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研究成果の概要(和文)：提案された計画のための不均一な触媒法を開発するために、私は(i)バイオマス由来の原料の価値ある化学物質への変換、(ii)バイオベースの芳香族炭化水素のメチル化、(iii)フェノール化、およびアミドの加水分解を含む触媒反応を実証しました/バイオベースの脂肪酸アミド、および(iv)金属酸化物に担持された金属触媒および機械学習に関する実験的および理論的研究は、将来の触媒設計を試みました。

研究成果の学術的意義や社会的意義

Catalytic transformation of biomass-derived feedstocks into valuable chemicals is a sustainable alternative to the chemical produced from fossil fuels. I have demonstrated Environmentally benign and low-cost methods using biomass-derived hydrocarbons and greenhouse gas carbon dioxide.

研究成果の概要(英文)：We have demonstrated catalytic reaction that includes (i) the transformation of biomass-derived feedstocks into valuable chemicals, (ii) the methylation of biobased hydrocarbons, (iii) experimental and theoretical studies on machine learning aided future catalysts design. Firstly, (1) acetalization of glycerol, (2) methanolysis of triglycerides, (3) selective alkenylation of oxindoles, and (4) phenolysis of amides to form esters are demonstrated. Second-year (1) catalytic methylation of biobased hydrocarbons, (2) hydrolysis of amides, (3) reverse water gas shift reaction, and (4) catalytic methylation of benzene analogs are explored by benign green catalytic methods, lastly (1) the role of metal in supported based catalysts, (2) metal-support interactions in metal-oxide-supported materials, (3) methylation of benzene using carbon dioxide and hydrogen, (4) machine-learning method to identify novel catalysts.

研究分野：グリーンサステイナブルケミストリーおよび環境化学関連

キーワード：Heterogeneous catalysis Biomass conversion

様式 C - 19、F - 19 - 1、Z - 19 (共通)

1. 研究開始当初の背景

Considering the environmental issues to build a sustainable society, the idea of green chemistry for chemical synthesis is a recent growing area, and developments in the catalytic process for green chemistry are essential. Furthermore, sustainable use of natural resources is currently a significant challenge in chemistry. In this context, biodiesel has emerged as an alternative to fossil fuels due to its environmental benefits. However, the rapid increase in biodiesel production resulted in increasing glycerol production as the byproduct, offering a promising opportunity for upgrading this waste material. Unfortunately, the developed catalytic methods for converting biomass-derived feedstock (e.g., glycerol, triglycerides) into chemicals suffer from low selectivity. Therefore, it is also desirable to upcycle the biomass to value-added chemicals such as fatty acid methyl ester (FAMES), fatty acid, fatty amides, hetero-amines (pyrazines), and biomass-derived hydrocarbons. In this sense, I propose the research subject of developing heterogeneous catalysts for oxidant-free conversion of biomass to fine chemicals.

2. 研究の目的

The development of heterogeneous catalysts that enable to upcycle of the biomass-derived feedstock in reaction with non-fossil chemicals (biomass-derived alcohols, aldehydes, ketones, phenols), and gases (CO_2 , NH_3) are aimed at this research. The high TON, atom efficiency, reusability, and easy catalyst-product separation of this heterogeneous method can lead to its practical use in biomass conversion. Specifically, we attempted the development of reactions, development of catalysts, and mechanistic establishment for these two types of reactions (i) the transformation of biomass-derived feedstocks triglycerides, glycerol, fatty amides, and phenols into valuable chemicals, and (ii) methylation of biobased hydrocarbons,

3. 研究の方法

In addition to catalytic reactions and experimental methods, we also established in situ / operando spectroscopy and computational scientific methods to demonstrate the structure-activity relationship for these proposed reactions. Based on this design guide, novel heterogeneous catalysts for the assigned reactions have been developed.

4. 研究成果

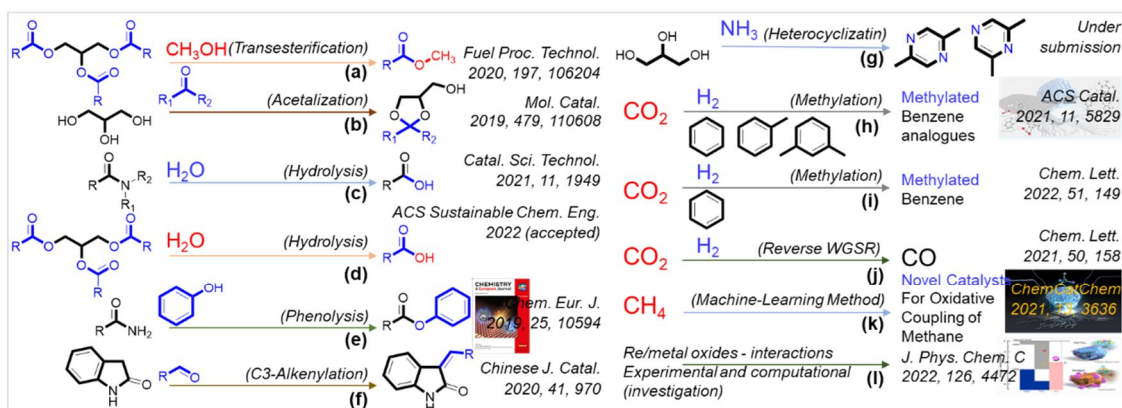


Figure 1. H β zeolite catalyzed transesterification of triglycerides and acetalization of glycerol (a) and (b); Nb $_2$ O $_5$ catalyzed hydrolysis of amides/fatty amides and triglycerides (c) and (d); CeO $_2$ catalyzed phenolysis of un-activated amides and selective alkenylation of oxindoles (e) and (f); Carbon supported Pt catalyzed Selective pyrazine synthesis (g); Combined TiO $_2$ supported Re and H β zeolite catalyzed methylation of *m*-xylene, toluene and benzene using CO $_2$ and H $_2$ (h); Combined MoO $_3$ (30)/TiO $_2$ supported Pt and H-Mordenite (SiO $_2$ /Al $_2$ O $_3$ = 90) catalyzed methylation of benzene (i); TiO $_2$ supported Re catalyzed reverse water gas shift reactions (j); Machine-Learning Method to identify the novel catalysts for oxidative coupling of methane (k); Experimental and theoretical investigation of metal (Re) support (metal oxides) interactions (l).

(a) Transesterification of triglycerides with methanol to get fatty acid methyl esters (FAMES): We present a simple heterogeneous catalytic method for the selective transformation of triglycerides into FAMES. For the methanolysis of trilaurin under reflux of methanol, a commercially available high-silica H β zeolite (H β -75, Si/Al=75) showed a higher yield of methyl laurate than other zeolite catalysts, metal oxides, and conventional heterogeneous and homogeneous catalysts. Under the optimized conditions, the method was widely applicable to transforming various triglycerides (C4–C18 frames) into the

corresponding FAMES (yields of 87–93%). The catalyst was furthermore reusable. Quantitative relationships between acidity, hydrophobicity, and reaction rates versus the Si/Al ratio of H β zeolite catalysts show a low affinity to glycerol, which arises from the hydrophobicity of the high-silica zeolites, is an important factor for controlling the catalytic activity.

(b) Acetalization of glycerol with aldehydes/ketones to get acetals/ketals: We demonstrated that a proton-exchanged *BEA zeolite with a high Si/Al ratio of 75 (H β -75) is an effective catalyst for the acetalization of glycerol with carbonyl compounds. This catalyst system was applicable to various substrates and reusable for at least four times with a slight decrease in activity. Furthermore, the turnover frequency, based on acid site concentration, increased as a function of H β Si/Al ratio, indicating the importance of the zeolite hydrophobic surface properties. The origin of the high efficiency exhibited by H β -75 is quantitatively discussed based on kinetic studies, hydrophobicity, and acid site concentration.

(c) Hydrolysis of amides/fatty amides into carboxylic acids/fatty acids: An efficient, simple, and versatile heterogeneous Nb₂O₅ catalytic system has been developed for hydrolysis of amides/fatty amides to carboxylic acids under relatively mild conditions. The quantity and strength of Lewis acid sites of Nb₂O₅ catalysts depended on their structure and morphology, where surface area, the number of Lewis acid sites, and their interactions with carbonyl groups decreased with an increase in catalyst calcination temperature. The catalytic performance can be attributed mainly to the facile activation of the C=O bond of the amides by surface Nb₅⁺ Lewis acid sites, which function even in the presence of basic inhibitors such as H₂O and NH₃. Moderated surface basicity was also found to play a role in the efficient progression of the reaction. This heterogeneous catalytic system accepted a broad scope of amide substrates (>40 examples; up to 95% isolated yield) along with catalysts reusability, additive-free, solvent-free reaction condition, and generation of ammonia and amines as byproducts from primary amides and secondary/tertiary amides respectively.

(d) Hydrolysis of triglycerides into fatty acids: We have developed a simple and highly efficient Lewis acid-promoted Nb₂O₅ catalyzed system for hydrolysis of triglycerides into fatty acids. Here, the catalysts' structure and morphology significantly exposed the quantity and strength of Lewis acid (LA) sites of Nb₂O₅ catalysts. A relatively low-temperature calcined (at 500 °C) Nb₂O₅ catalyst was found to be most reactive for the progress of this hydrolysis reaction. The comparative kinetic study suggests that the catalytic performance is attributed to the adroit activation of carbonyl bond by surface Nb₅⁺ LA sites even in the presence of hard bases like H₂O. This heterogeneous catalytic system adopted a broad scope of triglycerides (up to 99% isolated yield) with catalysts reusability, additive-free, and the generation of glycerol, a vital platform chemical byproducts.

(e) Phenolysis of unactivated amides into corresponding esters: The direct catalytic esterification of amides that leads to the construction of C-O bonds through the cleavage of amide C-N bonds is a desirable strategy in organic synthesis. We have developed a reusable catalytic method for the phenolysis of unactivated amides into the corresponding phenolic esters using CeO₂ as a catalyst. This catalytic system overcomes the typical stability issue of amides and phenols and thus offers a straightforward route to phenolic esters. This method is compatible with a wide range of substrates, including various functionalized amides and phenols. Results of kinetic studies suggest that the combined effects of the acid-base functions of catalysts are essential for the efficient progression of the C-N bond-breaking process. A plausible mechanistic study indicates that the rate-determining step of the reaction proceeds via a negatively charged transition state, in which the phenoxide engages in nucleophilic addition to the carboxylate species, followed by the transformation into the ester.

(f) Selective C3-alkenylation of oxindoles with aldehydes: We report herein that a commercially available CeO₂ is an active and reusable catalyst for the C3-selective alkenylation of oxindole with aldehydes/biomass derived aldehydes under solvent-free conditions. This catalytic method is generally applicable to different aromatic and aliphatic aldehydes, giving 3-alkylidene-oxindoles in high yields (87%–99%) and high stereoselectivities (79%–93% to *E*-isomers). This is the first example of the catalytic synthesis of 3-alkenyl-oxindoles from oxindole and various aliphatic aldehydes. The Lewis acid-base interaction between Lewis acid sites on CeO₂ and benzaldehyde was studied by in situ IR. The structure-activity relationship study using CeO₂ catalysts of different sizes suggests that a defect-free CeO₂ surface

is an active site for this reaction.

(g) Dehydrogenative synthesis of pyrazines from glycerol and ammonia: We report acceptorless dehydrogenative one-pot synthesis of pyrazines from glycerol and ammonia using a carbon-supported Pt catalyst (Pt/C). This reaction proceeds with high atom efficiency via sequential dehydrogenation, condensation, and dehydration steps, where two components undergo cyclization through selective C–N bond formations. The Pt/C catalyst is reusable, and the process gives complete conversion of glycerol to high yields (total 81%) of pyrazines (2,5 and 2,6 pyrazines). The effect of electronic states of various transition metals and the effect of Pt particle size reflect the catalytic activity as well as the turnover frequency per surface Pt atom.

(h) Methylation of benzene derivatives using CO₂ and H₂: In this work, the synthesis of methylated benzene derivatives via catalytic methylation of *m*-xylene using CO₂ and H₂ was demonstrated. Various combinations of hydrogenation catalysts and zeolites were tested and evaluated for this reaction. The combination of Re(1)/TiO₂ and H-β(40) exhibited the best performance, giving a relatively high yield and selectivity of methylated products. This catalyst system was also found to be compatible with the methylation of toluene under completely identical reaction conditions. In addition, machine learning approaches were used to analyze 328 experimental data points on the methylation of benzene, toluene, *m*-xylene, and naphthalene and to improve the performance of the catalytic methylation process. According to the PDP (Partial Dependence Plots analysis) based on the XGB (Extreme Gradient Boosting) prediction, among the investigated catalysts, the Re/TiO₂ catalyst with a Re loading amount of 1.8 wt % exhibited the best activity toward the methylation of benzene using CO₂/H₂.

(i) Methylation of benzene using CO₂ and H₂: Catalytic methylation of benzene using CO₂/H₂ was investigated using various combinations of supported metal catalysts and zeolites in a batch reactor. After a thorough catalyst screening process, the combination of Pt(3)/MoO_x(30)/TiO₂ and H-MOR (SiO₂/Al₂O₃ = 90) was the most efficient, achieving up to 39% benzene-based yield and 27% CO₂-based yield, without promoting the dearomatization of benzene, under the reaction conditions employed in this study (1 MPa CO₂, 5 MPa H₂, 1 mmol of benzene, 240 °C, 12 h).

(j) Reverse water gas shift reaction towards high CO selectivity: A Re catalyst supported on TiO₂ (Re/TiO₂) promoted the reverse water-gas shift (RWGS) reaction, exhibiting high selectivity toward CO. *Operando* X-ray absorption near edge structure (XANES) measurements revealed that the redox of Re species was responsible for the promotion of the reaction. The Re/TiO₂ catalyst also possessed remarkable durability and maintained >99.9% CO selectivity for more than 1000 h.

(k) Oxidative coupling of methane using an extrapolative machine-learning method to identify novel catalysts: We have constructed and analyzed an updated dataset consisting of 4759 experimental data points for the oxidative coupling of methane (OCM) reaction based on literature data reported before 2020 (~ 2019) using machine learning (ML) methods. Several ML methods, including random forest regression (RFR), extra trees regression (ETR), and gradient boosting regression with XGBoost (XGB), were used in conjunction with our proposed approach, in which elemental features are used as input representations rather than inputting the catalyst compositions directly. A recent research trend, namely, the extensive exploration of Mn/Na₂WO₄/SiO₂ catalyst systems in recent years due to their high activity and durability, was clearly reflected in the dataset analysis. An ML model for the prediction of the reaction outcome (C₂ yield) was successfully developed, and feature importance scores and Shapley Additive exPlanations (SHAP) values were calculated based on ETR and XGB, respectively, to identify the input variables with the most significant influence on the catalyst performance and observe how these important variables affect the C₂ yield in the OCM. The discovery and optimization of catalytic processes using ML as a “surrogate” model were explored, and promising catalytic system candidates for the OCM reaction were identified. Notably, the developed ML model predicted catalysts containing elements that do not appear in the OCM dataset. This clearly demonstrates the desirably high potential of our ML model to enable extrapolative predictions for ML-aided future catalysis research.

(l) Experimental and theoretical investigation of metal–support interactions in metal-oxide-supported rhenium materials: The activity and stability of supported metal catalysts is significantly

influenced by the interactions between the metal and the support, the so-called metal–support interactions (MSIs). Here, we present a systematic study using experimental and computational approaches to investigate MSIs between Re and oxide supports. Re dispersed on SiO₂, Al₂O₃, and MgO tends to be aggregated, whereas Re dispersed on TiO₂, V₂O₅, ZrO₂, Nb₂O₅, and CeO₂ exhibits a high degree of dispersion. Electronic properties such as the position of the conduction-band minimum (CBM) of the oxide support, a proxy for the electron affinity (EA), and the Fermi energy (EF) of the supported Re, a proxy for the work function (WF), were found to be of particular importance to govern the degrees of dispersion and aggregation of Re. Metal-oxide supports that have an EA that is larger than the WF of Re, such as TiO₂, V₂O₅, and CeO₂, can accept electrons from Re into their CB, inducing a strong MSI. This results in a large Re adsorption energy (E_{ads}) that leads to a high degree of dispersion for the supported Re metal. Our work describing the electronic interactions that govern the degree of dispersion will enable the synthesis of active and durable catalysts via the identification of suitable combinations of metals and oxides.

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5. 主な発表論文等

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〔産業財産権〕

〔その他〕

<http://www.cat.hokudai.ac.jp/shimizu/>

6. 研究組織

	氏名 (ローマ字氏名) (研究者番号)	所属研究機関・部局・職 (機関番号)	備考
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7. 科研費を使用して開催した国際研究集会

〔国際研究集会〕 計0件

8. 本研究に関連して実施した国際共同研究の実施状況

共同研究相手国	相手方研究機関
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