

論文 / 著書情報  
Article / Book Information

題目(和文)	アルケンエポキシ化用チタノシリケートゼオライト触媒の構造ならびに活性点分布に関する研究
Title(English)	Study on the structure and active site distribution of titanosilicate zeolites as catalyst for alkene epoxidation
著者(和文)	JIXinyi
Author(English)	Xinyi Ji
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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

(博士課程)  
Doctoral Program

## 論文要旨

THESIS SUMMARY

系・コース： 応用化学  
Department of, Graduate major in Chemical science 系  
and engineering コース

申請学位 (専攻分野)： 博士  
Academic Degree Requested Doctor of (Engineering)

学生氏名： JI XINYI  
Student's Name

指導教員 (主)： 野村淳子  
Academic Supervisor(main)

指導教員 (副)： 横井俊之  
Academic Supervisor(sub)

要旨 (英文 800 語程度)

Thesis Summary (approx.800 English Words)

In this thesis, I described the effects of Ti distribution and zeolite structure over the alkene oxidation. This thesis was composed of 5 chapters. In the chapter 1, I briefly introduced the definition of zeolite, the general introduction of titanosilicate and the introduction of typical titanosilicate Ti-MWW.

The main body of this thesis was divided into two sessions. The first session consisted of two chapters, chapter 2 and 3. This session was focused on the estimation method of Ti distribution in titanosilicate and its application in Ti-MWW zeolite. Firstly, a method for estimating the Ti distribution in the titanosilicate zeolite framework was successfully developed based on the epoxidation of 1-hexene (1-HX) and 2-methyl-2-pentene (2-MP). The index of 1-HX/2-MP, which was defined by the total yield of 1-HX to that of 2-MP, could indicate the Ti distribution: low value meant that more Ti species were located in large space like the supercage in the MWW framework. Ti distributions of various titanosilicates were estimated. The Ti distribution in Ti-MWW-D (prepared by direct-synthesis method) was different from that in Ti-MWW-P (prepared by post-synthesis method): Ti atoms in Ti-MWW-D were more located in the supercage and Ti atoms in Ti-MWW-P were more located in narrow channels. And the 1-HX/2-MP was suitable for the estimation of Ti distribution in micropore titanosilicate.

In chapter 3, the Ti distributions of various Ti-MWW-P zeolites synthesized under different Ti contents and crystallization periods were estimated by the 1-HX/ 2-MP index. At low Ti content and short crystallization time, Ti atoms were preferably located in the large space like the supercage and side pockets. At high Ti content and long crystallization time, Ti atoms would be more located in the narrow channels. Furthermore, based on the relationship between organic structure-directing agent (OSDA) and Ti species, the mechanism of the introduction of Ti atoms into the MWW framework by the post-synthesis method was briefly described. At last, Ti-MWW-P with special Ti distribution was proved to be a good catalyst in selective oxidation of 4-vinyl-1-cyclohexene (VCH).

In addition, in chapter 4, two novel CON titanosilicates were successfully synthesized by post-synthesis gas phase method. Ti-CON zeolite was firstly synthesized based on conventional microporous CON-type silicate De-B-CON under  $TiCl_4$  vapor treatment. Ti atoms were inserted into the defect sites in CON framework and performed tetrahedrally coordinated state. In order to improve the diffusion capacity, mesopores were introduced into the CON zeolite. The mesoporous CON-type titanosilicate Ti-Meso-CON was synthesized depended on mesopores contained CON-type silicate Meso-De-B-CON which was prepared by unique sequential alkaline and acid treatment. Ti-Meso-CON zeolite contained higher Ti content and ca. 10 nm mesopores. The obtained titanosilicates Ti-CON and Ti-Meso-CON were both applied into various alkene epoxidations. Ti-CON exhibited good catalytic activity in 1-hexene epoxidation but for the bulky alkene epoxidation, Ti-Meso-CON performed higher activity than Ti-CON owing to the existence of mesopores and high Ti content.

In conclusion, I summarized all the chapters. This thesis described the estimation method of Ti site distribution and the development of novel titanosilicates which had potential industrial applications.

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).

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