ABSTRACT: We aim to achieve resource recycling by capturing and using CO₂ generated in a chemical production and disposal process. We focused on CO₂ conversion to CO by the reverse water gas shift—chemical looping (RWGS-CL) reaction. This reaction proceeds in two steps (H₂ + MOₓ ⇌ H₂O + MOₓ−1; CO₂ + MOₓ−1 ⇌ CO + MOₓ) via a metal oxide that acts as an oxygen carrier. High CO₂ conversion can be achieved owing to a low H₂O concentration in the second step, which causes an unwanted back reaction (H₂ + CO₂ ⇌ CO + H₂O). However, the RWGS-CL process is difficult to control because of repeated thermochemical redox cycling, and the CO₂ and H₂ conversion extents vary depending on the metal oxide composition and experimental conditions. In this study, we developed metal oxides and simultaneously optimized experimental conditions to satisfy target CO₂ and H₂ conversion extents by using machine learning and Bayesian optimization. We used transfer learning to improve the prediction accuracy of the mathematical models by incorporating a data set and knowledge of oxygen vacancy formation energy. Furthermore, we analyzed the RWGS-CL reaction based on the prediction accuracy of each variable and the feature importance of the random forest regression model.

1. INTRODUCTION

A wide variety of chemicals are made from fossil fuels, such as crude oil, coal, and natural gas, through to petrochemical-based materials, such as ethylene and propene. Some of these chemicals are recycled after use, but most are incinerated and landfilled. A large quantity of carbon dioxide (CO₂) is released during the chemical manufacturing process and incineration of the chemicals. CO₂ is considered a cause of global warming.

One option to reduce CO₂ emissions is by carbon capture and storage (CCS), which is a process to separate and capture CO₂ generated during combustion and store it in a suitable place. CCS has the disadvantage that it requires a lot of energy to store CO₂. Carbon capture and usage (CCU) has therefore been attracting attention. CCU is a method of manufacturing petrochemical-based materials using captured CO₂ as a raw material. CCU reduces CO₂ emissions to the atmosphere and use of chemicals derived from fossil fuels; consequently, it is expected to achieve resource recycling. In addition, CO₂ conversion products can be sold as petrochemical-based materials, and thus, the CCU cost can be recovered. However, high thermodynamic stability is an obstacle to the use of CO₂, and therefore, hydrogen (H₂) is frequently used for CO₂ conversion because of its high energy content. CO₂ reacts with H₂ to form a wide variety of petrochemical-based materials, such as methane, methanol, carboxylic acids, and carbon monoxide (CO). In this work, we focused on the process of manufacturing CO, which can be used for production of chemicals, such as acetic acid, or as a synthesis gas with hydrogen to produce methanol or dimethyl ether.

The reverse water gas shift (RWGS) reaction produces CO from CO₂. CO₂ and H₂ react on a catalytic surface with CO and H₂O, as shown in the following reaction:

\[ \text{CO}_2 + \text{H}_2 \rightleftharpoons \text{CO} + \text{H}_2\text{O} \] (1)

Goguet et al. investigated the reactivity of the surface species present over a 2%Pt/CoO₂ catalyst during the RWGS reaction by a detailed operando spectrokinetic analysis. Chen et al. studied the reaction mechanism over a Cu catalyst by CO₂ hydrogenation. Wu et al. studied a Ni/SiO₂ catalyst and focused on the production of CO and CH₄. The RWGS reaction is characterized by a simple reaction mechanism and has been extensively studied; however, several disadvantages exist, such as the reaction efficiency being limited by...
equilibrium, the need for separation of the outlet gas, methanation, and the need for excess H₂. In this study, we focused on the reverse water gas shift—chemical looping (RWGS-CL) reaction. The RWGS reaction proceeds via the following two steps that are either spatially or temporally separated via a metal oxide (MOₓ), which acts as an oxygen carrier

\[ \text{H}_2 + \text{MO}_x \rightarrow \text{H}_2\text{O} + \text{MO}_{x-1} \]  

(2)

\[ \text{CO}_2 + \text{MO}_{x-1} \rightarrow \text{CO} + \text{MO}_x \]  

(3)

Hence, high CO₂ conversion can be achieved owing to low H₂O concentration in the second step, which causes an unwanted backward reaction in eq 1. Daza et al. investigated the effects of Cu and Sr doping on perovskites used as the metal oxide catalyst. Siriwardane et al. presented data on conversion for different coals with a chemical looping oxygen carrier, CuO–Fe₂O₃–alumina, over a range of conditions, including steam and various levels of reduction of the oxygen carrier. Here et al. combined a perovskite with a wide variety of supports, such as CeO₂, ZrO₂, Al₂O₃, SiO₂, and TiO₂, at industrial scale. Maiti et al. achieved 100% selectivity of CO generation at low temperature (450–500 °C) using La- and Ca-based perovskite oxides. Sun et al. proposed a one-pot method to synthesize dual-function materials that contained a sorbent coupled with a catalyst component. Ramos et al. discovered material properties that promote CO₂ conversion in a perovskite by doping with Co and Mn. Ma et al. presented several Co- and Mn-codoped ferrites for a midtemperature RWGS process. Kellar et al. searched for the best metal oxide from a process analysis based on thermodynamic data. Although the RWGS-CL reaction has been actively researched, it remains difficult to control because the reaction involves repeated thermochemical redox cycling, and the CO₂ and H₂ conversions vary greatly depending on the metal oxide composition and experimental conditions. Therefore, metal oxides with high CO₂ and H₂ conversions have not yet been developed. The objective of this study was to design metal oxides and simultaneously optimize experimental conditions to satisfy target CO₂ and H₂ conversion extents.

In conventional development of metal oxides, the next experimental candidate, including metal oxides and experimental conditions, is selected based on the experience and intuition of researchers. For example, if design variables for the RWGS-CL process are assumed to be 5 parameters, such as temperature and reactor size, with 10 candidates assumed for each parameter, and the metal oxides are assumed to have 15 candidates, the total number of combinations is 15 × 10⁵ (≈ 1.500 000), which requires a large number of experiments and is unrealistic. Therefore, we focused on machine learning to efficiently determine optimal metal oxides for this application. Mathematical models of Y = f(X) were constructed for CO₂ and H₂ conversions (Y) and descriptors for metal oxides and experimental conditions (X) using experimental results. Y-values were estimated by inputting X-values for metal oxides and experimental conditions into the models. By using models and estimating Y-values, it is possible to realistically handle 1 500 000 combinations of candidates. By selecting the next experimental candidates from estimation results, then an X that leads to target performances can be selected from a huge number of combinations of metal oxides and experimental conditions. In a typical mathematical model, the applicability domain (AD) is set, and candidates with desirable Y-values are selected within the AD. In the case of Gaussian process regression (GPR), the candidates considered include variability of the estimated Y-values, called Bayesian optimization. AD is useful for reliable prediction of X-values in interpolation regions, while Bayesian optimization is useful for exploring extrapolation in repeated experiments.

In addition to machine learning, we used transfer learning to improve the prediction accuracy of the mathematical models by incorporating a data set and knowledge of oxygen vacancy formation energy (OVFE).

2. METHOD

2.1. Data. The data comprised 520 samples that were investigated by Sekisui Chemical Co., Ltd. Test measurements were carried out at atmospheric pressure in a quartz tube microreactor, placed in an electric furnace. Typically, 200 mg of sample was packed between quartz wool plugs. First, the samples were heated to 650 °C under He. After this, the samples were reduced for 5 min in 100% H₂ flushed for 5 min under He, and then 100% CO₂ was flowed for 5 min, followed by another 5 min of He flushing. The total flow rate of the feed gas into the reactor was kept constant at 5.0 mL·min⁻¹. This process was repeated one time for a total of two cycles. There were 161 metal oxides. Experimental conditions included the metal, molar rates, precipitation method (sol–gel, coprecipitation, or solid phase), sintering temperature, heat-up time, temperature retention time, hydrogen excess, and reaction temperature. The outlet products were analyzed by a gas chromatograph–mass spectrometer (GC/MS). Consumption of H₂ and CO₂ is connected in that each molecule consumes and provides one oxygen atom. Hence, the molar amount of hydrogen consumption during reduction corresponds with the molar amount of carbon monoxide produced during CO₂ reoxidation. Additionally, the CO selectivity was almost 100% in this reaction. Therefore, H₂ conversion was calculated from the ratio of the amount of CO produced and the total amount of the inlet of H₂. For the purposes of this paper, the term of the CO₂ conversion was taken to mean the instantaneous conversion ratio of CO₂ gas to CO. Accordingly, the CO₂ conversion was calculated from the ratio of the amount of CO and CO₂ produced in 1 min after the starting of the reaction. For this reason, the conversion of CO₂ and H₂ was not the same. The CO selectivity was almost 100% in all experiments, which was because separating carbon and hydrogen prevents the generation of CH₄.

2.2. Descriptor Calculation. Data from the periodic table, Python Materials Genomics (pymatgen), and the Materials Project were used to represent structural information for the metal oxides. Periodic table data were collected from the Chemistry Handbook and paper and included atomic mass, electron count, peripheral electron count, atomic radius, ionic radius, ionization energy, electronegativity, work function, and surface energy. Pymatgen includes data for atomic mass, atomic orbitals, atomic radius, boiling point, density of solid, liquid range, melting point, molar volume, thermal conductivity, and electronegativity for metallic elements. The Materials Project data includes energy, energy per atom, volume, formation energy per atom, band gap, and density. Periodic table data and pymatgen descriptors were obtained for each atom, and weighted averages were determined using the molar rates; similarly, Materials Project
descriptors were obtained for each metal oxide, and weighted averages determined using the molar rates.

2.3. Regression Model. Partial least-squares (PLS),\textsuperscript{29} ridge regression (RR),\textsuperscript{30} least absolute shrinkage and selection operator (LASSO),\textsuperscript{31} elastic net (EN),\textsuperscript{32} linear support vector regression (LSVR),\textsuperscript{33} nonlinear support vector regression (NLSVR), linear Gaussian process regression (LGPR), random forest (RF),\textsuperscript{34} light gradient boosting machine (light-GBM),\textsuperscript{35} and GPR methods were used to build the mathematical models.

The prediction accuracy was evaluated by double cross-validation (DCV).\textsuperscript{36} The sample was first divided by the number of outer folds. One group was used for validation, and the remaining groups were used for cross-validation (CV) (inner folds) to optimize the hyperparameters and then predict the data for validation. This was done for all groups. The prediction accuracy of the mathematical model was evaluated by comparing the actual and predicted Y-values of the outer CV. For comparison, the coefficient of determination ($R^2_{DCV}$), mean absolute error (MAE$_{DCV}$), and root-mean-square error (RMSE$_{DCV}$) were used, as respectively given by

$$R^2_{DCV} = 1 - \frac{\sum_{i=1}^{n} (y^{(i)} - y^{(i)}_{DCV})^2}{\sum_{i=1}^{n} (y^{(i)} - y^{average})^2}$$

$$MAE_{DCV} = \frac{\sum_{i=1}^{n} |y^{(i)} - y^{(i)}_{average}|}{n}$$

$$RMSE_{DCV} = \sqrt{\frac{\sum_{i=1}^{n} (y^{(i)} - y^{(i)}_{DCV})^2}{n}}$$

where $y^{(i)}$ is the Y-value of the i-th sample, $y^{DCV}_{DCV}$ is the predicted Y-value of i-th sample by DCV, $y^{average}$ is the average of the Y-values, and $n$ is the number of samples.

2.4. Transfer Learning. Transfer learning is a method to improve the prediction accuracy and efficient learning of a mathematical model by transferring knowledge from other data. To improve the prediction accuracy of the mathematical model in this study, we used OVFE values, which are considered to be correlated with the extents of CO$_2$ and H$_2$ conversion. OVFE is the energy released when oxygen atoms are released from metal oxides and is typically calculated by density functional theory calculations. In this study, we used the OVFE values of 45 compounds reported by Deml et al.\textsuperscript{37}
In several transfer learning algorithms, we focused on the frustratingly easy domain adaptation, which can transfer knowledge by expanding data sets. The expanded data sets were generated in three parts: metal oxides, periodic table + pymatgen descriptors, and periodic table + Materials Project descriptors. In addition, the sintering temperature, shown in Figure 1(a), was varied. Therefore, metal oxides that have desirable Y-values can be estimated without the need for experiments. In the training data, was selected.

3. RESULTS AND DISCUSSION

3.1. Descriptor Calculations. The correlation coefficients between X and Y are shown in Figure 1 after calculating the

Figure 2. Measured and estimated values of CO2 and H2 conversion by double cross-validation. (a) Y = CO2 conversion, X = experimental conditions, periodic table, and pymatgen descriptors; (b) Y = H2 conversion, X = experimental conditions and pymatgen descriptors.

Table 2. Prediction Accuracy of Oxygen Vacancy Formation Energy (OVFE) Values for Each Variable

| model inputs                      | method | R2_{DCV} | MAE_{DCV} | RMSE_{DCV} |
|-----------------------------------|--------|----------|-----------|------------|
| periodic table                   | PLS    | 0.555    | 0.677     | 0.882      |
| pymatgen descriptors             | GPR    | 0.832    | 0.451     | 0.593      |
| Materials Project descriptors    | LSVR   | 0.840    | 0.398     | 0.530      |
| periodic table + pymatgen descriptors | GPR  | 0.886    | 0.335     | 0.446      |
| periodic table + Materials Project descriptors | PLS | 0.830    | 0.422     | 0.546      |
| pymatgen + Materials Project descriptors | GPR | 0.832    | 0.425     | 0.543      |
| all                              | PLS    | 0.852    | 0.393     | 0.510      |

3.2. Regression Analysis. Mathematical models were constructed between the X and Y values calculated in Section 3.1, and each was evaluated by DCV. The number of DCV inner folds was set to 5 and the number of outer folds was set to 161, which was the number of metal oxides. The mathematical models were validated by assuming that they would predict new metal oxides. The DCV-predicted results structural information for the metal oxides. There are no variables that highly correlated with Y. The maximum correlation coefficient was 0.37 for the atomic orbital energy and melting point, as shown in Figure 1(c). In addition, the data set has X variables that are highly correlated with each other, such as sintering temperature, heat-up time, and reaction temperature, shown in Figure 1(a).

3.3. Structural Analysis of Metal Oxides. After the estimated Y-values were obtained, the candidates for X were evaluated. If the mathematical model with the highest prediction accuracy was PLS, RR, LASSO, EN, LSVR, NLSVR, RF, or light-GBM, the AD was set, and the X-candidate that had desirable Y-values within the AD was selected. If the mathematical model with the highest prediction accuracy was LGPR or GPR, the X-candidate that had the highest probability of improvement (PI) value, i.e., the probability of exceeding the maximum Y-values in the training data, was selected.

Table 1. Prediction Accuracies of CO2 and H2 Conversions for Each Model Input

| model inputs                      | method | R2_{DCV} | MAE_{DCV} | RMSE_{DCV} |
|-----------------------------------|--------|----------|-----------|------------|
| periodic table                   | GPR    | 0.433    | 13.990    | 18.286     |
| pymatgen descriptors             | GPR    | 0.504    | 13.159    | 17.096     |
| Materials Project descriptors    | GPR    | 0.367    | 14.576    | 19.320     |
| periodic table + pymatgen descriptors | GPR | 0.441    | 13.701    | 18.158     |
| periodic table + Materials Project descriptors | GPR | 0.506    | 12.685    | 17.072     |
| pymatgen + Materials Project descriptors | GPR | 0.546    | 12.445    | 16.359     |
| all                              | RF     | 0.542    | 12.397    | 16.428     |

3.4. Evaluation of Metal Oxides. The candidates for Y were evaluated by expanding the original data set.
MAEDCV in eq 5, and RMSEDCV in eq 6 were calculated for each variable method, such as hydrogen excess, metal, and boiling point. The GPR model between the periodic table and pymatgen descriptors had the highest prediction accuracy, with $R^2_{DCV}$ of 0.886, because there are many variables with a relatively high correlation between OVFE and $X$. As atomic mass, solid density, melting point, and boiling point.

Figures 3 and 4 show the DCV-predicted OVFE. In Figure 3, most samples appear on the diagonal, and thus, the prediction accuracy is quite high. Figure 4 shows no relationship between CO$_2$ conversion and predicted OVFE values, but metal oxides with predicted OVFE values of 3 to 4 eV tended to have high H$_2$ conversion.

We then evaluated the prediction accuracies by DCV for each transfer learning method. The results are shown in Table 3. For both CO$_2$ and H$_2$ conversions, the prediction accuracy was found to be higher using transfer learning methods than using linear regression models. The GPR model was the most accurate, with $R^2_{DCV}$ of 0.886, because there are many variables with a relatively high correlation between OVFE and $X$.

### Table 3. Prediction Accuracy of CO$_2$ and H$_2$ Conversions for Transfer Learning Methods

| variable | CO$_2$ conversion | H$_2$ conversion |
|----------|-------------------|------------------|
|          | $R^2_{DCV}$  | MAE$_{DCV}$  | RMSE$_{DCV}$ | $R^2_{DCV}$  | MAE$_{DCV}$  | RMSE$_{DCV}$ |
| Section 3.2 | GPR 0.546 | 12.445 | 16.359 | GPR 0.733 | 6.683 | 10.233 |
| Method 1   | GPR 0.550 | 12.428 | 16.290 | GPR 0.737 | 6.712 | 10.160 |
| Method 2   | GPR 0.522 | 12.827 | 16.791 | GPR 0.723 | 6.798 | 10.434 |
| Method 3   | GPR 0.566 | 12.167 | 16.000 | GPR 0.730 | 6.771 | 10.296 |
was improved by using transfer learning Methods 1, 2, or 3. For CO$_2$ conversion, Method 3 had the highest prediction accuracy. Method 2 was not able to distinguish between the zero matrix and zero with no values; by excepting the zero matrix for autoscaling, Method 3 was able to make this distinction and therefore showed improved prediction accuracy. For H$_2$ conversion, Method 1 had the highest prediction accuracy, because it is considered to reflect a direct relationship between OVFE and H$_2$ conversion.

Figure 6. Random forest feature importance using transfer learning. (a) $Y = \text{CO}_2$ conversion; transfer learning Method 3; (b) $Y = \text{H}_2$ conversion; transfer learning Method 1.

Figure 7. Comparison between experimental (blue) and predicted (gray) Y-values.

Figure 8. Probability of improvement (PI) results. Red: Pareto optimal solution (53 samples); gray: all candidates (65,232,600 samples).

Figure 5 shows the CO$_2$ and H$_2$ conversions with highest DCV-predicted accuracy. Figure 6 shows the feature importance of RF. Comparing Figure 5 with Figure 2, by utilizing knowledge of OVFE, the prediction accuracy is slightly improved in the region where the value of $Y$ is large. In CO$_2$ conversion, the feature importance of $X_B$ in eq 7 is high. In Method 2, the feature of $X_B$ is not high, and thus, the data set with high feature importance of $X_B$ is considered to be
Table 4. Samples of Representative Pareto Optimal Solutions

| metal oxide | log PI [-] | $P_{\text{CO}_2}$ [-] | $P_{\text{H}_2}$ [-] | ratio of metal 1 [-] | ratio of metal 2 [-] | ratio of metal 3 [-] | precipitation method | hydrogen excess |
|-------------|------------|-----------------|-----------------|-----------------|-----------------|-----------------|------------------|-----------------|
| Mg-Cu-Ga    | −12.85     | 0.30            | $8.70 \times 10^{-4}$ | 0.16            | 0.41            | 0.43            | coprecipitation   | excess          |
| Mg-Cu-Ga    | −12.20     | 0.29            | $1.78 \times 10^{-3}$ | 0.18            | 0.48            | 0.34            | coprecipitation   | excess          |
| K-Cu-Ga     | −13.80     | 0.31            | $3.24 \times 10^{-4}$ | 0.17            | 0.49            | 0.34            | coprecipitation   | excess          |
| K-Cu-Ga     | −13.93     | 0.32            | $2.84 \times 10^{-4}$ | 0.19            | 0.5             | 0.31            | coprecipitation   | excess          |
| K-Cu-Ga     | −2.86      | 0.22            | 0.26             | 0.18            | 0.46            | 0.36            | coprecipitation   | less            |
| K-Cu-Ga     | −2.99      | 0.18            | 0.28             | 0.2             | 0.44            | 0.36            | coprecipitation   | less            |

Table 5. Samples with the Six Highest Log PI Values

| metal oxides | log PI [-] | $P_{\text{CO}_2}$ [-] | $P_{\text{H}_2}$ [-] | ratio of metal 1 [-] | ratio of metal 2 [-] | ratio of metal 3 [-] | precipitation method | hydrogen excess |
|--------------|------------|-----------------|-----------------|-----------------|-----------------|-----------------|------------------|-----------------|
| K-Cu-Ga      | −2.86      | 0.22            | 0.26             | 0.18            | 0.46            | 0.36            | coprecipitation   | less            |
| Mg-Cu-Ga     | −3.22      | 0.22            | 0.18             | 0.17            | 0.41            | 0.42            | coprecipitation   | less            |
| Ca-Cu-Ga     | −4.02      | 0.18            | 0.10             | 0.17            | 0.38            | 0.45            | coprecipitation   | less            |
| Cu-Ga-Sr     | −4.48      | 0.17            | 0.07             | 0.42            | 0.42            | 0.16            | coprecipitation   | less            |
| Mn-Cu-Ga     | −6.69      | 0.11            | 0.01             | 0.004           | 0.39            | 0.53            | coprecipitation   | less            |

Figure 9. Histogram of logarithm of probability of improvement (log PI).

suitable for transfer learning. In $H_2$ conversion, the feature importance of the predicted OVFE is high, and this variable may have increased the prediction accuracy.

3.4. Inverse Analysis. We created 65 232 600 candidate metal oxides as described in Section 2.5. The created candidates were input to the regression model with the highest prediction accuracy and the corresponding $Y$-values predicted. For $CO_2$ conversion, the GPR model was constructed between $X$, with experimental conditions, pymatgen descriptors, and Materials Project descriptors, and $Y$ using Method 3. For $H_2$ conversion, the GPR model used was constructed between $X$, with experimental conditions and pymatgen descriptors and $Y$ using Method 1. Figure 7 shows the predicted and experimental $Y$-values. We were able to find a candidate with predicted $Y$-values that exceeded the trade-off relationship in the experimental $Y$-values.

Both the $CO_2$ and $H_2$ conversion models used GPR, from which we calculated the PI using the predicted $Y$-values and their variances. Figure 8 shows the PI values. The 53 red samples are the Pareto optimal solution: six representative samples are shown in Table 4. The Pareto optimal solution is divided into two groups: one with high $CO_2$ conversion and the other with high $H_2$ conversion. Table 4 shows that these values are affected by the hydrogen excess value: when the hydrogen flow is insufficient, the extent of reaction between $CO_2$ and $MO_{x-1}$ in eq 3 is small because of low reaction between $H_2$ and $MO_{x-2}$ in eq 2; when the hydrogen flow is excessive, the reaction between $CO_2$ and $MO_{x-1}$ in eq 3 proceeds well, but the $H_2$ conversion decreases, and thus, PI values for $H_2$ conversion will be low. In Table 5, Cu and Ga, which are known to facilitate high $H_2$ conversion, were selected, and their molar composition was approximately 0.4. From the high RF feature importance shown in Figure 8, the melting point of Cu is larger than that of other metals and the atomic orbitals of both metals are relatively low, accounting for the high $CO_2$ and $H_2$ conversions.

PI values are probabilities, and therefore, we calculated the probability (log PI) that all $Y$ values exceed the existing $Y$-values by taking the logarithm for each $Y$ and adding these together. Figure 9 shows the histogram of log PI. Table 5 shows the five metal oxides with highest log PI values. As in Table 4, Cu and Ga were selected; the remaining metals tended to be alkali and alkaline-earth metals, which have a low number of electrons in the outermost electron shell. The state of electrons on the surface of the metal oxide is thought to be important for thermochemical redox cycling to occur. The candidates selected in Tables 4 and 5 were not in the training data. Experiments with the selected candidates are expected to develop metal oxides that exceed the $Y$-values in the training data set.

4. CONCLUSION

In this study, we constructed mathematical models using machine learning to predict $Y$-values, where $Y$ is $CO_2$ and $H_2$ conversions. Based on combinations of variables with the highest prediction accuracy and RF (random forest) feature importance, the prediction accuracy for $CO_2$ conversion was improved by adding electronic information and structural information for the metal oxides; $H_2$ conversion seems to depend on experimental conditions, such as hydrogen excess, metal, and metal composition. Furthermore, we focused on OVFE (oxygen vacancy formation energy) values, which are considered to be correlated with the $Y$-values, and conducted transfer learning. For $CO_2$ conversion, the prediction accuracy was improved by considering the relationship with $X$; for $H_2$ conversion, which is directly correlated with the predicted OVFE values, the prediction accuracy was improved by using the OVFE values as $X$. After constructing the mathematical models, we input 65 232 600 candidates as $X$-values that have not been experienced, and the corresponding $Y$-values were estimated without experiments. From the predicted $Y$-values, we were able to find candidates with predicted $Y$-values that exceed the trade-off relationship in the experimental $Y$-values. By selecting candidates for the next experiments from the
estimated results based on Bayesian optimization and updating the mathematical models with the experimental results, the candidate X metal oxides that will achieve the target Y-values are expected to be found with a small number of experiments. Although there are lots of experimental results published in the literature regarding CO₂ conversion using metal oxides, even when the experimental results are similar in terms of CO₂ conversion and metal oxides, the experimental results cannot be compared, the prediction accuracy of models cannot be compared, and the samples cannot be merged if experimental systems and experimental conditions are different. The use of samples from different experimental systems with techniques such as transfer learning would be a challenge for the future.

### AUTHOR INFORMATION

**Corresponding Author**

Hiromasa Kaneko — Department of Applied Chemistry, School of Science and Technology, Meiji University, Kawasaki-shi, Kanagawa-ken 214-8571, Japan; orcid.org/0000-0001-8367-6476; Phone: +81-44-934-7197; Email: hkaneko@meiji.ac.jp

**Authors**

Ryo Iwama — Department of Applied Chemistry, School of Science and Technology, Meiji University, Kawasaki-shi, Kanagawa-ken 214-8571, Japan; orcid.org/0000-0002-7718-4544

Koji Takizawa — Sekisui Chemical Co., Ltd., Osaka-shi, Osaka-fu 530-8565, Japan

Kenichi Shimmei — Sekisui Chemical Co., Ltd., Osaka-shi, Osaka-fu 530-8565, Japan

Eisuke Baba — Sekisui Chemical Co., Ltd., Osaka-shi, Osaka-fu 530-8565, Japan

Noritoshi Yagihashi — Sekisui Chemical Co., Ltd., Osaka-shi, Osaka-fu 530-8565, Japan

Complete contact information is available at: https://pubs.acs.org/10.1021/acsomega.2c00461

**Notes**

The authors declare no competing financial interest. Data and Software Availability: Research data are not shared since experimental data are considered proprietary by the organization and the algorithms of the proposed method are described in the manuscript.

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