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Article / Book Information

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種別(和文)	論文要旨
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（博士課程）
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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）
学生氏名： Student's Name	CAI, Yibing		審査員主査： Chief Examiner	横井 俊之	

要旨（英文 800 語程度）
Thesis Summary（approx.800 English Words ）

The development of porous materials has been attracting attention for various advanced applications in catalysis, adsorption, biosensors, energy storage, and so on. A key point in the field of catalysis is to design well-defined pore structure or introduce more active sites that could enhance the conversion of the reactants, yield and the selectivity of the target products. Among them, mesoporous carbon materials and extra-large pore zeolites are ideal candidates for these applications. This study explores two complementary strategies for enhancing catalytic performance through material design. In the first part (Chapters 2 and 3), mesoporous carbon replicas prepared using hard-template methods are investigated as support for Pt catalysts in C-methylation reactions. The influence of pore size variation and unique 3D pore connectivity on the catalytic performance are discussed. In the second part (Chapters 4 and 5), the interzeolite conversion (IZC) of extra-large pore aluminoborosilicate SSZ-53 zeolites are firstly explored, and the crystallization mechanism are investigated. A comparison between direct-synthesis and IZC is carried out, focusing on the Al distribution and catalytic activity in the acylation of 2-methoxynaphthalene.

In **Chapter 2**, silica nanospheres (SNSs) with different sizes of 6, 14, 22, 30 were synthesized using a modified sol-gel method with the usage of basic amino acid L-arginine, and the less-arranged SNSs were formed with addition of block copolymer P123, and adjustment of the pH in the suspension. Both types of SNSs served as templates in a hard-template method to produce different mesoporous carbon replicas (CRs), CR_FO_SNSs with 2D structure and CR_FOM_la-SNSs with 3D structure. The different stacking behaviors of the SNSs template on the porosity and surface area of CRs were investigated. Notably, CR_FOM_la-SNSs, derived from less-arrange SNSs template, exhibited a unique 3D structure with interconnected pore network, that showing a high BET surface area of 1400 ~ 1800 $m^2 g^{-1}$ and a large total pore volumes of 3 ~ 10 $cm^3 g^{-1}$. This highlights the superiority of the 3D mesopore structure and its potential as an excellent catalyst support.

In **Chapter 3**, the Pt catalysts loaded on the obtained CRs and commercial activated carbon (AC) were prepared using the EG method, where the ethylene glycol was used as the reductant. Pt nanoparticles on Pt/CRs had smaller particle sizes (~ 2 nm) and better dispersion than those on Pt/AC (~ 4 nm). The catalytic performance of all Pt catalyst was investigated on the C-methylation of 2-phenylethanol. Compared with Pt/AC, the catalytic performance

of the Pt/CRs was highlighted, showing the benefits of mesoporous structure while Pt/CR_FOM_la-SNSs performed better than Pt/CR_FO_SNSs. Among the synthesized catalysts, Pt/CR_FOM_la-SNS14 (mesopore size: 14 nm) showed the highest activity, achieving a product yield of 71.6%. This enhanced performance is not only attributed to the unique 3D mesoporous structure, but also the proper mesopore size, that affected the aggregation behaviors of Pt nanoparticles during the reactions.

In **Chapter 4**, the post-synthesis, direct-synthesis, characterization and catalytic performance of Al containing SFH-type zeolite SSZ-53 were discussed. The limitation of doping more Al into the SFH-type framework was found, where the sample with a Si/Al ratio in the synthesis gel is 60. The influence of Al incorporation on the catalytic performance was analyzed, where the acylation of 2-methoxynaphthalene was selected as the model reaction. Results indicated that the more Al was added, a higher conversion of 2-methoxynaphthalene and a higher selectivity of the target product, 1-acetyl-2-methoxynaphthalene, were obtained. This indicates that a new approach of synthesis method is required to improve catalytic activity.

In Chapter 5, interzeolite conversion (IZC) was employed to synthesize SFH-type zeolites with improved Al incorporation (Si/Al ratio of 60). The effects of the parent zeolite type, alkalinity, and synthesis temperature on the conversion process were systematically examined. Among them, MFI-type zeolite performed best as the parent materials, producing the Al-SSZ-53 samples with high purity and a satisfying Si/Al ratio of ~ 60. The time-resolved XRD and SEM analysis was conducted to understand the transformation behavior. It could be found that the synthesis time was shortened from 5 days in the direct-synthesis to 2 days by using IZC, and a special two-step pathway mechanism from MFI-type to SFH-type was explained by the solution-mediated theory. The Al distribution in the framework from solid ^{27}Al NMR was considered, where an additional peak appeared in the IZC sample, indicating a special Al-containing phase formed during the transformation process. The influence of Al distribution on physicochemical properties and the catalytic performance of 2-methoxynaphthalene were figured out.

To conclude, 3D mesoporous carbon supports with controlled pore sizes were successfully synthesized using the hard-template method, and SFH-type zeolites with higher Al content were obtained via interzeolite conversion (IZC). Both modifications successfully advanced the understanding of how pore structure and composition of porous materials influence the catalytic performance in reactions involving bulky molecules. The study provides valuable insights into the design of porous materials for developing more efficient catalysts.