

論文 / 著書情報
Article / Book Information

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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	応用化学 応用化学	系 コース	申請学位 (専攻分野)： Academic Degree Requested	博士 Doctor of	(工学)
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要旨 (英文 800 語程度)

Thesis Summary (approx.800 English Words)

This study focus on the synthesis and catalytic reaction of two medium-pore zeolites: **MFI**-type titanosilicate (TS-1) and the novel **TUN**-type aluminoborosilicate ([Al, B]-TUN). Medium-pore zeolites could selectively produce the target products because of their unique shape selectivity. TS-1 has been widely used in industrial field and intensively researched in academic field as an important oxidation catalyst. Yet, further improving the catalytic performance and reducing the cost still remained a challenge. In addition, [Al]-TUN (**TUN**-type aluminosilicate) is another medium-pore zeolite similar to the **MFI** topology but it has unique structure features different from **MFI** as well. These interesting features endow [Al]-TUN with promising and novel catalytic applications. Although [Al]-TUN has been studied in various acid and redox reactions, the synthesis of [Al]-TUN was limited to a narrow Si/Al ratio resulting in restricted acidic properties and applications. Therefore, this thesis will deal with the improvement of TS-1 epoxidation activity and the modification of the acidic properties of **TUN**-type aluminosilicate. There are total five chapters:

Chapter I, the general introduction of zeolites and two medium-pore topologies: **MFI** and **TUN**. A brief history of zeolite was introduced firstly and followed by three important features of zeolites, the topology, the framework compositions and the resulting acidic properties. Finally, the applications of zeolites were introduced. The introduction of **MFI**-type titanosilicate and aluminosilicate (TS-1 and ZSM-5) and **TUN**-type aluminosilicate ([Al]-TUN) was also based on the above mentioned order.

Chapter II, the synthesis of TS-1 zeolites with high epoxidation activity. In this chapter, the effect of hydrolysis conditions and gel compositions on the catalytic performance of TS-1 were studied. I found that the temperature and duration time during the hydrolysis of Si and Ti sources have an important impact on the epoxidation performance of the TS-1, which has been long ignored in previous studies. In addition, the different amount of OSDA (organic-structure directing agents) and Ti content in the synthesis gel were studied and a new recipe for synthesis of highly active TS-1 was proposed. The different morphology, particle size and the Ti coordination were also discussed.

Chapter III, the synthesis of novel [Al, B]-TUN zeolites and application in *n*-hexane cracking. In this chapter, firstly, I tried to synthesis [B]-TUN. The results show that it is very difficult to directly hydrothermal synthesis of [B]-TUN. Under the current synthesis conditions, **TUN**-type zeolite synthesis prefers the Al atoms in the framework other than the B. Secondly, based on the above mentioned experiences, I tried to synthesis [Al, B]-TUN in a low Si/Al ratio gel. In such cases, the framework B content in the final solids were still limited to a low level. Finally, by applying a high Si/Al ratio gel, [Al, B]-TUN with a higher B content in the final solids was successfully synthesized. The different Al and B content, coordination, the subsequent acidic properties and cracking reaction behaviors were studied.

Chapter IV, the post-treatment of [Al, B]-TUN and its high performance in *n*-hexane cracking reaction. Firstly, a three-steps post-treatment method including steaming, HNO₃ acid and ammonium hexafluorosilicate (AHFS) treatment was applied to [Al, B]-TUN. This strong treatment method removed most of the strong acid sites. Secondly, two relatively moderate post-treatment methods, HNO₃ acid treatment and steaming treatment, were adopted, respectively. Through a proper post-treatment method, a novel **TUN**-type zeolite with high reactivity, high selectivity to propylene and stable catalyst duration was achieved. The relationship between the acidic properties and cracking reaction behaviors were studied.

Chapter V, the general summary and acknowledgement.

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 1copy of 800 Words (English).