Title	ハイスループット実験と自動記述子設計を用いたメタンドライリフォーミング触媒の探索
Author(s)	杜, 文涛
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Description	Supervisor: 谷池 俊明, 先端科学技術研究科, 博士

Abstract

DU Wentao (2220433)

Conventional catalyst development has largely relied on trial-and-error experiments, which are time-consuming and costly. To tackle this challenge, data-driven approaches, particularly machine learning (ML), have gained prominence in accelerating catalyst discovery and optimization. However, two key challenges must be overcome for the practical application of ML in catalyst development. First, complex materials like solid catalysts often lack sufficient experimental data for effective ML training. Second, designing comprehensive descriptors for materials typically requires deep domain knowledge. The advent of high-throughput experimentation (HTE) offers a powerful solution to the data scarcity issue in catalysis research. Additionally, a recently developed automatic feature engineering (AFE) technique effectively mitigates the need for prior system knowledge, addressing the challenge of descriptor design. Dry reforming of methane (DRM) is an important catalytic process, typically requiring temperatures above 700 °C for high reactant conversion. However, such conditions cause catalyst sintering and deactivation. Operating at 500 °C or lower presents a promising alternative, as it can mitigate catalyst degradation and reduce environmental impact. Nevertheless, under these milder conditions, undesirable side reactions tend to occur, resulting in carbon deposition. To overcome these challenges, the development of active, stable, and selective catalysts demands a multi-element design strategy. Therefore, this thesis aims to present an approach for developing multi-element DRM catalysts without prior knowledge, leveraging a combination of HTE, ML, and AFE within an adaptive experimental design framework.

The presence of a high-quality, large-scale, and consistent catalyst dataset is essential for effectively applying ML to explore the extensive materials space and achieve efficient catalyst design. Therefore, in **Chapter 2**, I present the acquisition of an unbiased training dataset for DRM at 500 °C, comprising 256 γ-Al_2O_3-supported multi-element catalysts. These catalysts were prepared through HTE by randomly combining 17 elements selected from the periodic table without preconceptions. The resulting data were analyzed from multiple perspectives to extract meaningful insights into catalyst design and catalysis. Overall, this chapter highlights the effectiveness of unbiased exploration in establishing a robust dataset for ML and identifying valuable catalyst design guidelines.

Catalysis research is often hindered by the limited size of available datasets. This poses challenges for training expressive ML models, which typically require numerous tunable parameters to capture complex trends. Additionally, the diversity and complexity of catalysts make it difficult to design comprehensive descriptors based on physicochemical intuition. To overcome this, in **Chapter 3**, a two-step ML approach with AFE was introduced to generate effective descriptors directly from composition, enabling simple models to accurately capture performance trends without prior knowledge. Using the unbiased DRM dataset constructed in Chapter 2, an active learning loop was implemented by integrating AFE, farthest point sampling (FPS), and HTE. This iterative framework expanded the compositional space from 17 to 45 elements and guided efficient exploration of the catalyst landscape. Finally, this approach enabled the construction of a robust predictive model for identifying superior catalysts across vast material space.

Low-temperature DRM is explored as a promising route due to its lower energy demands and improved economic feasibility. However, it remains susceptible to carbon deposition mainly via CO disproportionation. Carbon accumulation deactivates catalysts by blocking active sites and hindering reactant flow, increasing pressure drop and safety risks. To address this, in **Chapter 4**, I investigated high-performance catalysts identified through the active learning loop in Chapter 3. Thermogravimetric–differential thermal analysis (TG-DTA) was conducted after 6 hours of reaction at 500 °C to assess carbon deposition. The analysis reveals key relationships among catalytic activity, carbon formation behavior, and composition, offering essential guidance for designing highly active and carbon-resistant catalysts.

In summary, catalyst discovery has long been constrained by human preconceptions and domain knowledge, which limit exploration to narrow, well-understood regions of design space. This dissertation establishes a transformative strategy that promises to remove these constraints. Through a fully data-driven framework that integrates HTE, ML, and AFE, this study opens access to previously inaccessible areas of the materials landscape and accelerates the identification of high-performing catalysts. Furthermore, by leveraging predictive models and addressing key challenges such as carbon deposition, this study not only deepens our understanding of catalyst behavior but also provides actionable design guidelines for the development of highly active and durable catalysts.

Keywords: High-throughput experimentation, automatic feature engineering, dry reforming of methane, multi-element catalyst design, carbon deposition