Catalyst Informatics Based on Element Substitution Strategies for Systematic Catalyst Development

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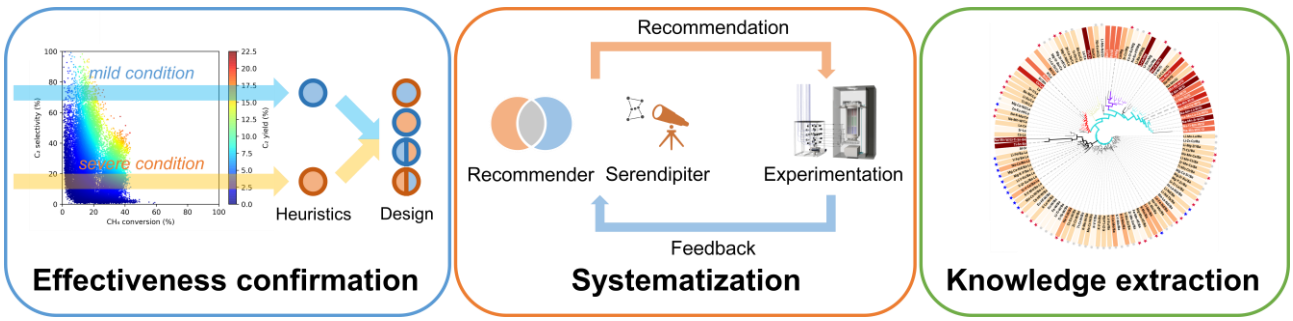
Solid catalysts are crucial in industry for producing a diverse range of chemical compounds. However, their working mechanisms are complex, influenced by many variables across different spatial and temporal scales. This has prevented rational catalyst design based on researchers' understanding; historically, most practical catalysts have been developed through empirical trials and errors, often accompanied by serendipitous discoveries. However, the traditional trial-and-error method of catalyst development is essentially ad hoc and cost intensive. In this thesis, I attempted to systematize catalyst discoveries based on a strategy of elemental substitution in a data-driven manner, aiming at establishing a reproducible and generalizable method for discovering high-performance catalysts. In doing so, a catalyst dataset featured with both high quality and high quantity was essential. A dataset for oxidative coupling of methane, previously acquired by us through high-throughput experimentation, was adopted as a training dataset [1]. The same experimental system was also used to validate concepts for individual chapters.

In **Chapter 2**, combinatorial rules for superior methane conversion under mild conditions and those for suppressing deep oxidation under severe conditions were extracted from the training data. These rules were successfully combined to design novel OCM catalysts. Key findings include temperature-dependent catalyst design guidelines, the importance of support properties, and the effectiveness of mixed supports like La_2O_3 and BaO. This chapter validated the effectiveness of catalyst design based on combinations of catalyst components.

In **Chapter 3**, to systematize catalyst discovery based on component combinations, an adaptive sampling loop has been designed with implementing a catalyst recommender system which can estimate catalyst performance through the substitution of component combinations. The system is based on the evidence theory, a mathematical framework to quantify the certainty of a hypothesis by combining multiple pieces of evidence and capable of managing uncertainties. It recommends catalysts according to the degree of belief that the catalyst is good or uncertain, which is equal to controlling the exploration/exploitation trade-off. The adaptive sampling led to the discovery of various high-performing catalysts. Moreover, by finding discernible patterns within serendipitous catalysts (unexpectedly high-performing catalysts), a catalyst serendipiter system was developed. The serendipiter system was designed to predict the occurrence of serendipity based on catalyst compositions and prediction outcomes from various classifiers, working in tandem with the recommender system. The system was exploited to induce serendipitous catalysts.

In **Chapter 4**, a method to visually extract the combinatorial knowledge for catalyst design from a set of data with different backgrounds was introduced based on a phylogenetic tree. The developed method was not only able to visualize the history of catalyst development in OCM, but also clarified a significant contribution of my research (Chapters 2 and 3) to widen the scope of high-performing catalysts. Moreover, these visualizations highlighted that each catalyst system follows different rules of elemental combinations to achieve good performance. The insights gained from this analysis were exploited to design promising mixed oxide-based catalysts, a category that has been relatively underexplored in the history of OCM catalyst development.



In this thesis, systematic and efficient methods for data-driven catalyst discovery based on combinatorial rules have been developed. Due to the un-necessity of specific descriptors, the methods are generally applicable to other catalysis, and any cases where materials are developed from combinations of components.

[1] T. N. Nguyen, S. Nakanowatari, T. P. N. Tran, A. Thakur, L. Takahashi, K. Takahashi, T. Taniike. *ACS Catal.*, **2021**, 11, 3, 1797–1809.

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