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High-Throughput Screening and Literature Data Driven Machine Learning Assisting Discovery of La₂O₃-based Catalysts for Low-Temperature Oxidative Coupling of Methane

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Abstract

Three-element component La₂O₃-based catalysts for the oxidative coupling of methane (OCM) are examined for this study using machine learning (ML) approaches such as support vector regression (SVR) and random forest regression (RFR). This validation was conducted while assuming that the three-element component (M1–M2–M3) resulting in high C₂ yield predicted by ML is helpful to ascertain the appropriate component to promote the unique nature of La₂O₃ itself: the low-temperature OCM feature. The combined use of an open-source high-throughput screening (HTS) database and SVR assisted the discovery of three-element component La₂O₃-based OCM catalysts of 11 types with C₂ yield (> 5.0%) appearing at 450°C. Then, to predict more unique component La₂O₃-based OCM catalysts from the outer field of screening elements at HTS experimentation, HTS and literature databases were applied for SVR and RFR. This combined approach discovered 11 additional combinations of three-element component La₂O₃-based OCM catalysts affording C₂ yield (> 5.0%) appearing at 450°C.

1. Introduction

Because of the increasing availability of natural gas resources such as shale gas and methane hydrate, catalytic transformation of methane has attracted strong attention in industrial and academic fields. The oxidative coupling of methane (OCM) discovered in the 1980s is one attractive process for producing C₂ hydrocarbons of ethylene (C₂H₄) and ethane (C₂H₆) directly from methane (CH₄) [1-2]. However, using conventional processes of catalyst discovery based on scientists' intuition and experience, the desired performance criteria of a C₂ yield higher than 30% and superior C₂ selectivity of 80% with an appropriate C₂ yield sufficient to generate economically rational industrial plant systems have persisted as great challenges for the past 40 years [3-6].

Along with the recent intense growth of data science management and machine learning (ML) engineering, a common interesting query has arisen: “How can such new technologies aid catalyst discovery more effectively in comparison with conventional method?”. *Catalyst Informatics* has been adopted to develop next-generation understanding as a trend of data used for catalyst discovery [7-10]. Indeed, some successful discoveries of homogeneous catalysts have been achieved [11-12]. Such discoveries rely on the nature of homogeneous catalysis, which is fundamentally attributable to the design of an active metal center and a coordinating ligand. When applying a heterogeneous (solid) catalyst, greatly challenging issues arise for application because heterogeneous catalysis depends on numerous catalyst features such as composition, morphology (size, shape, crystallinity, etc), oxidation state, defect, strain, support nature, additive element, and stability under active conditions. In fact, ascertaining the relations among physicochemical features as determined by their reactivity and by conventional characterizations is extremely complicated. Moreover, even if some clear relation is identified between those characteristic features and reactivity, it remains controversial whether the relation can serve in a common role as a global descriptor. In other words, the model of representing heterogeneous catalysis feature using ML algorithm networking is an extremely important and persistent subject [13].

Since 2017, the author has explored how to apply the ML engineering to produce benefit information for catalyst scientists under close collaboration with ML engineers and data scientists, as presented later in Acknowledgements. During initial stages of research, results demonstrated that a well-defined random forest regression (RFR) based on a large dataset from the literature including 1,868 catalyst data can propose 56 undiscovered catalyst components and corresponding reaction conditions for OCM to achieve C₂ yield higher than 30% [14]. It is particularly interesting that the RFR also revealed the hidden key importance of OCM catalysis among the 11 descriptors of the preparation method, reaction temperature, CH₄ pressure, O₂ pressure, contact time, cation1, cation1 mol%, cation2, cation2 mol%, catalyst support, and catalyst support mol%. Those findings indicated ML approaches as effective tools for discovering unreported catalysts and for elucidating hidden effects such as catalyst components and reaction conditions on catalyst performance. However, at the next research stage in validation, a difficulty arose in achieving target OCM catalyst performance with a high C₂ yield over 30% among the predicted catalyst components [15]. One more impressive experience was conducted on tracing the catalyst behaviors under different reaction conditions by interpolate filling using ML engineering with a small experimental dataset. At this time, the author examined the OCM reaction over a well-known Na-Mn-W/SiO₂ catalyst for 156 data points collected one-by-one using a conventional fixed-bed reactor for experimentation. Then, ML was implemented to treat experimental conditions in high dimensions described with five reaction

parameters: reaction temperature, CH₄/O₂ ratio, CH₄ and O₂ gas concentrations (CH₄ + O₂ conc.), total flow rate, and catalyst weight. At this time, extra tree regression (ETR) was found to represent the trend of experimental data accurately [16]. This representation predicted that the target of C₂ yield features over Na-Mn-W/SiO₂ can help to indicate the best reaction conditions to afford the best performance. Therefore, ML was demonstrated as a powerful approach for ascertaining and then elucidating the best experimental conditions in multiple dimensions. Other earlier studies of reaction network representation [17-18] and hidden rule identification by item-set mining [19] have also provided attractive findings for improvement of scientists' understanding of some catalysis features. According to these shared experiences with collaborators, the author has found that ML and data management engineering skills hold great potential to reveal hidden trends of data in both large and small datasets, but exact prediction of the outer area performance is apparently still challenging. It is worth describing that the former is apparently difficult by human senses, but the latter has been achieved by conventional catalyst scientists without such a complete understanding, in catalyst development and modification history. Therefore, arranging the ML-aided catalyst discovery with more appropriate methods for generating unexpected components possessing high performance at target reactions is truly desired.

Along with the intense growth of data analysis and predictions that have been made using ML and data management engineering tools, it has become necessary to collect huge amounts of tailor-made datasets with high efficiency. Several parallel reactor systems have been developed in a fixed bed catalytic reactor [20-23]. The combined use of tailor-made datasets for experimentation that have complied using systematic high-throughput screening (HTS) experimentation and ML and data management engineering skills have opened up different protocols for understanding catalysis features at catalyst discovery [24-29]. One earlier study used OCM catalyst discovery from a bias-free and consistent process for HTS at OCM. They described their approach as "Catalyst discovery from Scratch" [30]. For this protocol, three active elements (M1–M2–M3) were selected randomly from Li, Na, Mg, K, Ca, Ti, V, Mn, Fe, Co, Ni, Cu, Zn, Sr, Y, Zr, Mo, Pd, Cs, Ba, La, Ce, Nd, Eu, Tb, Hf, W, or "none" to make 28 selections. Then, these were co-impregnated onto the support, which was also selected randomly from MgO, gamma-Al₂O₃, SiO₂, Ca(OH)₂, anatase-TiO₂, ZrO₂, Ba(OH)₂, La₂O₃, or CeO₂ to make nine selections. The dried powder was calcined at 1000°C for 3 h to supply mixed-oxide form. Then totally 300 catalysts were prepared and checked for reactivity under various reaction condition using the systematic HTS system. The generated bias-free catalyst library and data analysis with visualization, ML engineering, and constructed networking provided a perspective for views on OCM catalysis features that were different from those assisted with conventional catalyst chemistry methods [13, 31-33]. It is proposed that those studies based on

systematic HTS may alter the style of catalyst discovery. However, the utility of such powerful tools of HTS has limited their popularity because of associated costs for construction and operation at present situation. Nowadays, the databases also can be accessible from the literature and/or an on-line open platform such as CADS [34-35]. Therefore, a need exists for discussing a means of applying open-resource datasets collected by different researchers including HTS datasets to assist various researchers during catalyst discovery and modification.

Herein, the author specifically examines the low-temperature OCM character derived La_2O_3 catalyst [19]. Reportedly, lanthanum-based catalysts such as La_2O_3 nanorod [36], $\text{La}_2\text{O}_3\text{CO}_3$ nanorods [37], and $\text{La}_2\text{Ce}_2\text{O}_7$ perovskite [38-39] have shown especially low onset temperatures of OCM: 420-550°C. However, multi-element-supported La_2O_3 features have rarely been investigated [22]. Therefore, the author believes that ML-aided catalyst investigation revealing a new multi-element-supported La_2O_3 catalyst possessing much unique lower temperature OCM feature has become a notable achievement [40].

2. Experimental

Three-element-supported La_2O_3 catalysts were prepared using co-impregnation method with an aqueous solvent of mixed metal resources at 50°C for 6 h. The loading amount for each metal was fixed at 0.3 wt%, in theory. After evaporating the water solvent, the remaining mixed powder was dried at 110°C. Then it was calcined at 600°C for 3 h. No-metal-supported La_2O_3 (bare), denoted as none/ La_2O_3 , was treated using the same protocol. The OCM reaction was performed with as-prepared catalyst (50 mg) in a conventional fixed-bed reactor system (downstream) with a furnace ($L = 270$ mm) in 50°C intervals at 400-850°C. Pre-treatment was applied at 400°C for 1 h under an O_2 flow (20.0 ml min^{-1}). Then, a typical OCM stream composed of $\text{CH}_4/\text{O}_2/\text{N}_2 = 12.0/6.0/3.0 \text{ ml min}^{-1}$ was used to evaluate the OCM reactivity. It is noteworthy that a standard Na-Mn-W/ SiO_2 gave a significant C_2 yield of 24.8% at 800°C, whereas none/ La_2O_3 served C_2 yield of 11.2% at 500°C

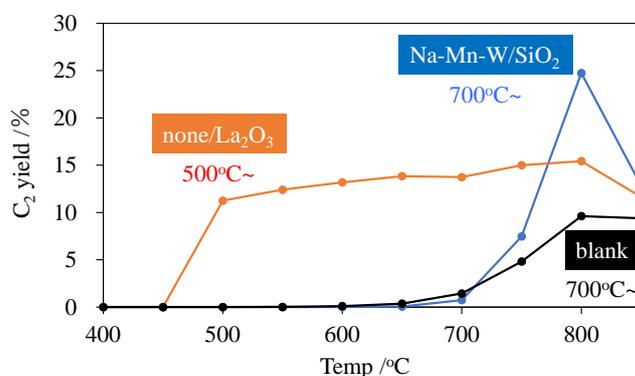


Figure 1. Trends of typical OCM catalyst at present reaction condition at $\text{CH}_4/\text{O}_2 = 2.0$.

under the conditions ($\text{CH}_4/\text{O}_2 = 2.0$) shown in **Figure 1**. The findings indicated clearly that the former had a high potential at higher temperature. The latter possessed a unique feature as a lower onset temperature in OCM reaction.

Then, ML prediction was conducted using the open-source HTS data (40,330 points by 350 catalysts) collected by a collaborator. The data were from surveys of OCM catalyst potential at 700–1000°C under various reaction conditions of the CH_4/O_2 ratio, total flow volume, and $\text{CH}_4 + \text{O}_2$ conc. [20, 30]. To describe the catalyst composite information of three-element-impregnated catalyst (M1–M2–M3/Support), a one-hot encoding manner was used. Binary numbers 1 and 0 were assigned to the corresponding index cell for composite element and support. The scheme was implemented in accordance with reaction conditions and the corresponding C_2 yield (**Figure 2**). Furthermore, for extracting appropriate trends of data, the data for validation were selected in accordance with the following conditions: total flow greater than 20 mL min^{-1} , $\text{CH}_4 + \text{O}_2$ conc. over 85%, and $\text{CH}_4/\text{O}_2 = 2.0$. A difficult issue exists for HTS dataset-driven ML at this study: because the HTS data were obtained for higher temperatures (700–1000°C) but not for lower temperature trends (approx. 400–600°C) of OCM performance, direct validation of predicted C_2 yield might have insufficient guidance to discover low-temperature La_2O_3 -based OCM catalysts. To overcome this shortcoming, the author revisited the research history made with collaborators using ML engineering tools (*vide spuria*) for interpretation of the meanings of the trends covered by ML regression models. Consequently, the assumption that the ML regression field might emphasize friendly element combinations (M1–M2–M3) for OCM showing higher C_2 yield was applied for this study to ascertain components that are useful for enhancing the unique characteristics for La_2O_3 .

Cat. Comp.	[Cat. comp. described with a one-hot encoding manner]							[Reaction condition]				[Target]
	Ba	Ca	Mn	Co	Na	Al_2O_3	SiO_2	Temp.	P_{CH_4}	P_{O_2}	P_{balance}	C_2 yield
BaCaMn/ Al_2O_3 :	1	1	1	0	0	1	0	1273 K	0.56	0.29	0.15	x %
CaMnCo/ SiO_2 :	0	1	1	1	0	0	1	973 K				y %
MnCoNa/ SiO_2 :	0	0	1	1	1	0	1	1173 K				z %

Figure 2. Example of one-hot encoding implementation at this study.

3. Results and Discussion

First, a support vector regression (SVR) model was applied to represent C_2 yields at 40,330 points (350 catalysts) covered by HTS. The data points derived from nine catalysts were removed because of extraordinary trends in selectivity. This model score at cross-validation of train-and-test examination was 0.86. Then, specifically examining the M1–M2–M3/ La_2O_3 category at the constructed SVR field predicted 41 three-element combinations at La_2O_3 -based catalysts with

features affording C_2 yields higher than 16.01% at higher temperatures. The TOP 20 were selected from the SVR field at La_2O_3 support. Then their potential for use as a low-temperature OCM based on La_2O_3 was examined, as shown in **Figure 3**. The expected score of C_2 yield at the SVR field, the predicted C_2 value was higher to the left side in the figure, did not contribute directly to the enhancement capacity of La_2O_3 character over that of low onset temperature. However, an interesting finding was that M1–M2–M3 components of 11 types can afford C_2 yield at 450°C, which was a lower onset temperature than the 500°C observed for none/ La_2O_3 . The components (C_2 yield at 450°C) were YEuHf (13.1%), CaYHf (10.1%), CaYEu (15.1%), CaYLa (15.0%), CaYBa (13.6%), MgNdHf (9.5%), CaEuHf (13.2%), CaYNd (10.2%), SrNdHf (9.7%), CaNiY (6.1%), and MgEuHf (11.4%). Although the remaining nine validations were not matched with the assumption, one was more likely to find undiscovered and effective catalysts with such *indirect* ML effectively from different areas of the collected datasets. Indeed, the hit rate for finding the low-temperature OCM catalysts was 55% (11 appearances per 20 validations), which is apparently a highly attractive rate.

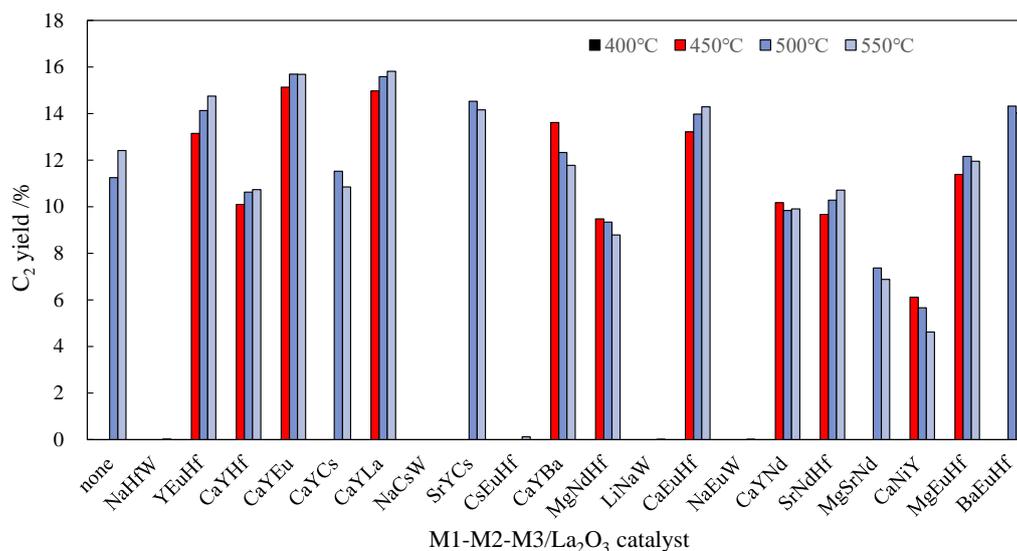


Figure 3. Plots of C_2 yield over M1–M2–M3/ La_2O_3 selected from TOP 20 category in the SVR field based on HTS data. The order of element from left to right was corresponding to the predicted C_2 yield from high to low values at the SVR field.^[10,40]

Next, implementation of data from the literature combined with the tailor-made HTS data was examined. The HTS data were compiled by 28 selections of elements and 9 selections of supports, as described above. Therefore, the output area of ML prediction using a one-hot encoding method was limited within such original indexes. To investigate more undiscovered La_2O_3 -based catalyst, data from the literature of 1,802 points (1,286 catalysts) [3] were added for subsequent ML prediction approaches. At that time, 59 catalysts derived from Na-Mn-W based catalyst in HTS [20] were excluded. Also, to eliminate data biases derived from the Na-Mn-W-like category and various

reaction conditions at HTS experimentations, the best C_2 yield values were applied for constructing an ML regression model from 300 random HTS data [30]. Then, the score at renewed SVR became lower to be 0.45 at the train-and-test examination. That might be attributed to bias in the literature data, which were inferred from different reactor systems and conditions at variety laboratories. However, when this challenge implies a different possible area, it would be an interesting strategy for three-element component enhancing the La_2O_3 nature as a low onset temperature OCM catalyst. Random forest regression (RFR) was also applied to the processed data: its score was 0.51. The individual TOP 12 of M1–M2–M3/ La_2O_3 components selected from SVR and RFR fields at the predicted C_2 yield were prepared and examined for those features at OCM. It is particularly interesting that MgSrYb (C_2 yield at 450°C: 12.1%) and GaSrYb (14.9%) from SVR assistance and GaTbHf (7.5%), ZnGaHf (3.8%), GaEuHf (7.5%), GaYEu (5.5%), ZnGaY (7.2%), ZnGaEu (9.3%), GaYbHf (6.3%), GaSrEu (7.6%), GaSrY (8.3%), GaSrHf (1.8%), and GaEuTb (8.6%) from RFR assistance were newly identified as low-temperature OCM catalysts based on La_2O_3 in each of 12 validations, as shown in **Figure 4**. Only the MgSrNd duplicated earlier experimentation by SVR using HTS data (**Figure 3**), although this was inactive at 450°C. Although RFR was composed mainly by GaHf derivatives, results showed that the addition of literature data into the HTS datasets can be an effective approach for emphasizing the outer area capacity in ML prediction. An additional 11 catalysts possessed C_2 yields higher than 5.0% at 450°C.

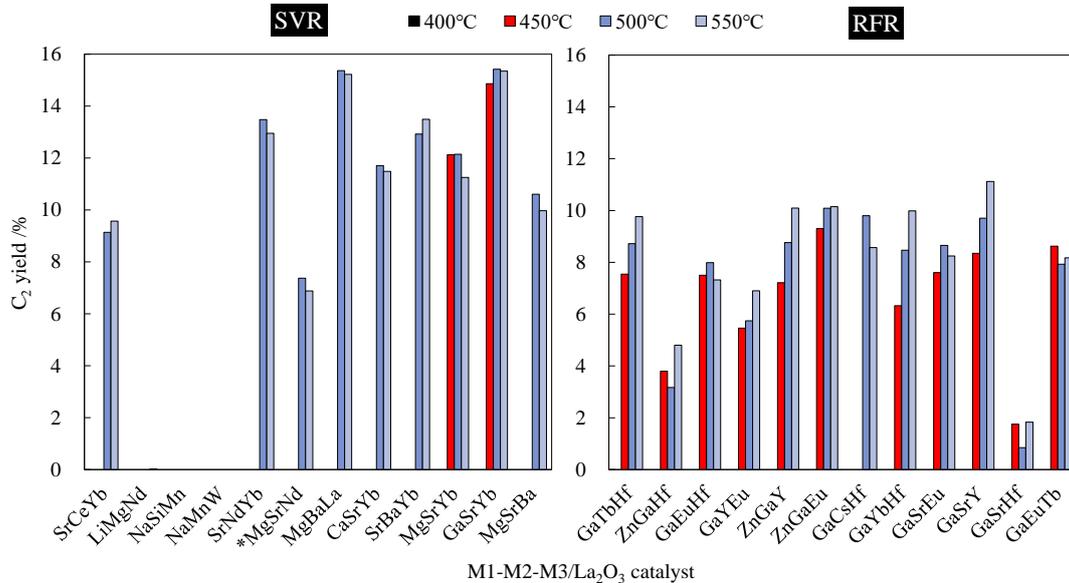


Figure 4. Plots of C_2 yield over M1–M2–M3/ La_2O_3 selected from TOP 12 category in the SVR and RFR field based on HTS and literature data. The order of element from left to right corresponding to the predicted C_2 yield from high to low values, respectively, at SVR and RFR field.^[40]

To emphasize the important potential of 24 as-revealed La_2O_3 -based catalysts, the selected 10 catalysts affording C_2 yields higher than 10.0% at 450°C under $\text{CH}_4/\text{O}_2 = 2.0$ were examined further to assess the OCM performance at $\text{CH}_4/\text{O}_2 = 3.5$, which condition is a stricter condition for CH_4 activation at low temperatures. Under the present conditions, none/ La_2O_3 showed an onset temperature of 600°C for appearance of C_2 yield (1.2%). **Figure 4** shows that all components retained a positive effect on the La_2O_3 feature of a low onset temperature in OCM at $\text{CH}_4/\text{O}_2 = 3.5$. It is noteworthy that the two catalysts of CaYHf and MgEuHf exhibited C_2 yields at 450°C of 3.6% and 3.1%, respectively, even under the $\text{CH}_4/\text{O}_2 = 3.5$ condition. Additionally, the two catalysts of CaYBa and YEuHf respectively showed attractive C_2 yield of 8.3% and 2.8% at 500°C . These results indicate that the as-revealed La_2O_3 catalysts promoted positive potential for further investigations, such as performance improvement and mechanistic study at next stage in conventional methods used for catalyst chemistry.

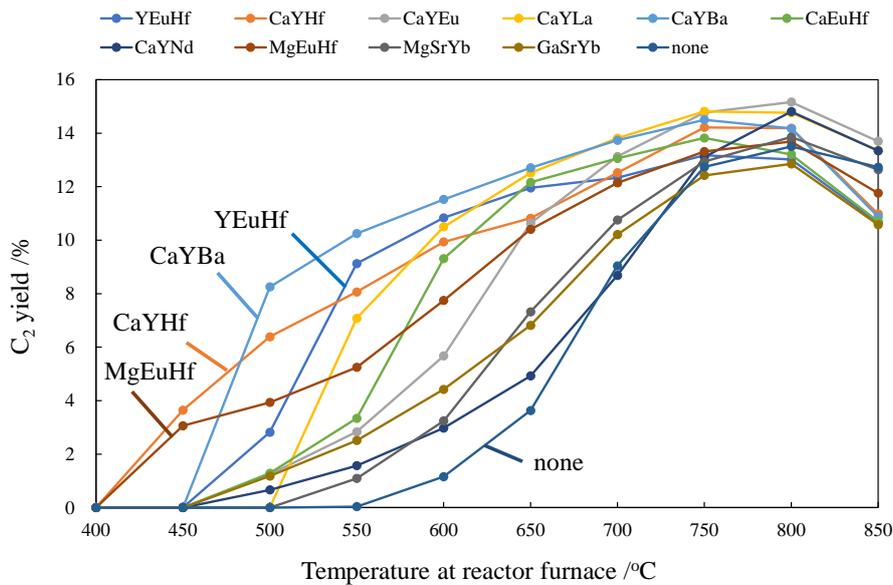


Figure 4. Plots of C_2 yield as a function of reactor temperature over selected 11 types of M1–M2–M3/ La_2O_3 at $\text{CH}_4/\text{O}_2 = 3.5$.^[40]

4. Conclusions

Specifically examining the unique OCM features derived from La_2O_3 catalyst, this investigation of multiple components supported La_2O_3 catalyst was conducted by ML engineering for the combined use of open-resource HTS datasets collected by a collaborator [20, 30] and literature data summarized in the review article [3]. To overcome mismatching between the target temperature area (approx. $400\text{--}600^\circ\text{C}$) and the collected data area (mainly at around $700\text{--}1000^\circ\text{C}$) in the dataset

resources, *indirect* ML prediction was used. Investigation was done based on the hypothesis that the M1–M2–M3 combinations giving a high C₂ yield at high temperature predicted by a direct ML prediction are helpful for finding familiar multiple components on the La₂O₃ support, consequently enhancing the unique nature of low onset temperature in OCM for La₂O₃ itself. It is particularly interesting that three elements of 11 types in 20 validations were found to be effective M1–M2–M3 components when using HTS data-driven ML with SVR. Moreover, the combined use of HTS and data from the literature further revealed additional 11-13 types of active M1–M2–M3 component La₂O₃-based catalysts at 450°C. This finding can promote further studies of improvement, further revealing the roles of the respective components at next step in conventional styles of catalyst chemistry. Accordingly, this report presents a successfully applied approach for discovering heterogeneous catalysts with ML assistance [40]. The use of ML to identify knowledge from trends of data to supply new ideas for conventional scientists has also been presented in reports of our recent studies [41-43].

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