Advancements in Material and Process Design through Machine Learning: Application to Epoxy-Based Adhesive Materials

Authors: *Keisuke Ando*, Yoshiyuki Kamo**

**Advanced Technology R&D Center*

Abstract

During the development of materials technologies supporting higher performance and reliability of electrical devices and reduction of environmental impact, design parameters such as the chemical composition of raw materials and manufacturing processes need to be adjusted. Yet changes in design parameters can dramatically alter the microstructure and state of materials, so developing the material with the desired structure and state requires tremendous time through trial and error.

Accordingly, we developed a method that first estimates promising design parameter regions via machine learning using known experimental data, and then selects as the next experimental point the condition with high predictive uncertainty and sparse experimental data, thereby efficiently clarifying promising design parameter regions. By applying this method to the development of epoxy-based adhesive materials, we also reduced the number of experiments by about 80% compared with conventional practices. This technique is anticipated to be applicable not only for the development of new materials, but also for various uses such as improving existing materials and manufacturing processes.

1. Introduction

Materials technology is a cross-disciplinary foundational technology that has the potential to lead to solutions for a wide range of industrial and societal challenges[(1)]. In recent years, there have been active efforts into research and development of material technologies incorporating Materials Informatics (MI) across industry, government and academia, around the world[(1)(2)]. At Mitsubishi Electric, we manufacture electrical device products that deliver value in every environment, from home appliances to space. Achieving greater added value for these devices, such as improved performance, enhanced reliability, and reduced environmental impact, requires the development of new materials. After generating candidate materials, design parameters such as the chemical composition of raw materials and manufacturing processes need to be adjusted in materials development so that they can be put to practical use in devices; however, even slight differences in conditions can lead to dramatic changes in a material's microstructure and state. Therefore, if promising design parameter regions that yield the desired material can be identified from the outset, efficient and innovative development is anticipated.

Here, we examined a method leveraging MI that actively and efficiently clarifies promising design parameter regions from known experimental results, taking an epoxy monolith sheet used for novel epoxy-based adhesive materials as a specific example. In an epoxy monolith sheet, the microstructure changes depending on the weight ratio of three raw materials, and there are also weight ratios of raw materials at which an epoxy monolith sheet cannot be obtained. Formation of the epoxy monolith sheet is also strongly influenced by the reactivity of the raw materials, so it also depends on differences in the chemical structure of the raw materials. Therefore, identifying design parameter regions such as the raw material weight ratios at which an epoxy monolith sheet can be obtained has in the past required experimental trial and error and a tremendous amount of time.

In this paper, we developed an MI method that first estimates promising design parameter regions using machine learning with known experimental data, and then selects as the next experimental point as the next experimental point the condition with high predictive uncertainty and sparse experimental data, thereby efficiently clarifying the promising design parameter regions—we then outline a case study applying the method to an epoxy monolith sheet.

2. Method for Estimating Promising Design Parameter Regions

For objectives like this one, under the premise that conditions such as the weight ratio of multiple raw materials are variable, one approach of clarifying parameter regions that yield the desired material is to use active learning to estimate phase diagrams that show thermodynamic equilibrium according to the temperature and composition of the material. Dai et al. showed that by using Bayesian optimization based on posterior prediction with a Gaussian process, the number of sampling points required in estimating a phase diagram with two phases can be greatly reduced(3). Terayama et al. showed that uncertainty sampling, which focuses exploration on uncertain regions near phase boundaries, makes efficient sampling possible even for phase diagrams with multiple phases(4). The phase diagram estimation addressed by these prior studies handle the thermodynamic equilibrium, so thermodynamic calculation can often be leveraged, and consequently a relatively large number of samplings can be tolerated. On the other hand, for materials like the epoxy monolith that are characterized by changes in microstructure depending on the manufacturing process, experimental verification is required, so there are severe constraints on the number of samplings. Therefore, to efficiently estimate promising design parameter regions, for uncertainty sampling, we introduced a procedure that preferentially proposes conditions that are far from the training data that have been tested to date.

2.1 Estimation framework

Figure 1 shows the framework for estimating promising design parameter regions. Here, we illustrate the case where the ratios of three raw materials A, B, and C are selected as the design parameters. The ternary diagram in Fig. 1 represents the raw material weight ratio; the closer to the vertex for raw material A, the greater the amount of raw material A.

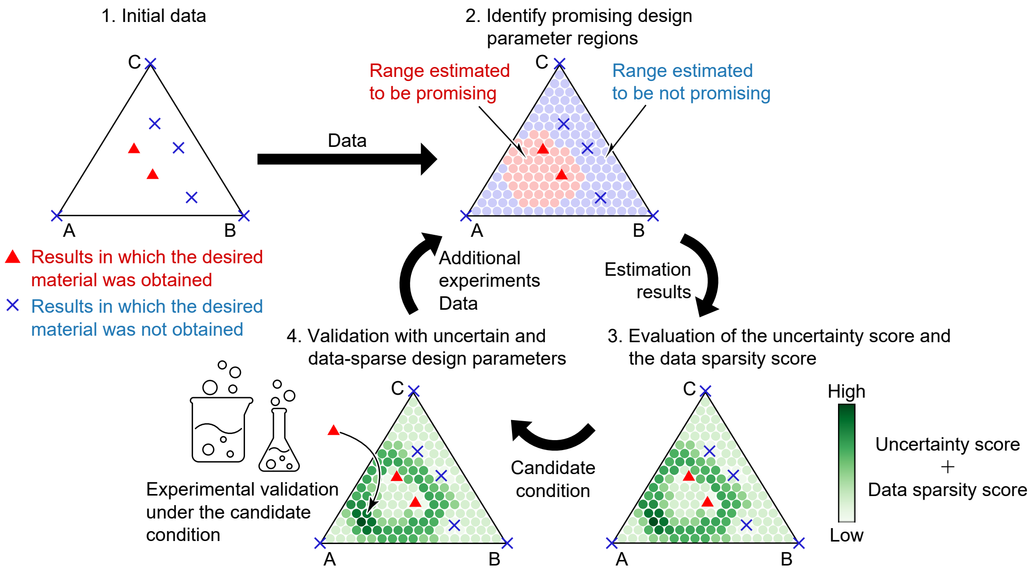

Fig. 1 Framework for identifying the promising design parameter regions

First, we prepare two types of outcomes as the initial training data: cases in which the desired material was obtained and cases with undesirable outcomes. Here, given that it is obvious that weight ratios that lack the required raw materials yield undesirable results, we added to the training data that undesirable results are obtained at the vertices of the ternary diagram.

Next, by performing machine learning using this training data, we built a classification prediction model to determine whether the design parameters are promising, and estimated promising design parameter regions. Based on these estimation results, we evaluated the uncertainty score and, as a penalty term, calculated the proximity to the training data, thereby evaluating the data sparsity score. The uncertainty score is a metric that evaluates the uncertainty in the prediction model's classification of whether the design parameters are promising, and given that the boundary between the region estimated as promising and the region estimated as undesirable is calculated to be the most uncertain, the entire perimeter of the boundary is evaluated as equally uncertain. Therefore, designing an objective function that adds the data sparsity score to the uncertainty score makes it possible to uniquely propose candidate conditions that are both uncertain and have not yet been experimentally tested. Using the candidate conditions proposed by this method, we carried out experimental validation and checked whether the desired material could be obtained. By adding these results to the training data and repeating the cycle of estimation, evaluation and verification, we also clarified the promising design parameter regions.

2.2 Preliminary verification of the estimation framework

To verify the operation of this method, we defined a virtual ground truth and tested whether the promising design parameter regions could be estimated. The interior of the ellipse shown with the black line in the figure is the region assumed as the ground truth, and Fig. 2 shows that the red region on the left denotes the region estimated as promising. It can be seen that in the first cycle, the estimated region does not coincide with the ground truth. Figure 2 on the right shows the distribution of the sum of the uncertainty score and the data sparsity score calculated from the estimation results. At the boundary between the region estimated as promising and the region estimated as undesirable, the sum of the uncertainty score and the data sparsity score is rated highly, and the asterisks are proposed as candidate conditions to be verified next. When a candidate condition falls within the region assumed as the ground truth, we added virtual data indicating that the desired material was obtained; when it falls outside, we added virtual data indicating that the desired material was not obtained.

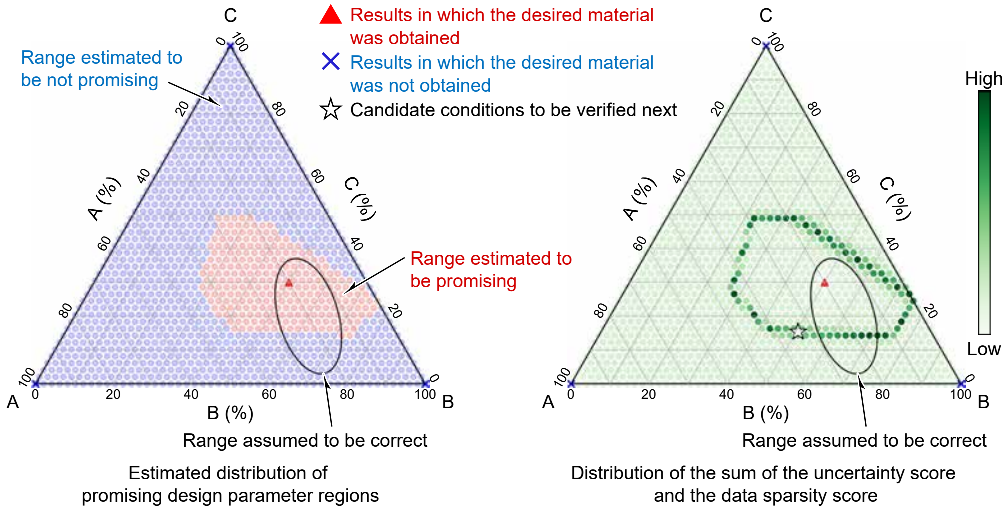

Fig. 2 Results of the estimation for the virtual data in the first cycle

The estimation results after repeating the estimation framework for 13 cycles are shown in Fig. 3. With the addition of virtual data 13 times, the region estimated as promising showed results very close to the ground truth, confirming the potential to derive promising design parameter regions with few experiments.

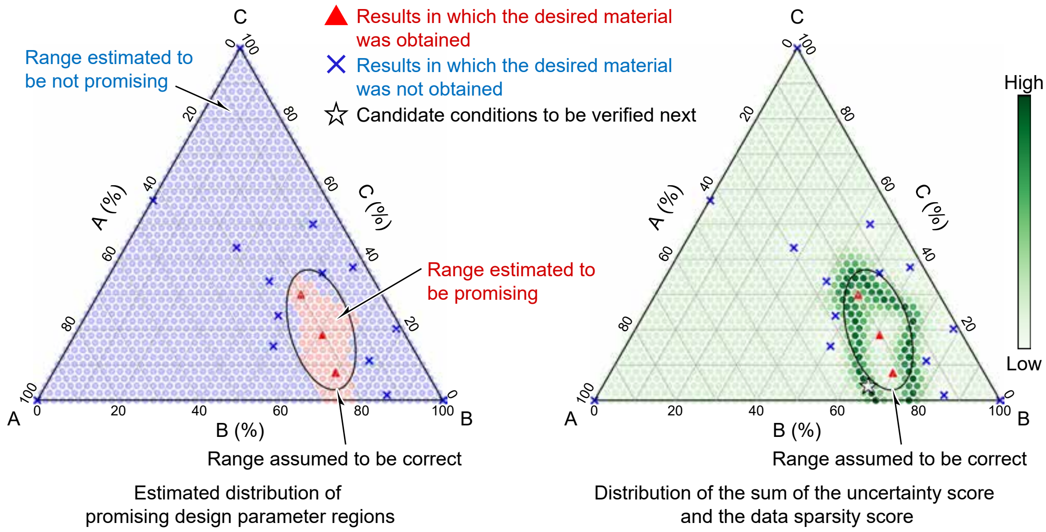

Fig. 3 Estimation results for the virtual data after 13 cycles

3. Application to the Development of New Epoxy-Based Adhesive Materials

In this chapter, we outline a case where this method was applied to an epoxy monolith sheet used in new epoxy-based adhesive materials.

3.1 Epoxy monolith sheet used in new epoxy-based adhesive materials

Epoxy-based adhesive materials, widely used in electric motors, semiconductor devices, and aerospace devices, are materials with high strength and excellent heat resistance, but they also have the drawback of being resistant to plastic deformation and brittle. In electrical devices, components repeatedly expand and contract due to temperature cycles during operation and shutdown, so greater flexibility of epoxy-based adhesive materials is required. We developed a sheet adhesive material in which an epoxy monolith sheet with internal continuous pore is impregnated with adhesive components, and have shown that by making the flexible epoxy monolith sheet function as a stress-relief layer at the bonded joint, it contributes to improving the heat cycle resistance of epoxy-based adhesive materials[(5)], and we are pursuing research and development toward further performance enhancement.

The epoxy monolith sheet is a thin film characterized by a co-continuous pore structure in which a mesh-like epoxy framework and voids are each three-dimensionally interconnected (Fig. 4)[(6)]. To achieve an epoxy monolith sheet, at least three raw materials need to be mixed: an epoxy resin, a curing agent and a porogenic solvent, and the microstructure varies depending on differences in the raw material weight ratio. If the raw-material weight ratio is not within an appropriate range, the epoxy framework becomes discontinuous and the sheet cannot maintain a self-supporting integrity, or the voids become discontinuous and cannot be impregnated with adhesive components, and it fails to function as an adhesive material. Therefore, clarification was needed of the raw material weight ratios that produce an epoxy monolith sheet, characterized by a mesh-like epoxy framework and voids that are each three-dimensionally interconnected.

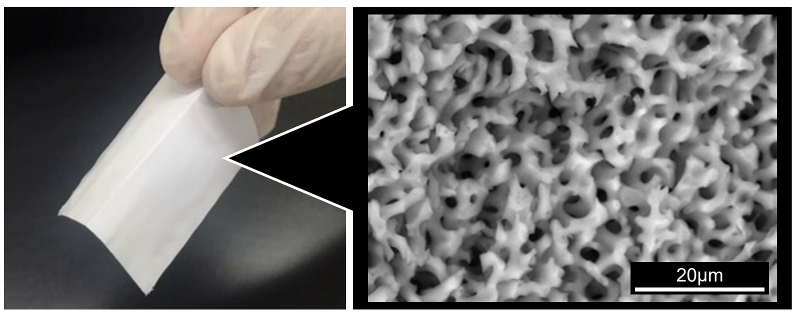

Fig. 4 Surface appearance and cross-sectional SEM image of the epoxy monolith sheets

3.2 Method for estimating promising design parameter regions to obtain an epoxy monolith sheet

In this paper, we focused on epoxy monoliths prepared using 4,4’-methylenebis (cyclohexylamine) (BACM) as the curing agent and poly (ethylene glycol) (PEG) as the porogenic solvent, with 2,2’-bis (4’-glycidyloxyphenyl) propane (BADGE) or 1,3-bis (N,N-diglycidylaminomethyl) cyclohexane (TETRAD-C) as the epoxy resin. For these two epoxy monolith systems, we set the weight ratios of the three raw materials as the design parameters, and estimated the promising design parameter regions for obtaining epoxy monolith sheets. First, we used as initial training data the result at a PEG concentration of 70 wt% where an epoxy monolith was formed in preliminary experiments, and the results for each raw material alone, where no monolith forms (the vertices of the ternary diagram). Based on the preliminary verification in section 2.2, while adding the results of additional experiments to the training data, we repeated the estimation framework for 13 cycles.

4. Estimated Results for Promising Design Parameter Regions where Epoxy Monolith Sheets are Obtained

Figure 5 shows the design parameter regions, estimated after 13 cycles, where epoxy monoliths are obtained. Focusing on the distribution of the experimental conditions, similar to the results of the preliminary verification in section 2.2, the experimental points that are not promising for monolith fabrication are distributed so as to surround the entire periphery of the ranges estimated to be promising for monolith fabrication, from which we consider that the promising design parameter regions have been estimated with high accuracy. In particular, given that the promising design parameter ranges differ for the monolith using BADGE as the epoxy resin and the monolith using TETRAD-C, we are able to estimate design parameter regions that reflect differences in the types of raw materials. By clarifying such promising design parameter regions, we became able to design materials after identifying the overall trends in epoxy monolith synthesis with a small number of experiments. Moreover, experiments through 13 cycles revealed that PEG concentrations of 40 wt% or lower did not yield epoxy monoliths. Because the number of experiments conducted at these concentrations was limited, even before it was definitively established that monoliths could not be obtained, we were able to focus on more promising experimental conditions. Whereas 66 additional experiments would traditionally have been required to test all conditions in 10 wt% increments, using this method we were able to identify the promising design parameter regions with 13 additional experiments. We reduced the number of experiments by about 80%, demonstrating that this method is effective for streamlining research and development (Fig. 6).

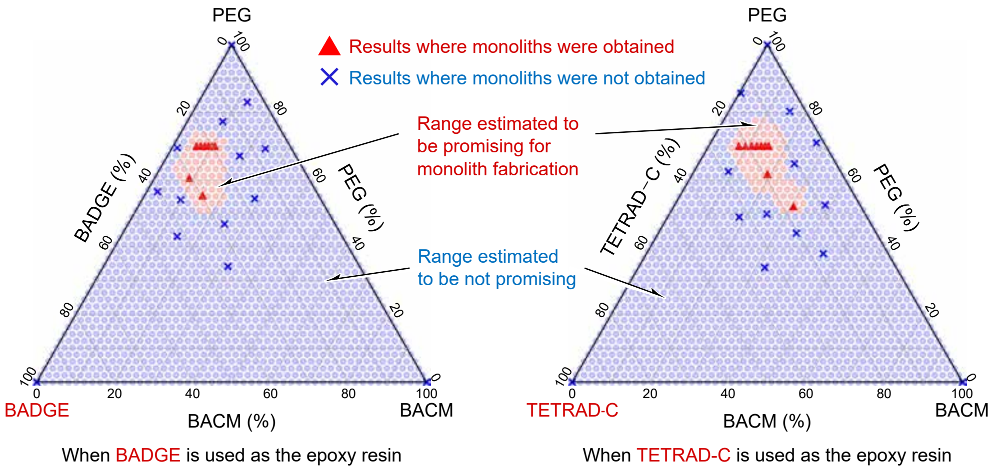

Fig. 5 Design parameter regions where the epoxy resin is obtained

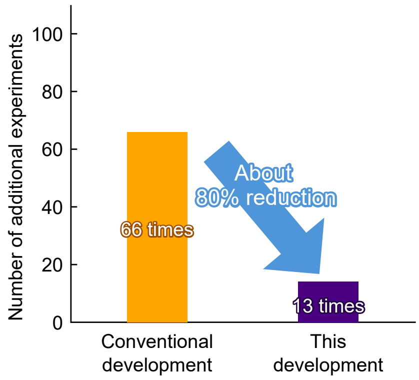

Fig. 6 Reduction in the number of experiments required to identify the promising design parameter regions

5. Conclusion

For efficient research and development of innovative materials, we developed a method that uses machine learning to efficiently select design parameters. Even in systems that include many design parameters, we identified a method to accurately estimate design parameter regions with a small number of experiments, and by applying this method to the development of new epoxy-based adhesive materials, we reduced the number of experiments by around 80% compared with conventional development. Given that this method can be extended to higher dimensions with three or more types of design parameters, we will proceed to apply it to even more complex and advanced materials development.

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