学 位 論 文 内 容 の 要 旨

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学 位 論 文 題 名

Development of Highly Active and Reusable Niobium-Based Complex Oxide Catalysts for the Dehydration of Biomass-Derived Sugars
（バイオマス由来糖類の脱水反応に高活性かつ再利用可能なニオブ系複合酸化物触媒の開発）

Biomass is a renewable carbon source critical for achieving carbon neutrality. Among biomass-derived products, furfural - obtained via acid-catalyzed dehydration of xylose - is recognized as a key platform molecule due to its wide applicability in biofuels, polymers, and fine chemicals. However, traditional catalytic systems such as amorphous niobium oxide (Nb_2O_5), while active, suffer from poor structural stability and loss of catalytic sites upon thermal regeneration, which hinders long-term usability. This dissertation aims to address these limitations by developing structurally stable, crystalline Nb-based complex oxide catalysts modified with phosphate to enhance both acidity and reusability in furfural production.

Chapter 1 introduces the challenges in furfural synthesis, summarizing catalytic strategies including homogeneous and heterogeneous systems. It highlights the promise of Lewis acid catalysis using Nb_2O_5, while identifying the need for improved stability, acidity control, and side-reaction suppression. The design rationale for crystalline complex oxides such as $CaNb_2O_6$, $SrNb_2O_6$, and $BaNb_2O_6$ is presented, emphasizing the structural advantages of alkaline earth metal incorporation and phosphate modification.

In Chapter 2, $CaNb_2O_6$ is synthesized using the amorphous metal complex (AMC) method and modified by phosphoric acid treatment. Characterization via XRD, XPS, BET, and FTIR-pyridine reveals that phosphate modification increases the number of Lewis acid sites (LAS) while introducing a small amount of Brønsted acid sites (BAS). In biphasic xylose dehydration, P–$CaNb_2O_6$ exhibits enhanced furfural yield and selectivity compared to both unmodified $CaNb_2O_6$ and P-Nb_2O_5. This improvement is attributed to the suppression of humin formation and the stabilization of active LAS. The catalyst also demonstrates superior reusability over multiple cycles, retaining structural integrity and catalytic performance after calcination.

Chapter 3 expands this approach to other alkaline earth metals (Sr, Ba), synthesizing $SrNb_2O_6$ and $BaNb_2O_6$ analogs via the same AMC method. Characterization shows that increasing cationic radius weakens Lewis acidity and enhances basicity, consistent with observed decreases in catalytic selectivity. Phosphate treatment consistently decreases LAS density but suppresses side reactions, leading to moderate gains in furfural selectivity. Among all tested catalysts, P–$CaNb_2O_6$ achieves the best balance of LAS distribution and stability, establishing it as the optimal candidate.

Chapter 4 summarizes the findings and presents future perspectives, emphasizing that structural regulation via cation selection and phosphate surface engineering effectively modulates the acid-base properties of crystalline metal oxides. This approach offers a promising platform for designing robust solid acid catalysts for biomass valorization and furfural production under industrially relevant conditions.