Design of thermodynamically stable perovskites using machine learning

Zhilong Song [1], Xiwen Chen [1], Sae Dieb[2], Masashi Ishii[2]

Soochow university [1], MaDIS National Institute for Materials Science [2]

E-mail: DIEB.Sae@nims.go.jp

Perovskite materials have attracted much attention in the past few years and have been widely used in solar cells, light emitting diodes, lasers, photocatalysis, etc. [1] Especially, the photoelectric conversion efficiency of perovskite solar cells increased from 3.8%[2] in 2009 to 25.2%[3] in 2019. However, the long-term stability of perovskite materials is a big challenge for its large-scale commercialization.

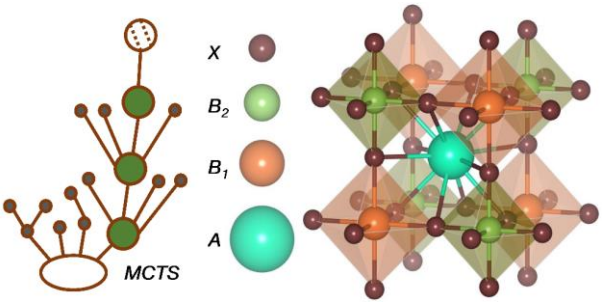

Figure 1 Double perovskite structure

We utilized a machine learning based optimization method named Monte Carlo tree search (MCTS)[4] and MCTS in combination with reinforcement learning (policy gradient) to search for the double perovskite material with best thermodynamic stability. MCTS has shown competitive efficiency in materials design problems[5]. As shown in Figure 1, the double perovskite structure has A, B_1, B_2 and X site which occupied by different elements. Different elements configuration of four sites formed 420 kinds of double perovskite structures and the decomposition energy of them were calculated in advance through first-principle calculation performed by VASP software. The SCF energy convergence and ionic convergence value were set to 10^{-5} eV and the calculation for each structure takes 20-30min.

The X axis of Figure 2 means the number of double perovskite structures that had been searched while the Y axis refers to the best decomposition energy obtained. The result in this figure is the average result of 20 runs. MCTS (blue line) found the structure with higher decomposition energy and had a better efficiency compared to Random Search(green line). The structure with highest decomposition energy ($Cs_2AgTlBr_6$) was obtained by MCTS almost every time after searching less than 50 structures.

A modified version of MCTS that integrated a reinforcement learning technique: policy gradient (yellow line) was also tested. Generally, this method can let the tree choose the nodes to expand wisely with the help of policy gradient and it had successfully improved the search result and efficiency in other optimization tasks. However, it didn't outperform the initial MCTS because the policy gradient needs data to train and 50 data obtained in this task was not enough for it.

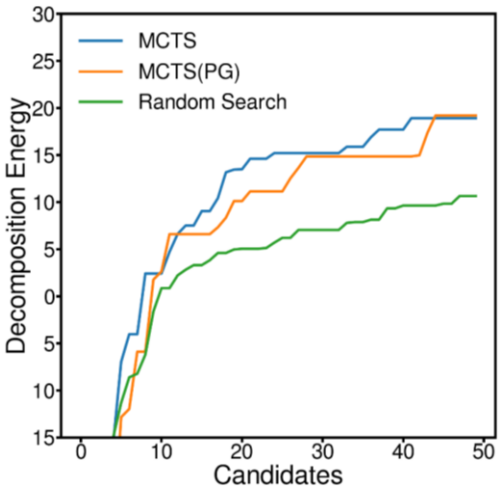

Figure 2 Comparison of MCTS and Random Search

In summary, the MCTS efficiently found the most thermodynamically stable double perovskite structure in the pre-calculated data using only elements type information. Next, we will perform the MCTS and first-principle calculation simultaneously to explore more perovskite structures with high decomposition energy and MCTS(PG) may help improve the efficiency with more data.

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