

論文 / 著書情報
Article / Book Information

題目(和文)	
Title(English)	Development of Heterogeneous Catalysts for Alcohol Transformations
著者(和文)	SUARSIHEndah
Author(English)	Endah Suarsih
出典(和文)	学位:博士(工学), 学位授与機関:東京工業大学, 報告番号:甲第12870号, 授与年月日:2024年9月20日, 学位の種別:課程博士, 審査員:原 亨和,鎌田 慶吾,北野 政明,山本 隆文,伊澤 誠一郎
Citation(English)	Degree:Doctor (Engineering), Conferring organization: Tokyo Institute of Technology, Report number:甲第12870号, Conferred date:2024/9/20, Degree Type:Course doctor, Examiner:,,,,,
学位種別(和文)	博士論文
Category(English)	Doctoral Thesis
種別(和文)	論文要旨
Type(English)	Summary

論文要旨

THESIS SUMMARY

系・コース： Department of, Graduate major in	Materials Science and Engineering	系 コース	申請学位 (専攻分野)： Academic Degree Requested	博士 Doctor of	(Engineering)
学生氏名： Student's Name	Endah Suarsih		審査員主査： Chief Examiner	Michikazu Hara	

要旨 (英文 800 語程度)

Thesis Summary (approx.800 English Words)

In the approach to achieve green and sustainable production of chemicals, the ideal synthesis in terms of catalytic activity, selectivity, atom-, and step efficiency should be satisfied. [44] This approach often uses a catalysis pathway, and the reduction of toxic reactants, stoichiometric amount of additives, waste, energy, and number of operations. Experimentally, the one-pot method is one of the most powerful attempts to approach greener synthesis which allows the construction of complex structures from simple, benign, and readily available starting materials. In this sense, the utilization of alcohols that can be transformed into activated intermediates is a great advantage.

Alcohol is a readily available and relatively unreactive starting material to produce chemicals. Their abundance, readily available, easy handling, and environmentally benign properties make alcohol convenient to be utilized for a greener synthesis approach. Thus, a variety of research has been dedicated to developing efficient catalysts for alcohol transformations. The study in this thesis investigates heterogeneous catalysts based on Co and Pd for the formation of C-C and C-N bonds using alcohols as starting materials. The main purpose of this thesis is to develop an efficient catalyst for green alcohol transformation protocols namely borrowing hydrogen and acceptorless dehydrogenative coupling protocols. Moreover, the utilization of heterogeneous catalysts for this one-pot borrowing hydrogen and acceptorless dehydrogenative coupling methods will make this strategy even more promising.

In chapter two, the formation of the C-C bond through α -alkylation of ketone with alcohols via borrowing hydrogen protocols over Co-MgO/TiO₂ exhibited excellent catalytic performance. The catalyst managed to give higher activity than the noble metal catalysts including Ru, Pd, and Pt, and the direct cooperation of Co, MgO, and TiO₂ resulted in a high activity of the catalyst as well as highly selective to produce the desired alkylated product. The investigation about catalyst recyclability suggests that the stirring rate greatly affected the reuse experiment. In addition, the developed catalyst has successfully been applied for various scopes of ketones and primary alcohols with different functionalities generating a moderate to an excellent yield of corresponding alkylated products. Controlled experiments revealed that the co-deposited MgO turns to enhance the hydrogenation of the alkene intermediate.

Chapter three discusses the utilization of the developed catalyst for the direct amination of primary alcohols with NH₃ which shows that the Co-MgO/TiO₂ catalyst is also applicable for the formation of C-N bond formation to give the corresponding primary amines in high selectivity under milder conditions than the reported heterogeneous Co catalysts. Moreover, the catalyst is allowed to selectively produce either primary, secondary, or tertiary amines by simply changing the reaction conditions.

In chapter four, an alternative Pd/TiO₂ catalyst is developed for the synthesis of N-heteroaromatics compounds demonstrating the synthesis of pyrazines. Currently, the catalyst shows remarkably high catalytic performance and stability for the benchmark reaction system. The catalyst managed to be used for 4 reactions run without significant decrease in activity and selectivity. The role of both palladium metal nanoparticle and support is important for the reaction sequence to proceed well.

備考：論文要旨は、和文 2000 字と英文 300 語を 1 部ずつ提出するか、もしくは英文 800 語を 1 部提出してください。

Note：Thesis Summary should be submitted in either a copy of 2000 Japanese Characters and 300 Words (English) or 1 copy of 800 Words (English).

注意：論文要旨は、東工大リサーチリポジトリ(T2R2)にてインターネット公表されますので、公表可能な範囲の内容で作成してください。
Attention: Thesis Summary will be published on Tokyo Tech Research Repository Website (T2R2).