Development of crystal structure prediction methods using machine learning

Tomoki YAMASHITA

Top Runner Incubation Center for Academia-Industry Fusion,

Nagaoka University of Technology

1603-1 Kamitomioka-machi, Nagaoka, Niigata, 940-2188

We search for solid electrolyte materials that can be synthesized in the systems of Li-Mg-SiO_4 using crystal structure prediction (CSP) by CrySPY [1]. We investigate whether it is possible to synthesize structures with high ionic conductivity by changing the ratio of Li to Mg from Li_2MgSiO_4. We performed CSP simulations with random search. In the CSP simulations, first-principles calculations were carried out using the VASP code [2], and a generalized gradient approximation was employed for exchange-correlation functional. The compositions calculated in this study were Li_2MgSiO_4, $Li_{1.5}Mg_{1.25}SiO_4$, and $Li_{2.5}Mg_{0.75}SiO_4$. The number of searching trials for each composition was 200, which were conducted using the ISSP supercomputer. Our calculated formation energies showed that Li_2MgSiO_4 can be synthesized, however, $Li_{1.5}Mg_{1.25}SiO_4$ and $Li_{2.5}Mg_{0.75}SiO_4$ are unlikely to be synthesized, leading to phase separation. These results are consistent with our experiments. Therefore, it is difficult to synthesize solid electrolytes in the system of Li-Mg-SiO_4 except for Li_2MgSiO_4.

We have also studied on algorithm development for CSP. Look Ahead based on Quadratic Approximation (LAQA), which is one of the selection-type algorithms we previously developed [3], can control the optimization priority of the candidates. We have proposed an improved score of LAQA, where the stress term is added to overcome the drawbacks of the previous score. CSP simulations by this improved algorithm are performed to investigate the searching efficiency for typical materials such as Si (16 atoms), Al_2O_3 (10 atoms), NaCl (16 atoms), and $SrCO_3$ (20 atoms). We used the CrySPY code for the CSP simulations. For the system of $SrCO_3$, we employed the VASP code to evaluate the energy and to carry out the local structure optimization using the ISSP supercomputer. For the other systems, the QUANTUM ESPRESSO was also used. The total-energy calculations were performed using the density functional theory with the projector-augmented wave method. The generalized gradient approximation was used for exchange-correlation functional. Figure 1 illustrates the

number of total optimization steps required to find the most stable structure, obtained by the CSP simulations using random search (RS), Greedy, and LAQA. The searching efficiencies of LAQA with the weight parameter of stress w_S of 10 (red bars in Fig. 1) are not always the best, however, it demonstrates high searching efficiency on various data sets. In other words, the introduction of the stress term makes LAQA more robust and versatile [4].

References

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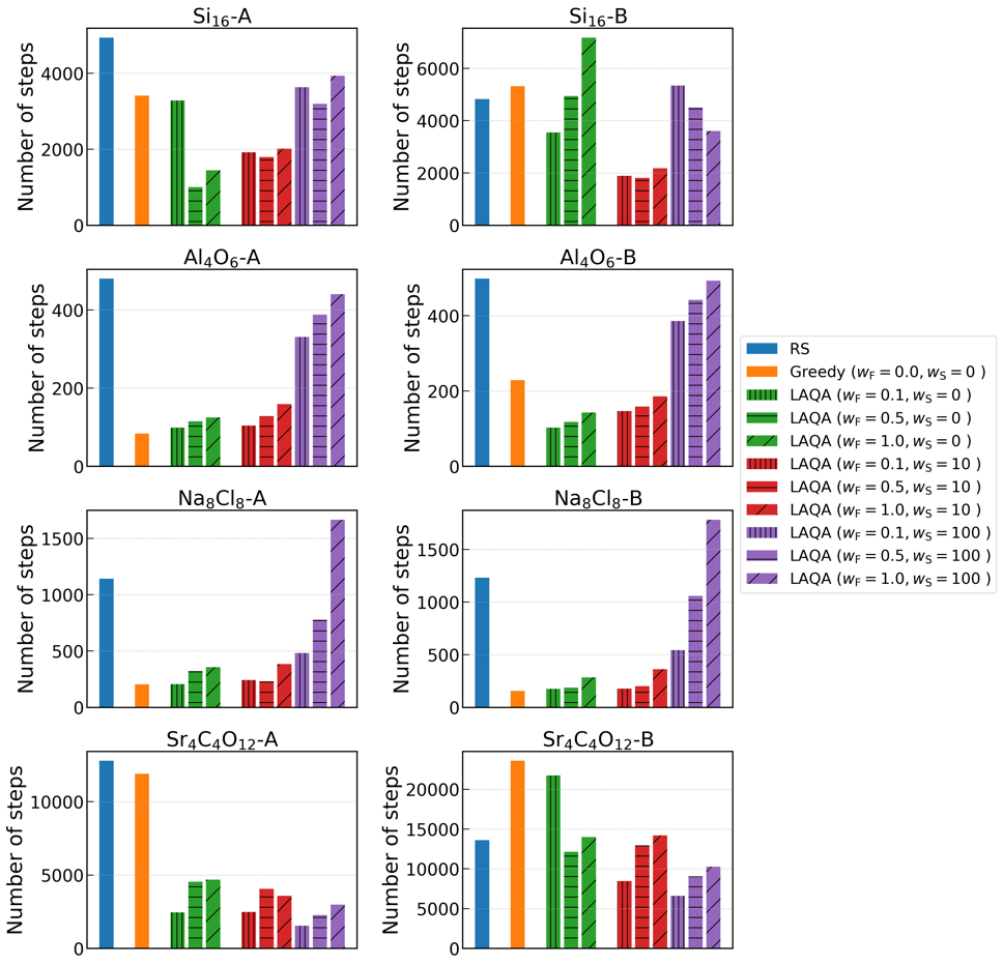

Fig. 1: Number of total optimization steps required to find the most stable structure.