JAIST Repository
https://dspace.jaist.ac.jp/

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
Citation	
Issue Date	2025-09
Type	Thesis or Dissertation
Text version	none
URL	http://hdl.handle.net/10119/20091
Rights	
Description	Supervisor: 谷池 俊明, 先端科学技術研究科, 博士

JAIST
JAPAN
ADVANCED INSTITUTE OF
SCIENCE AND TECHNOLOGY

Japan Advanced Institute of Science and Technology

氏名	DU, Wentao		
学位の種類	博士（マテリアルサイエンス）		
学位記番号	博材第 618 号		
学位授与年月日	令和 7 年 9 月 24 日		
論文題目	High-throughput experimentation and automatic feature engineering for catalyst discovery in dry reforming of methane		
論文審査委員	谷池俊明	北陸先端科学技術大学院大学	教授
	長尾祐樹	同	教授
	西村俊	同	准教授
	本郷研太	同	准教授
	長藤圭介	東京大学	教授

論文の内容の要旨

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

論文審査の結果の要旨

従来的な固体触媒開発は、その複雑性から実験的な試行錯誤に依ってきた。近年、機械学習などの AI 技術を用いたデータ駆動型の触媒開発が注目を集めている。その鍵は、機械学習に資する規模と質、一貫性を備えたデータと、触媒の本質を捉えた記述子（触媒材料を数値表現したもの）である。しかし、探索空間が未知かつ広大であるほど、データがない、記述子がわからない、というジレンマがあり、データ駆動型触媒開発は比較的既知な空間での最適化にとどまっている。

本論文は、独自のハイスループット実験（HTE）と記述子生成技術を組み合わせて用いることで、既知から完全に解放されたデータ駆動型触媒開発を実現する研究成果をまとめたものである。モデル反応として、メタンと二酸化炭素から水素を取り出すメタンドライリフォーミングに注目した。

第 2 章では、DRM 用のハイスループット実験系を構築した。周期表から無作為に抽出した複数の元素を任意組成で組み合わせた触媒を想定し、300 個もの触媒の低温 DRM の性能データを短期間で取得した。得られたバイアスのないデータを、様々な統計・機械学習手法で分析し、Ni や白金族元素といった従来の活性金属に、Li、Al、Nb、Hf などのプロモーターを組み合わせることで高活性や炭素析出抑制ができることを明らかにした。本章で明らかになったプロモーターのほとんどは文献未報告であり、既知にとらわれない探索の重要性が浮き彫りになった。

第 3 章では、探索を周期表全体に拡張し（探索空間は 10 億個程度の触媒を含む）、HTE と記述子生成をループさせる大規模な触媒探索を実施した。ループを経るほど、訓練データの多様性が拡張され、記述子や機械学習モデルがより高精度かつロバストになる。こうして得られたモデルは、広大な空間から多様な高性能触媒を高確率で提案できる。水素収率が基準の 20%を超える触媒を、実験的に 100 個以上発見した。これらは文献で報告された 35 個の高性能触媒より単に多いだけでなく、化学組成においても極めて多様であった。

第 4 章では、得られた高性能触媒に対して炭素析出挙動を評価し、触媒失活要因の解析を行った。Rh の添加が炭素形成を顕著に抑制することが示され、さらに Pt、Cu、Y、Pd などの元素の添加によっても炭素蓄積が抑制されることが確認された。

本研究は、完全なデータ駆動型触媒開発を初めて実証したものであり、DRM にとどまらず、今後の触媒開発に新たな展望をもたらすことが期待される。よって、本論文は博士（マテリアルサイエンス）の学位論文として十分に価値あるものと認めた。