Data-Driven Flux-Method Crystal Growth of Inorganic Materials Using Bayesian Predictive Simulation

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Crystallographic characteristics such as crystallinity, crystal outline, and size are related to physicochemical phenomena occurring on the surface and inside of materials, and their control directly affects material performance. The flux method is a powerful technique for developing high-performance crystalline materials, but its growth guidelines are not well established. Therefore, it is necessary to find the optimum conditions in a huge experimental space based on various experimental factors. Thus, it takes several years to develop crystals. Recently, we have addressed flux process informatics (FPI), which is a data-driven approach to the flux method. There are many issues to be solved to achieve high-accuracy FPI system. In this study, we constructed an adaptive design of experiments (ADOE) system that can be used for FPI.

The ADOE system is a Bayesian optimization cycle consisting of (I) acquisition of experimental data, (II) modeling by Gaussian process regression, (III) conduction of virtual experiments over 10000 ways, and (IV) proposal of experimental conditions based on the acquisition function. In this time, a layered perovskite oxide $Ba_5Nb_4O_{15}$ (BNO) with an anisotropic crystal structure was selected as one of the model materials. As the explanatory variables, experimental conditions, including raw material amounts, flux species, and heating conditions were used. Two types of crystal sizes were used as the objective variables to describe anisotropic crystal shape of BNO. The number of training data was about 70.

Fig. 1 shows the simulation method of ADOE. The all dataset was divided into 10 as a training data and others as a test data. A model was created using the training data, and the test data with high-likelihood one was selected as the goal candidate. If the selected test data was different from the goal, the data was added to the training data and continued the exploration until finding the goal. The cycles of modeling were output per 1 trial and scored the efficiency of ADOE after 100 trials. Comparing 100 random experiments, the average number of cycles by ADOE was 6 times smaller, indicating the good efficiency of this system. In this presentation, we will also discuss the experimental results after applying the ADOE system, and contributing mechanism of each factor on the crystal growth.

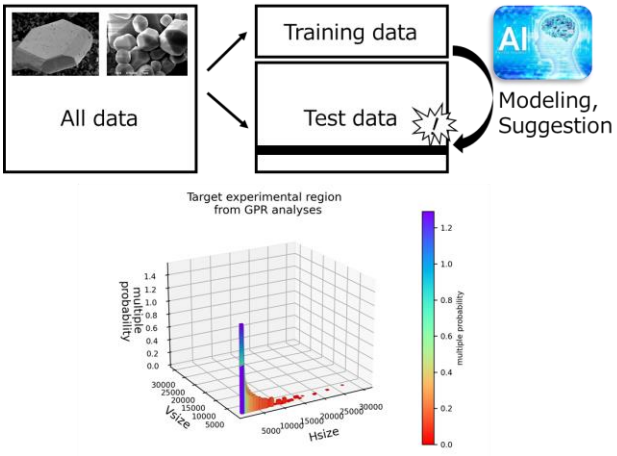

Fig. 1 Prediction simulation flow using ADOE.

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