater.* 18 (2019) 732–739.