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Hydride Content Control of Perovskite Oxyhydride $\text{BaTiO}_{3-x}\text{H}_x$ Supported by Image-Based Machine Learning

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ExtraTrees

ABSTRACT

Hydride content in perovskite oxyhydrides represents a crucial parameter that governs properties of the materials. However, the synthesis of the oxyhydrides with precise control over the hydride content x remains a challenging task because of the time-consuming and/or destructive analytical methods typically used for the determination of x . Here, we report an image-based ML system for prediction of the hydride contents x in a perovskite oxyhydride $\text{BaTiO}_{3-x}\text{H}_x$ using a picture of powder material as a quick evaluation method of hydride content x . The ML system, which employs the *ExtraTrees* algorithm, enabled the prediction of x with a mean absolute error of 0.013 and a low running cost. Although a similar ML system constructed by convolutional neural networks (CNN) that we used in the previous study demonstrated comparable accuracy in the prediction, the system with *ExtraTrees* was more computationally efficient, thus was applied as a non-destructive and quick analytical method for hydride content x . Using the ML system, detailed profiles of thermal hydrogen release of $\text{BaTiO}_{3-x}\text{H}_x$ were quantitatively analyzed to demonstrate the feasibility of fine-tuning the hydride content within $0 \leq x \leq 0.4$ by adjusting the processing temperature. In the case of powder samples comprising mixtures of the oxyhydride with different hydride contents, the ML-based prediction provided an almost averaged value for x . The present results demonstrate an application of image-based ML for the fine-tuning of oxyhydride materials.

INTRODUCTION

Inorganic oxides with multiple anions, such as oxyhalides, and oxynitrides, have attracted considerable attention owing to their superior functionalities, which may not be absent from conventional oxides.¹ Perovskite oxyhydrides, in which the O^{2-} anions are partially replaced by H^- ions, have various properties and demonstrated applications, including catalysis of ammonia

synthesis,^{2–4} CO₂ methanation,⁵ and hydrogenation of organic compounds.^{6–8} Oxyhydrides have also been used as precursors for the synthesis of other mixed-anion perovskites,^{9–11} playing a pivotal role in this research field. The hydride content of oxyhydrides is a crucial factor that governs their properties. However, existing studies are focused on investigating the activation energies of H₂ release from oxyhydrides rather than fine-tuning their hydride content.¹²

Controlling the hydride content x of oxyhydrides remains a challenge. Optimizing the reaction conditions to achieve a desired x requires numerous cycles of synthesis and analysis, and thus current methods are still time-consuming. The standard procedure for synthesizing BaTiO_{3– x} H _{x} involves heating BaTiO₃ and CaH₂ at 500–580 °C for 4–7 days in a sealed tube,¹³ making it difficult to monitor the reaction course by sampling small portions of the reaction mixture. Furthermore, quantitative analytical methods for determining the hydrogen content are limited. While direct refinement of the hydride content via Rietveld analysis of neutron diffraction patterns is powerful,¹⁴ it requires considerable time or a high-flux neutron source for accurate analysis. Thermal desorption spectrometry (TDS) can be used to quantitatively analyze the hydride content of oxyhydrides by determining the amount of hydrogen gas desorbed upon heating.^{15,16} Because analytical samples for TDS are decomposed during the measurement, this method is unsuitable for non-destructive or on-site analysis of the hydride content. X-ray diffraction (XRD) and thermogravimetry (TG) are more convenient alternatives; however, their precision may not be adequate owing to their indirect nature.

Recently, machine learning (ML) has been used for predicting various properties of perovskite materials, and expected to accelerate the discovery of new materials.¹⁷ The formation energy of perovskite materials was predicted by means of ML through the connection of atoms in the crystal.¹⁸ In the field of materials science, image-based ML has also been used for predicting

material properties.¹⁹ For instance, the infrared (IR) spectrum images of proteins were predicted rapidly by ML using structural descriptors.²⁰ We have recently developed a ML method that uses images to predict the mixing ratios of solid chemicals.²¹ Once a suitable ML model is built using a training data set containing 50–300 images with experimentally determined mixture ratios, the mixing ratios of organic/inorganic compounds, crystal polymorphs, and enantiomers can be rapidly predicted from an image, with mean absolute error (MAE) of approximately 5%. This system is also applicable to the non-destructive and consecutive analysis of solid-state chemical reactions. Given these advantages, we envision that rapid, non-destructive analysis based on ML will lead to reliable conditions for preparing oxyhydride samples with controlled hydride contents. Here, we report a facile method based on image-based ML for the quantitative analysis of the hydride content of perovskite oxyhydride, $\text{BaTiO}_{3-x}\text{H}_x$. Analysis of thermal hydrogen release under various conditions indicated that the final hydrogen content, x , can be finely tuned in the range of $0 \leq x \leq 0.4$ by selecting the appropriate annealing temperature. The ML system was also applicable in the prediction of averaged hydride contents when powder materials were composed of the oxyhydrides with different hydride contents.

RESULT and DISCUSSION

To construct an ML model for analyzing the hydride content of $\text{BaTiO}_{3-x}\text{H}_x$, we first collected a training data set (Figure 1). Powder material with $x = 0.53$ was prepared by heating BaTiO_3 (particle size approximately 100 nm) and CaH_2 at 560 °C for 2 days (see ESI for the detailed procedure).¹³ Authentic samples with various x values were synthesized by heating the initial

material ($x = 0.53$) under random conditions (300–500 °C for durations of 15 min to 10 h). Then, the experimental hydride content x of each sample was analyzed using TDS (see ESI for details). Total 230 images (approximately 1 mg each; Figure 1) were captured from 23 authentic powder samples using a microscope camera. It should be noted that the stir-to-picture cycle¹⁰ allows the quick acquisition of 10 different images (within a few minutes) from each authentic sample. Among these, 207 images were used for model making, while the others were used for testing. The ML model was initially constructed by using convolutional neural network (CNN) as implemented in PyTorch.²² While our previous ML system using CNN was designed for the prediction of mixing ratio r with a range of $0 \leq r \leq 1$,²¹ the ML model for the present study was constructed to output hydride content x in a range of $0 \leq x$ without setting the upper limit. As the color of the oxyhydride changed in accordance with hydride x , an alternative ML model was also prepared for comparison of the running cost. A screening process based on several pixel-intensity-based features was employed to evaluate the accuracy of the ML model (see Figures S2–S19). After the screening, it was found that a ML model constructed by the ExtraTrees²³ regression using the histogram of pixel intensity values (the brightness levels at individual pixels) as explanatory variables can be a computational cost-effective alternative. Additional details regarding ML model making are provided in the Supporting Information.

The accuracy of the ML predictions was evaluated using an observation–prediction plot, which was generated using 23 test images that were excluded from the training data set (Figure 2). The ML model constructed using CNN provided predictions with an MAE, root mean squared error (RMSE), and coefficient of determination R^2 of 0.0089, 0.011, and 0.99, respectively. The prediction accuracy was significantly better than that of previously reported solid mixture predictions; for example, prediction of the sugar/dietary salt mixing ratio using a model built with

300 training images was characterized by an MAE, RMSE, and R^2 of 0.039, 0.046, and 0.97, respectively.¹⁰ Although the process of extracting feature variables from image data using CNN differs from visual recognition by human eyes, the remarkable increase in the prediction accuracy can be attributed to the apparent color change upon hydride release.

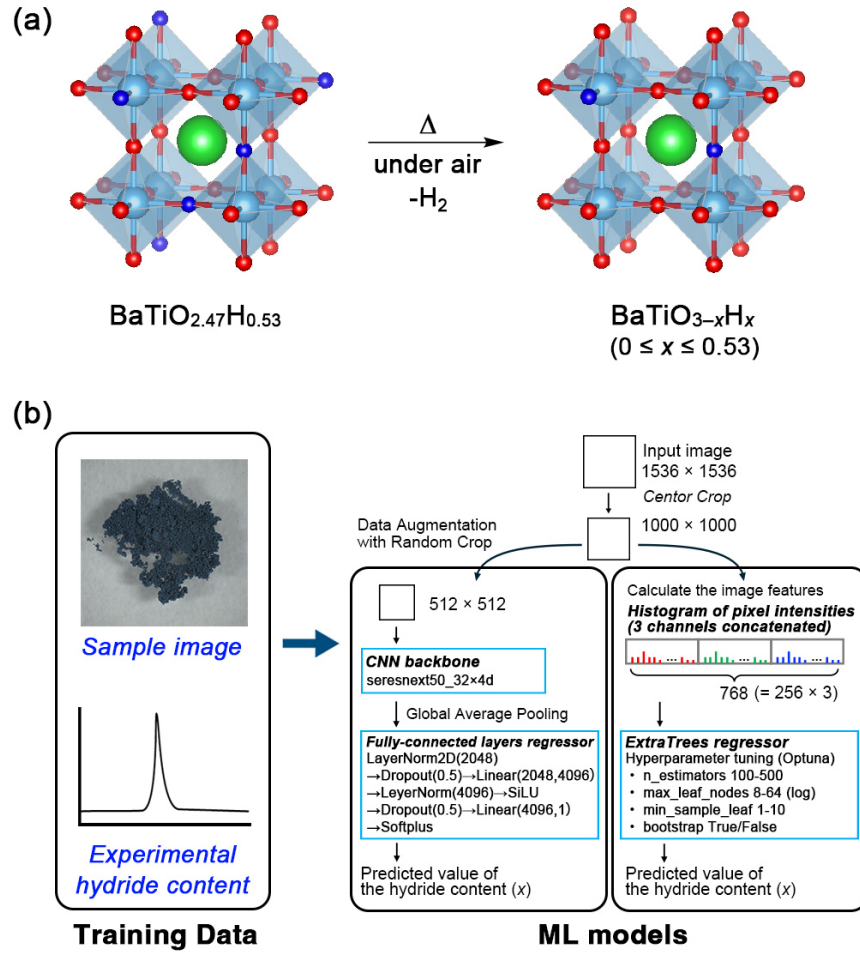


Figure 1. (a) Schematic representation of thermal hydrogen release from perovskite oxyhydride $\text{BaTiO}_{2.47}\text{H}_{0.53}$. The structures were drawn using VESTA.²⁴ Green, gray, red, and blue spheres represent Ba, Ti, O, and H, respectively. (b) Workflow of ML model making for the prediction of hydride content x .

Although the prediction accuracy of the ML model using CNN was acceptable for quantitative analysis of hydride content x , an alternative ML model constructed using *EtraTrees* exhibited better computational efficiency while maintain a comparable level of accuracy. The *ExtraTrees* regression model with feature representation C (the histograms of pixel intensity value) predicted x with MAE, RMSE and R^2 of 0.013, 0.017 and 0.99, respectively, which represents the optimal accuracy. In this ML model, the original images, which had a resolution of 1536×1536 pixels, were center-cropped to a size of 1000×1000 pixels and the intensity values were normalized. Given the favorable cost-to-accuracy ratio, we employed the ML model using the *ExtraTrees* regression in the following studies.

Using the ML-based hydride-content prediction system, we analyzed the profiles of hydrogen release from $\text{BaTiO}_{3-x}\text{H}_x$ ($x = 0.53$ for the initial sample) at various temperatures. The oxyhydride sample was heated in an electronic furnace under aerobic conditions for up to 5 h. Small aliquots of the samples (approximately 1 mg) were photographed at certain intervals during heating. Changes in the hydride content versus heating time were plotted for each reaction temperature.

When the initial sample ($x = 0.53$) was heated to 500 °C, the hydride content x decreased quickly, resulting in the formation of a faded colored sample with $x = 0.031$ after 1 h (Figure 3). Over a period of 5 h, the value of x gradually approached nearly 0, indicating release of most of the hydrogen content. A steep decrease in the hydride content was also observed during the initial 1 h of heating at 400 °C, although x remained at 0.18 even after 5 h of heating at 400 °C. Heating the sample at 300 °C for 5 h resulted in a smaller decrease in the hydride content, reducing x to approximately 0.4.

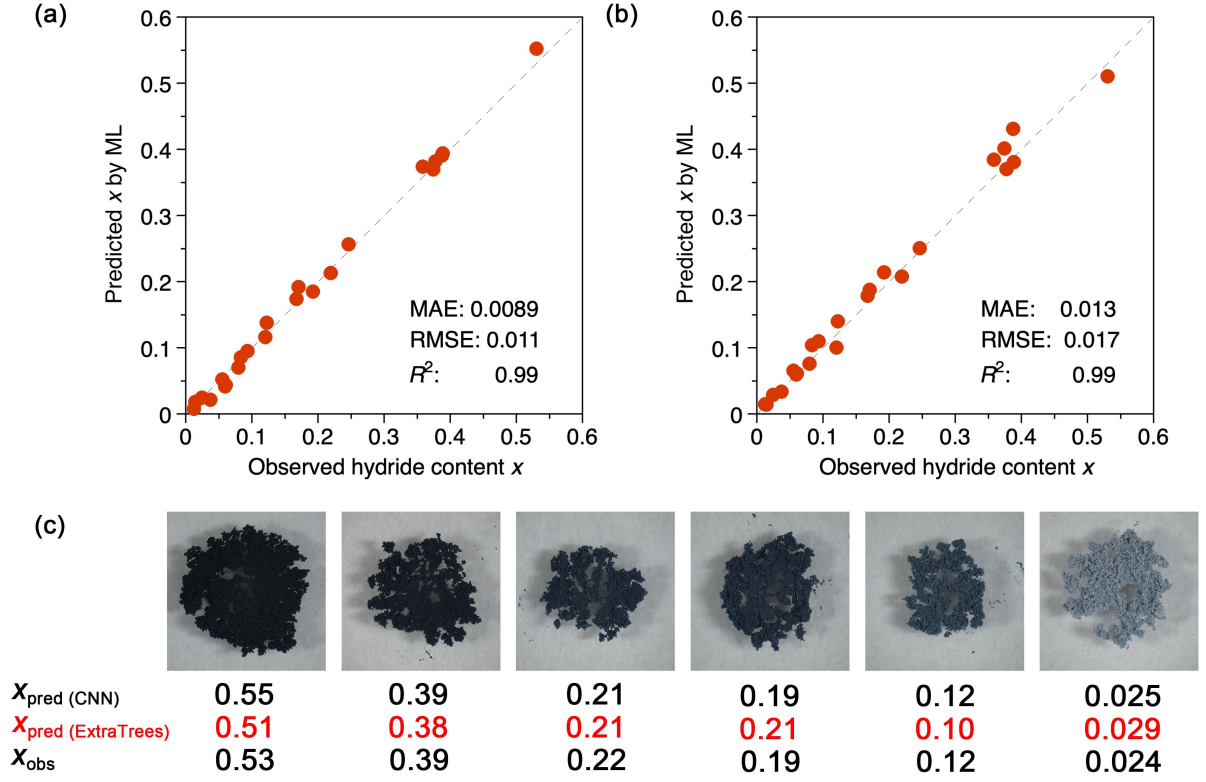


Figure 2. Observation–prediction plots for the prediction of hydrogen content x of $\text{BaTiO}_{3-x}\text{H}_x$ (a) using CNN and (b) using ExtraTrees. (c) Selected examples of image-based ML prediction results ($x_{\text{pred (CNN)}}$ or $x_{\text{pred (ExtraTrees)}}$ are the predicted value from image-based ML using CNN or ExtraTrees, respectively, and x_{obs} is the experimental value determined using TDS).

When the initial sample was heated at temperatures with smaller increments (300, 325, 350, 375, 400, 425, 450, 475, and 500 °C) for 5 h, the final hydride content x changed to 0.38, 0.36, 0.33, 0.25, 0.18, 0.12, 0.059, 0.022, and 0.013, respectively (Figure 3b). Although the hydrogen release profiles between 300 and 325 °C were similar, x decreased by approximately 0.05 when the annealing temperature was increased to 350 °C. In the range of 350–450 °C, the final hydride content x changed proportionally with the annealing temperature, with a slope of $2.7 \times 10^{-3} \text{ K}^{-1}$. However, there was only a slight difference in the profiles obtained at 475 and 500 °C, which were similar to those observed at 300–325 °C.

The validity of the ML-based prediction for the analysis shown in Figure 3a was also confirmed through TDS analysis. For samples prepared at (i) 300 °C for 4 h, (ii) 400 °C for 3 h, and (iii) 450 °C for 2 h, hydride content x determined experimentally using TDS was 0.37, 0.19, and 0.083, respectively. The corresponding ML-prediction values ($x = 0.40, 0.21, \text{ and } 0.11$ for (i), (ii), and (iii), respectively) were as accurate as those obtained with the test images shown in Figure 2a.

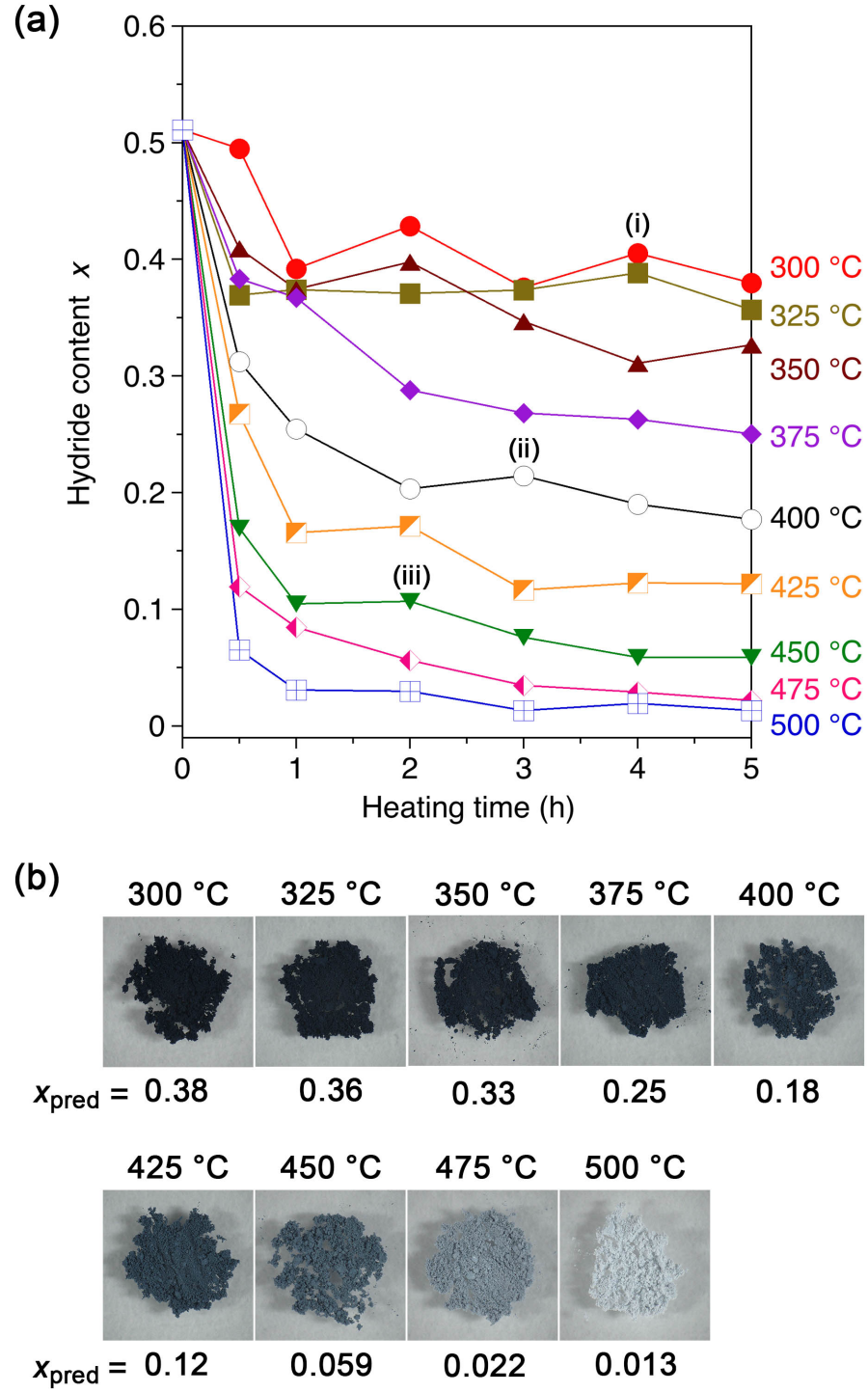


Figure 3. (a) Changes in the hydride content of oxyhydride $\text{BaTiO}_{3-x}\text{H}_x$ (initial $x = 0.53$) prepared at various temperature analyzed using image-based ML. (b) Photographs of samples after 5 h of heating at 300–500 °C with ML-predicted x_{pred} values.

Next, we applied complex temperature programs to control the hydride content x . The initial $\text{BaTiO}_{3-x}\text{H}_x$ sample was first heated at 300 °C for 3 h. Then, the temperature was increased to 400 °C for another 3 h, followed by additional heating at 500 °C for 3 h (Figure 4a). The hydride content x changed from $x = 0.49$ (initial) to 0.46 (300 °C), 0.32 (400 °C), and 0.022 (500 °C) in an almost stepwise manner. When the sample pre-heated at 350 °C for 3 h ($x = 0.39$) was treated at 300 °C for 3 h, the hydride content x remained unchanged (Figure 4b). However, when the temperature was raised to 400 °C, x decreased to 0.29 and remained almost constant throughout the additional stage of heating at 300 °C for 3 h. After the final stage of heating at 450 °C for 3 h, the final value of x was 0.12, which was comparable with that of the sample obtained by heating the initial sample at 450 °C for 3 h (Figure 4b and 3a).

The annealed samples maintained their hydride content for over 1 d at room temperature. When the sample with $x = 0.44$, prepared by heating at 300 °C for 3 h, was left at room temperature for 1 d, the hydride content remained unchanged (Figure 4c). Nonetheless, treating the sample at 400 °C led to a gradual decrease in x . After heating for 5 h, x reached a constant value of approximately 0.2. Although the detailed mechanism of the temperature-dependent decrease in the hydride content of $\text{BaTiO}_{3-x}\text{H}_x$ remains unclear, the ML-based analysis has provided a more detailed temperature vs hydrogen release relationship than previous studies.¹³ As we observed that the final hydride content in the temperature range of 350–450 °C changed linearly, oxyhydrides $\text{BaTiO}_{3-x}\text{H}_x$ with $0 \leq x \leq 0.4$ can be prepared in a predictable manner. More importantly, the non-destructive analysis using image-based ML facilitates quick monitoring during the reaction without the loss of sample quantity, allowing the fine tuning of x .

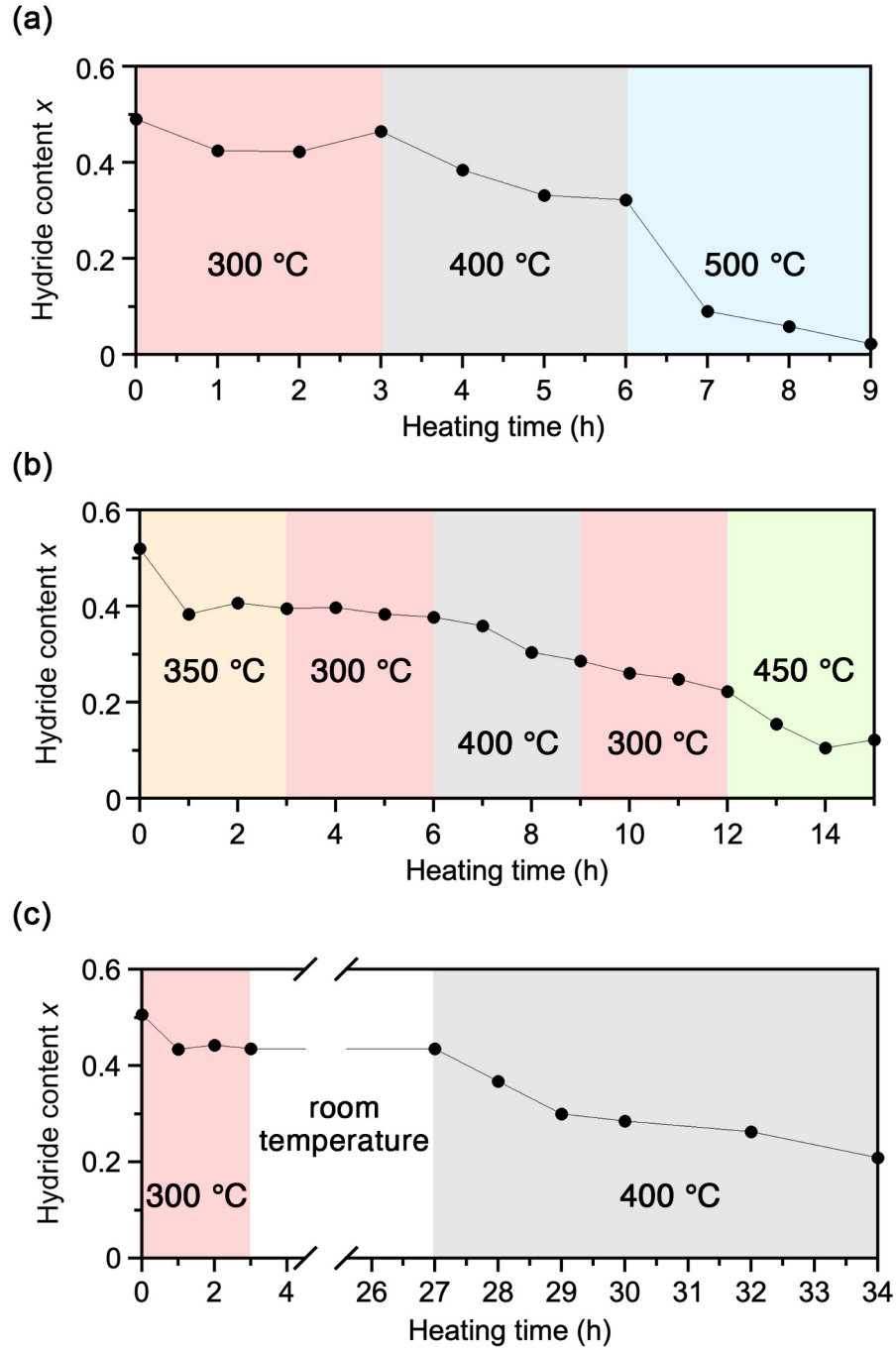


Figure 4. Changes in hydride content x of oxyhydride $\text{BaTiO}_{3-x}\text{H}_x$ (initial $x = 0.53$) prepared with a series of temperature programs, including: fluctuations in the heating temperature, ranging (a) from 300 °C to 400 °C, and 500 °C; (b) from 350 °C to 300 °C, 400 °C, 300 °C, and finally 450 °C; (c) from 300 °C to room temperature and 400 °C.

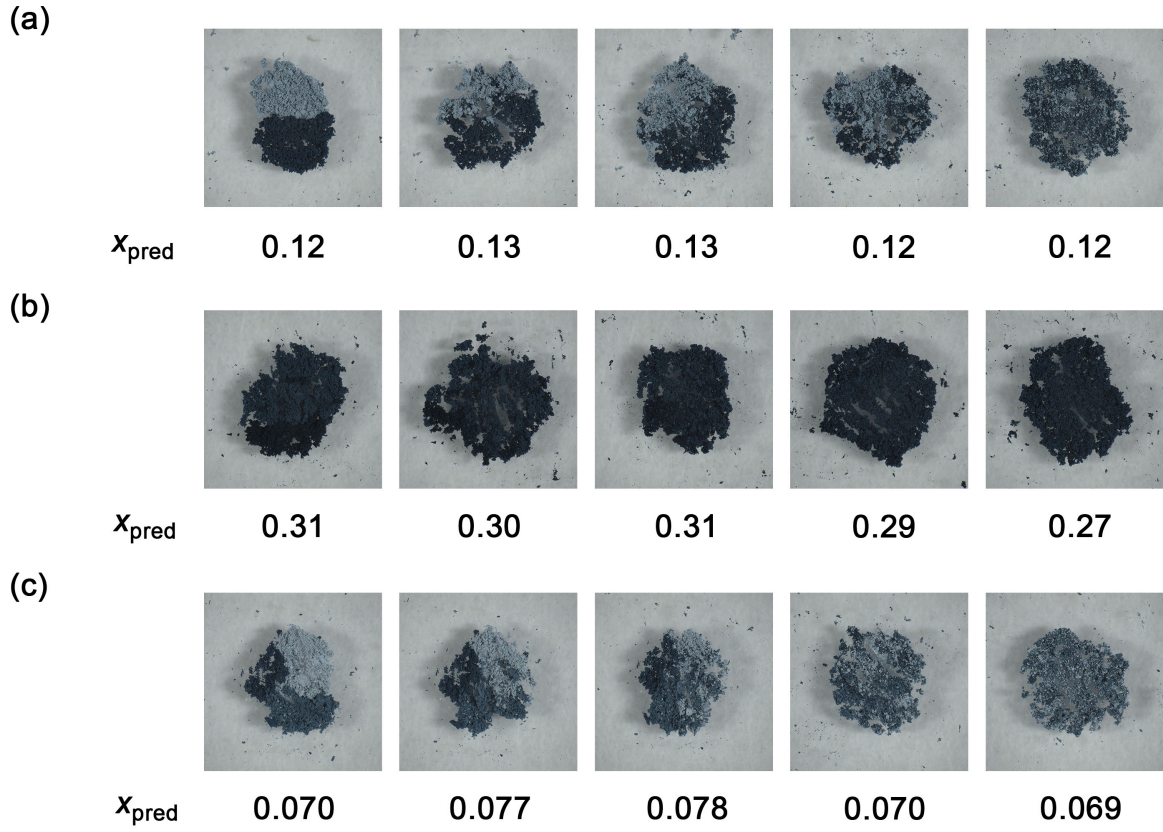


Figure 5. Hydride content prediction using (a) a 1:1 (wt/wt) mixture of oxyhydrides with $x = 0.035$ and 0.29 at various mixing states (left: before mixing with a spatula; right: well mixed). Analogous experiments using oxyhydride mixtures of (b) $x = 0.26$ and 0.51 in a 7:3 (wt/wt) ratio, (c) $x = 0.022$, 0.12, and 0.25 in a 5:3:2 ratio, respectively.

Oxyhydride synthesis often results in the formation of heterogeneous samples comprising domains with different hydride contents x . To expand the scope of ML-based hydride content analysis, we conducted the prediction experiments using mixtures of $\text{BaTiO}_{3-x}\text{H}_x$ powders having different hydride contents x (Figure 5). Two authentic samples with $x = 0.035$ and 0.29 were placed on a glass plate in a 1:1 ratio (wt/wt), and a picture was taken without mixing (Figure 5a, left). The ML-prediction output exhibited a hydride content of $x = 0.12$, which is somewhat proximate to the theoretically averaged x of 0.16 . Upon stirring the solid samples with a spatula, several pictures were taken at various stages of the mixing process. The ML-prediction of x for these images exhibited almost constant values independent of the stirring conditions. In the case of a 7:3 (wt/wt) mixture of $x = 0.26$ and 0.51 , respectively, the predicted x values exhibited slight fluctuations in the range of $0.27 \leq x \leq 0.31$. Despite the tendency to underestimation, the ML-prediction again demonstrated values roughly consistent with the theoretical average value ($x = 0.34$). The analysis of a three-component system, composed of oxyhydrides with $x = 0.022$, 0.12 , and 0.25 in a weight ratio of 5:3:2, revealed that the predicted x ranged from 0.069 to 0.078 . These values are also in agreement with the theoretical value ($x = 0.097$). It is also noteworthy that the ML model constructed using CNN exhibited slightly higher accuracy of prediction than the *ExtraTrees* model (Figure S21), despite the high computational cost. Although the margin of error for the ML-prediction of mixtures is relatively larger than that for homogeneous samples, it was demonstrated that the ML model can be used to estimate the averaged hydride content in heterogeneous or well-mixed oxyhydride mixtures, despite the lack of images of such mixtures in the training dataset.

CONCLUSION

In summary, we developed an image-based ML system to predict the hydride content of oxyhydride $\text{BaTiO}_{3-x}\text{H}_x$ from photographs of powder samples. The sufficiently low MAE of the ML system allowed it to be used as a non-destructive, quantitative analytical method that is more efficient and faster than conventional methods such as powder X-ray diffraction and TDS. Sequential analysis during the thermal release of hydrogen revealed the characteristic temperature dependence of the hydride content, which can be used to prepare oxyhydrides with different hydride contents in the range of $0 \leq x \leq 0.4$ in a predictable manner. In terms of computational cost efficiency, the ML model using the *ExtraTrees* regression was found to be more efficient than that using CNN. The ML prediction system was also demonstrated to be applicable for the estimation of averaged hydride contents in heterogeneous oxyhydride samples. Although the ML model developed in this research was designed for predicting the hydride contents of $\text{BaTiO}_{3-x}\text{H}_x$ only, a similar system could be used to analyze various mixed-anion compounds. Despite the necessity for a substantial training data set (>200 pictures with >20 experimental values), the accuracy of the prediction is attractive as it can be used as an analytical method that does not require any expensive spectrometer. It is also worth noting that evaluating multiple machine learning models is important for selecting an appropriate model for individual compounds. Given these advantages, ML-based analysis of materials will become more popular and then support the discovery of new structures, reactions, and properties of solid compounds.

ASSOCIATED CONTENT

The Supporting Information is available free of charge on the ACS Publications Web site.

Synthetic procedures, TDS spectra, Details of machine learning, compound images for machine learning (PDF).

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