

Synopsis of the PhD thesis entitled:

“Mesoporous catalysts of hydrodecyclization and isomerization of hydrocarbons”

by M.Sc. Karolina Grzeszczak

The aim of this study was to:

1) Identify the influence of the preparation conditions on the texture and morphology of mesoporous materials.

Study in this part of work included:

a) the influence of surfactant type on the properties of mesoporous materials

Cationic (quaternary ammonium bromides, DMPM-11 and M₂M-9) and sugar surfactants (decanol-N-methyl glucamide, MEGA); obtained in the Department of Organic and Pharmaceutical Technology in Faculty of Chemistry, Wrocław University of Technology were used as a template in synthesis of silica materials. Nonionic ethoxylated alcohols with alkyl chains of different lengths and different number of ethoxy groups (commercially available under the trade name Rokanole) and ethoxylated castor oil (commercially available under the trade name Rokacety) were used in the synthesis of silica-alumina materials. The premise of this part of the research was to obtain mesoporous materials that are characterized by a narrow pore distribution and high specific surface area (above 1000 m² / g).

It has been shown that the materials synthesised with the DMPM-11, M₂M-9 MEGA are characterized by a narrow pore diameter and specific surface area less than 1000 m² / g. From this materials, only material obtained with DMPM-11 as a surfactant has a texture similar to the SiSBA-15 materials. Materials obtained with Rokanol DB7 (7EO (C12 - C15), Rokanol T10 (8,5EO (C16 - C18)) Rokanol T6 (6EO (C16 - C18)), and Rokacet R40 (40EO-castor oil) have also a narrow pore diameter distribution and surface area greater than 1000 m²/ g. Among the materials synthesized with surfactants from PCC Rokita the greatest degree of arrangement had material synthesised with Rokanol T10.

b) the influence of synthesis method on porous materials properties (AISBA-15 and AIMCM-41)

The aims of these research was to increase the degree of substitution of Si atoms with Al atoms in AISBA-15 synthesized with aluminum sulfate and also to increase the pore diameter of AISBA-15 materials synthesized with methylcellulose as co-temple. In the typical synthesis conditions of MCM-41 NaOH is used for pH adjustment. It was assumed that the use of NH₄OH for pH adjustment will increase the acidity of the material, and that the change of pH regulator should not have a significant impact on the AIMCM-41 materials structure. The results showed that the modified method of synthesis allows to increase the incorporation of Al into the structure of AISBA-15. The material obtained by the modified method of synthesis was characterized by about four times higher Si / Al ratios in comparison to the material prepared according to the procedure adopted in the present study as a standard procedure. It was also found that the introduction of methylcellulose during synthesis of AISBA-15 increased the diameter of the pores. Materials obtained with the addition of methylcellulose had a less ordered structure. Material AIMCM-41 obtained with NH₄OH was characterized by a higher acidity, larger specific surface area and also degree of arrangement was improved compared to AIMCM-41 obtained with using NaOH.

2) Determining the influence of the support on the activity and selectivity of platinum catalyst in the hydrogenation reaction (1-methylnaphthalene), and hydrodecyclization reaction (decalin)

AISBA-15, ZrSBA-15, AIMCM-41, ZrMCM-41 and Al₂O₃ was used as a support in this part of research. Platinum catalysts obtained by using aluminum-containing supports (AISBA-15, AIMCM-41) had a higher hydrogenating activity (greater selectivity of methyldecalin) than the catalysts prepared with use of supports containing zirconium (ZrSBA-15 ZrMCM-41). Also higher activity and selectivity to ring opening products had catalyst containing aluminum than catalyst containing zirconium.

3) Determining the influence of the support on the activity and selectivity of platinum catalyst in hydrocracking and isomerization reaction (n-hexadecane)

In this part of research, new materials prepared with ethoxylated alcohol (Rokanole), AISBA-15 materials, SAPO-11, ZSM-22 microporous materials and micro-mesoporous material (containing AISBA-15 and SAPO-11, designated as H-1) were used as a support of platinum catalysts. H-1 material was obtained by introduction of SAPO-11 during ALSB-15

synthesis.

In these study also catalysts obtained by mechanical mixing of the microporous catalysts (Pt/ AISBA-15 + Pt/SAPO-11) and (Pt/AISBA-15 + Pt/ZSM-22) was used in hydroconversion of n-hexadecane. The platinum catalysts that support was obtained with a Rokanole has a comparable activity and selectivity to isomerization products in n-hexadecane conversion as Pt/AISBA-15catalysts.

It was found that the synthesis method of AISBA-15 and alumina precursor used in AISBA-15 synthesis affects platinum catalysts activity. The platinum catalyst obtained by using aluminum sulfate as a precursor had higher activity in n-C16 conversion than a catalyst prepared with aluminum isopropoxide. Platinum catalysts obtained with materials that was synthesised with ethoxylated alcohols (Rokanol DB7 and Rokanol T10) had higher activity in n-C16 conversion when compared to the Pt/AISBA-15 (synthesised with standard method). Platinum catalyst with micro-mesoporous (H-1) support had lower activity and comparable maximum isomerization yield to the Pt/AISBA-15 catalyst (standard method synthesis). The activity of catalysts (Pt/AISBA-15 + Pt/SAPO-11) and Pt/AISBA-15 was comparable.