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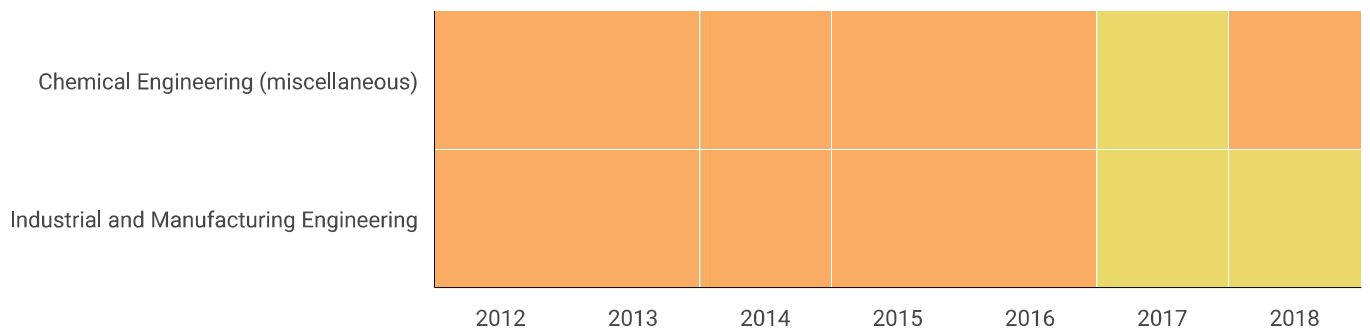


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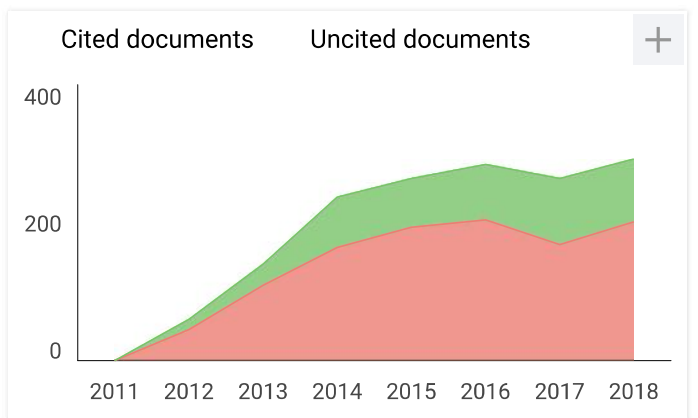
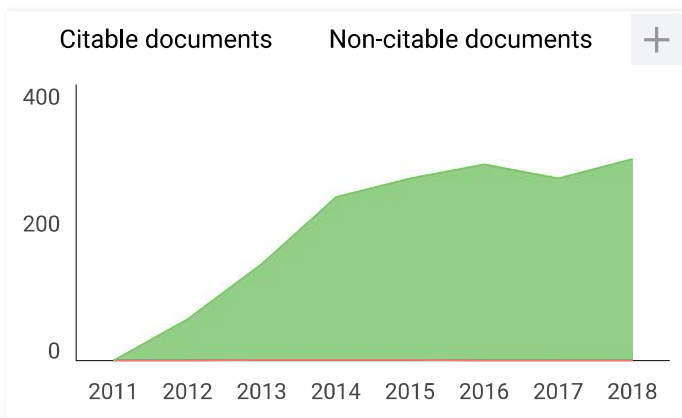
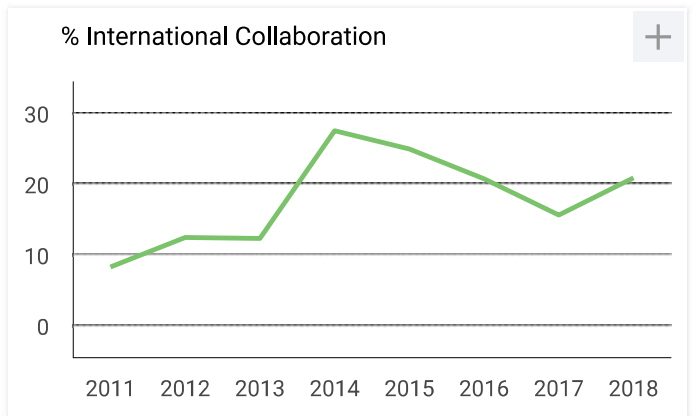
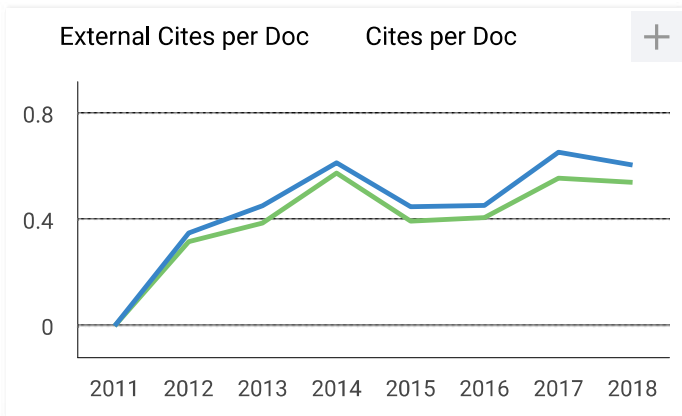
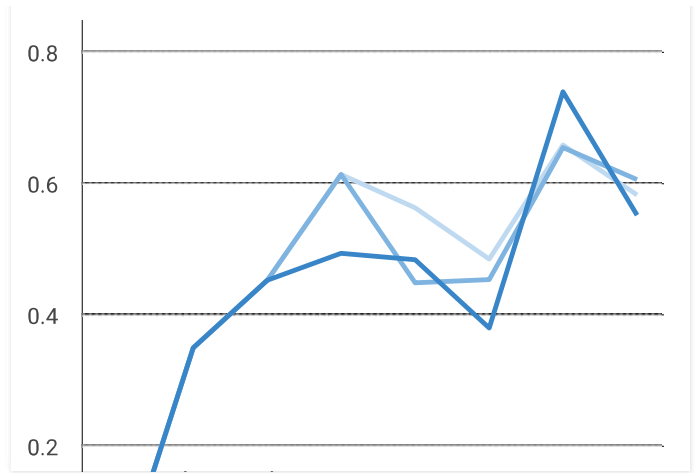
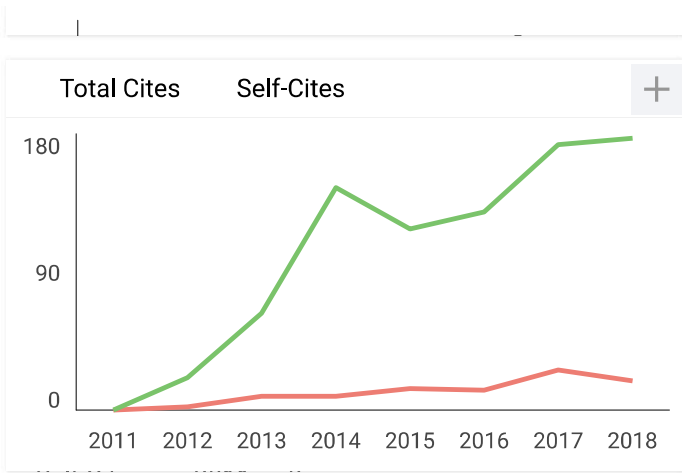
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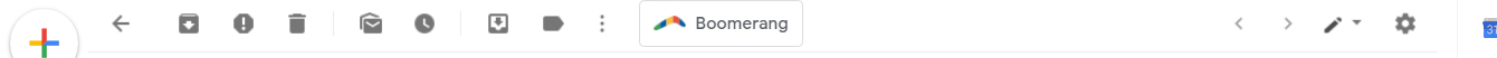
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Thank you very much for the preprinted our paper.  
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We put a yellow mark on the part of the paper that needs to be fixed:  
1) caption at Figure 1 (page 1146)  
2) table 1 (some numbers, need to replaced , with .) (page 1147)  
3) some numbers, need to replaced , with . (page 1147)  
4) "ca" needs to be deleted or replaced with "around"  
(page 1147)  
5) please add caption a) and b) at the Figure 4 (page 1148).

Thank you.  
Best regards,

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Department of Chemistry  
Faculty of Science and Technology  
Universitas Airlangga  
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Nonaktifkan untuk: Inggris ✕

Dear Hartati,  
Urgently answer me:  
- page 1145 Fig. 1 "modernite" or "mordenite"  
- page 1146: I do not understand what to change concerning 445,53; 545,82; 781,12  
Prof. B. Koumanova



**Hartati Dodit** <hartatidodit@gmail.com>  
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**Bogdana Koumanova**  
Please, answer my questions!

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Kam, 28 Sep 2017 09.22 ☆ ↶ ⋮

Dear Prof. B. Koumanova

I apologize for my less obvious explanation.  
- page 1145 Fig. 1 : please replace with "mordenite" (mordenite is a zeolite mineral with the chemical formula:  $\text{Ca}_{10}(\text{Na}_2\text{K}_2)\text{Al}_2\text{Si}_4\text{O}_{24}\cdot 7\text{H}_2\text{O}$ ).  
- It should be written: 445.53; 545.82; 781.12 (because 445,53; 545,82; 781,12 are Indonesian stlie).

Thank you very much

## ORGANIC TEMPLATE FREE HIERARCHICAL ZSM-5 PREPARED BY DESILICATION

Hartati, Alfa Akustia Widati, Aning Purwaningsih,  
Alfinda Novi Kristanti, Anggarani Nur Oktavia

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### ABSTRACT

Organic template free hierarchical ZSM-5 were synthesized using desilication. X-Ray Diffraction (XRD), Fourier Transform Infra Red (FTIR) spectrometry, and  $N_2$  adsorption/desorption analytical techniques were applied to characterize the physicochemical properties of the solid synthesized. Hierarchical ZSM-5 of a molar composition of  $100SiO_2:1.25Al_2O_3:1800H_2O$  was obtained at a hydrothermal temperature of  $175^\circ C$  for 36 h. The analytical results showed that the hierarchical ZSM-5 had a mesopore surface area, a pore volume, and a pore diameter of  $42.752 m^2/g$ ,  $0.186 cc/g$ , and  $3.810 nm$ , respectively.

**Keywords:** hierarchical ZSM-5, desilication, organic template free.

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### INTRODUCTION

Zeolites are microporous crystalline solids of unique properties such as a thermal stability, a high acidity, a good selectivity, and a high ion exchange capacity. Therefore, they can be used as catalysts, adsorbents, and ion exchange agents [1]. Natural zeolites often contain impurities that can cover the pores or active sites, so activation is required for their elimination [2].

Synthetic zeolites are generally made through a crystallization process of sodium aluminosilicates gels. In this case, a mixture of sodium silicates and sodium aluminates [3] is the source of silica and alumina for zeolites synthesis. Synthetic zeolites are commonly applied in industries because of their high purity and uniformity of particle size.

In general, ZSM-5 zeolites are synthesized using structure directing agents (SDA). Tetraprophylammonium hydroxide (TPAOH) is the most effective substance as a SDA [4]. Although TPAOH provides a good crystal structure, it also causes economical, health, and environmental problems because it is expensive, toxic

[5], and promotes air pollution in the the course of the calcination process [6, 7].

Most of ZSM-5 zeolites are prepared of a micropore size. Microporous ZSM-5 has limitations in respect to molecular diffusion due to steric effects, especially in case of bulky molecules [8]. There are many attempts to overcome this problem referring to the development of mesoporous ZSM-5 and hierarchical (microporous and mesoporous) ZSM-5. Meso-sized pores are expected to facilitate the reactants mass transport into the active site of the zeolite.

Mesopores can be obtained in zeolites with the addition of a cationic surfactant as a mesophase template. The surfactant can be eliminated, as is done by Goncalves et al. [9], Barakov et al. [10], Hartati et al. [11], or Jian et al. [12] upon completion of the synthesis process. However, the use of cationic surfactant results in air pollution, and which is why alternative methods are needed. Desilication is an easy method applied to the synthesis of various types of mesoporous zeolites, for example Mobile Five-1 (MFI) [13], Mordenite (MOR) [14], and Beta Polymorph A (BEA) [15]. Desilication can provide a controlled mesoporous structure maintaining Bronsted acidity [13].

In this research, ZSM-5 was synthesized using tetraethylorthosilicate (TEOS) and sodium aluminosilicate without structure directing agent or seeds. The resulting microporous ZSM-5 was then converted into a hierarchical (mesoporous and microporous) ZSM-5 through a desilication process.

## EXPERIMENTAL

Anhydrous sodium aluminate ( $\text{NaAlO}_2$ , Sigma Aldrich, 50 %), tetraethylorthosilicate (TEOS, Merck, 99 %), sodium hydroxide (NaOH, Merck, 99 %), ethanol ( $\text{C}_2\text{H}_5\text{OH}$ , Sigma Aldrich, 99.8 %) and distilled water were used. Approximately 0.375 g of NaOH were dissolved in 10 mL of distilled water and then gradually mixed with 0.256 g of  $\text{NaAlO}_2$  in a plastic beaker. The solution was then added dropwise to 14.1 mL of TEOS as a silica source and 10 mL of distilled water. Thus a mixture of a molar composition of  $10\text{Na}_2\text{O} : 100\text{SiO}_2 : 1.25\text{Al}_2\text{O}_3 : 1800\text{H}_2\text{O}$  was obtained. It was stirred for 5 h and aged for 19 h. Subsequently, it was transferred to a stainless steel autoclave. The latter was sealed and heated at  $175^\circ\text{C}$  of a varying time (24 h, 36 h, 48 h, and 72 h). Then the solids obtained were washed with distilled water to pH 7 using centrifugation techniques and dried in an oven for 24 h at  $100^\circ\text{C}$ . Thus microporous ZSM-5 was obtained.

The desilication process was carried out by introducing 0.176 g of microporous ZSM-5 to a mixture of 20 mL of water and 10 mL of ethanol placed in a plastic beaker. Then 20 mL of 0.125 M NaOH were added dropwise. During this process the plastic beaker was covered with aluminum foil to prevent evaporation. The mixture was transferred to a polypropylene bottle and heated for 24 h at  $100^\circ\text{C}$  [16]. The product obtained was washed with distilled water until pH 7 and dried for 24 h at  $80^\circ\text{C}$ .

The structure of ZSM-5 was determined by X-Ray Powder diffraction (XRD) at a wavelength  $1.54056 \text{ \AA}$  using a Philip Analytical X'Pert PRO of  $2\theta = 5 - 50^\circ$  and Cu  $K\alpha$  radiation. FT-IR spectra (Shimadzu IR Prestige 21 spectro-photometer) were used to identify the structure of ZSM-5 and the functional groups present. The pore properties prior to and following the desilication were studied using nitrogen absorption/ desorption (Nova Quantachrome 1200E).

## RESULTS AND DISCUSSION

The X-ray diffraction patterns of ZSM-5 synthesized in absence of an organic template with hydrothermal time varying at  $175^\circ\text{C}$  is shown in Fig. 1. It is evident that no ZSM-5 but an amorphous solid is formed within 24 h. The time increase to 36 h results in ZSM-5 synthesis. This is verified by the sharp peak at  $2\theta$  of about  $7^\circ - 9^\circ$  and  $22^\circ - 25^\circ$  (Fig. 1b). Fig. 1b shows a peak at ca  $6^\circ - 7^\circ$  which is characteristic peak of mordenite [14]. The sample obtained within 48 h (Fig. 1c) shows a diffraction pattern similar to that in Fig. 1b, but with an additional peak at ca  $27^\circ$  indicating the formation of quartz [17]. At 72 hours, Quartz dominates ZSM-5 in case of 72 h hydrothermal time as evident from Fig. 1d.

The data presented shows that the hydrothermal time increase leads to a lower crystallinity of MFI structure and promotes the formation of quartz and modernit. This is so because no structure directing agent of MFI is used [18]. Furthermore, quartz formation is facilitated by high NaOH concentrations of NaOH as reported by Prasetyoko et al. [19]. The characterization by FTIR spectra is aimed at determining the constituent bonds of the zeolite framework. Peaks appear at  $468.67 \text{ cm}^{-1}$ ,  $796.55 \text{ cm}^{-1}$ ,  $1097.4 \text{ cm}^{-1}$  and  $1629.74 \text{ cm}^{-1}$  appear in

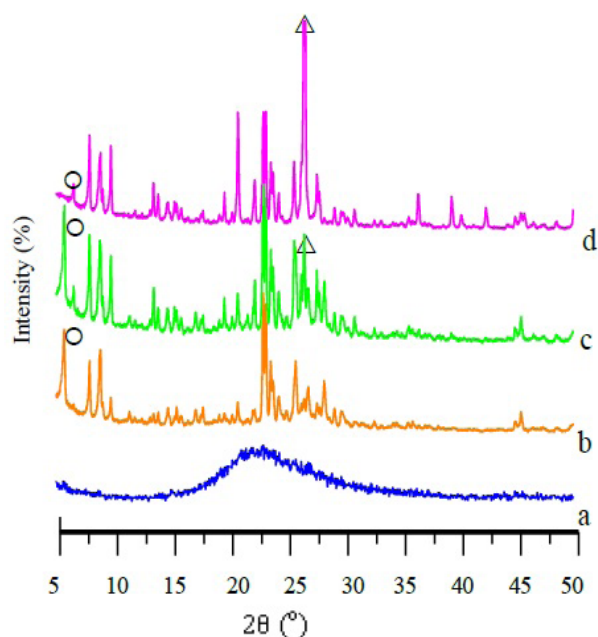


Fig. 1. XRD pattern of ZSM-5 synthesized at  $175^\circ\text{C}$  for a) 24 h; b) 36 h; c) 48 h; d) 72 h (○ - mordenite, △ - quartz).

Table 1. FTIR data referring to ZSM-5 synthesized.

Hydrothermal Time	Wavenumber (cm <sup>-1</sup> )					
	Bending vibration of Si-O and Al-O	Five membered ring	Symetric stretching vibration of Si-O-Si	Asymmetry stretching vibration of Si-O-Si	TO4 tetrahedron units	bending vibration of -OH
24 h	468.67	-	796.55	1097.42	-	1629.74
36 h	445.53	545.82	781.12	1080.06	1236.29	1629.74
48 h	457.10	545.82	781.12	1083.92	1238.21	1629.74

case of hydrothermal time of 24 h. Peaks appear at 445.53 cm<sup>-1</sup>, 545.82 cm<sup>-1</sup>, 781.12 cm<sup>-1</sup>, 1080.06 cm<sup>-1</sup>, 1236.29 cm<sup>-1</sup> and 1629.74 cm<sup>-1</sup> when the treatment is within 36 h. The peaks in the spectra recorded in case of hydrothermal time of 48 and 72 h are outlined at identical wave numbers, i.e. at 457.10 cm<sup>-1</sup>, 545.82 cm<sup>-1</sup>, 781.12 cm<sup>-1</sup>, 1083.92 cm<sup>-1</sup>, 1238.21 cm<sup>-1</sup> and 1629.74 cm<sup>-1</sup>. These results are similar to those reported by Cheng et al. [5]. The band at around 450 cm<sup>-1</sup> illustrated in Fig. 2 shows the bending vibration of Si-O and Al-O, the band at around 1100 cm<sup>-1</sup> is indicative of the asymmetry stretching vibration of Si-O-Si, that at around 800 cm<sup>-1</sup> is ascribed to the symmetric stretching vibration of Si-O-Si, while the band at around 550 cm<sup>-1</sup> is typical for the five membered ring of the pentasil structure. All FTIR data obtained is listed in Table 1. It is evident that no ZSM-5 is synthesized within hydrothermal time of 24 h, i.e. no bands at around 550 cm<sup>-1</sup> and 1200 cm<sup>-1</sup> characteristic of MFI structure are seen. Thus the FTIR data obtained support those of XRD analysis.

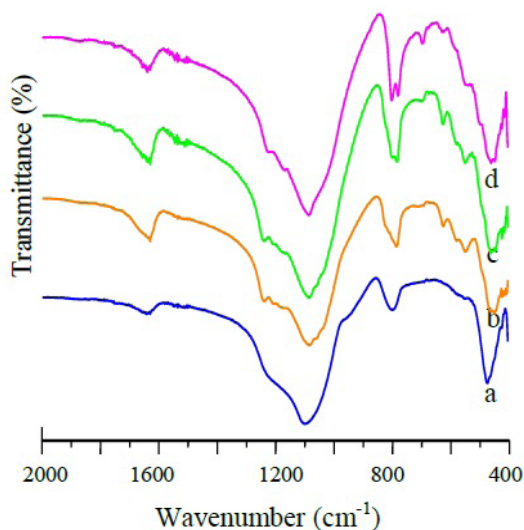


Fig. 2. FTIR spectra of ZSM-5 synthesized at 175°C for a) 24 h; b) 36 h; c) 48 h; and d) 72 h.

Desilication is a post-treatment of a zeolite aimed at a selective extraction of silicon atoms from the zeolite framework. It is worth adding that the the aluminum framework does not disrupt during this process because of the basicity provided by NaOH presence. In this research, the desilication is carried out using NaOH solution. Sodium hydroxide is a good desilicating agent when compared to organic bases such as tetrapropyl ammonium hydroxide (TPAOH) and tetrabutyl ammonium hydroxide (TBAOH) because the desilication process with their participation is considered less selective [20].

The XRD diffraction patterns in Fig. 3 show specific peaks at ca 7° - 9° and 23° - 25°. This provides the conclusion that the process conducted decreases the zeolite crystallinity but still preserves ZSM-5 MFI structure. The NaOH treatment affects crystal morphology through facilitation of mesopores formation. In fact the morphological changes of the zeolite crystals increase with NaOH concentration increase [21]. According to Groen et al. [22], the optimum conditions of desilication refer to 0.2 M NaOH, temperature of 338 K and duration of 30 min.

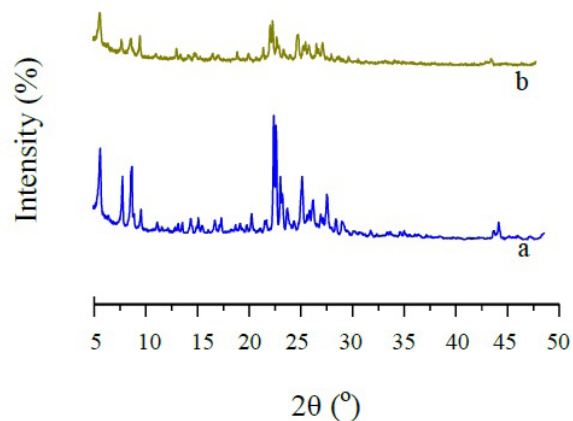


Fig. 3. XRD pattern of ZSM-5 a) prior to desilication and b) after desilication.

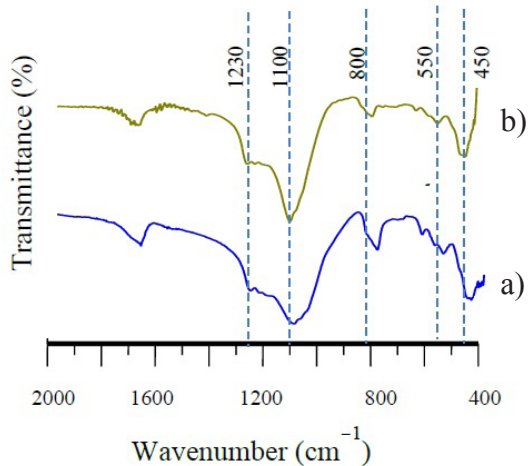


Fig. 4. FT-IR spectra of ZSM-5: a) prior to desilication and b) after desilication.

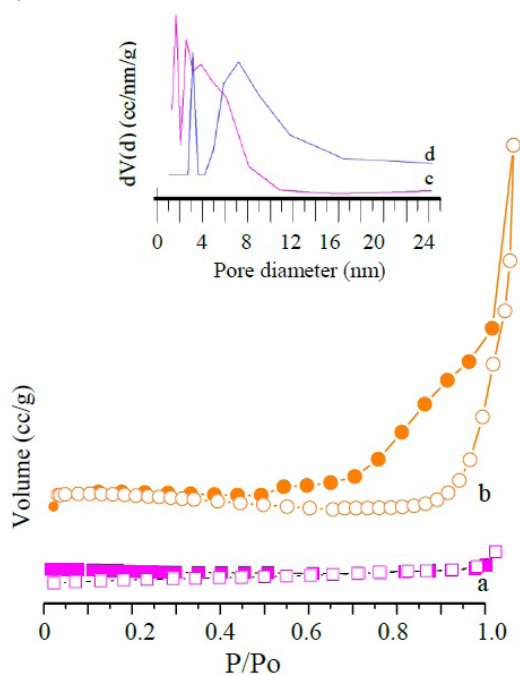


Fig. 5. Nitrogen adsorption/desorption isotherm of ZSM-5 a) prior to desilication and b) after desilication ; pore diameter curve: c) before desilication and d) after desilication).

The FTIR spectra are shown in Fig. 4. A similarity of the pattern is observed in Figs. 4a and 4b. This indicates that the framework structure is not changed during the desilication process.

The nitrogen adsorption/desorption curves of a sample of ZSM-5 prior to and after desilication are shown in Fig. 5. In Fig. 5 was also listed about curves of pore size distribution of ZSM-5 after desilication

process. It is evident that the curve of ZSM-5 prior desilication is of type I, which is typical for microporous solids. The insert in Fig. 5c shows that the pore diameter is ca 3.436 nm, while the surface area of the mesopores is ca 65.797 m<sup>2</sup>/g (69.44 % of total surface area). It is also evident that the pore volume is ca 0.232 cc/g, while the micropores surface area is ca 28.954 m<sup>2</sup>/g. Fig. 5b indicates that the curve of ZSM-5 after desilication corresponds to type IV. The latter is typical for mesoporous solids (Groen et al., 2005). The distribution of the pore diameter is between 3.8 nm and 8 nm (Fig. 5d), while the mesopores surface area is ca 42.752 m<sup>2</sup>/g (86.84 % of total surface area). The pore volume is 0.186 cc/g, while the micropores surface area is found equal to 6.475 m<sup>2</sup>/g. It is small but nevertheless it shows that ZSM-5 considered differs from that prior desilication.

## CONCLUSIONS

Hierarchical ZSM-5 can be synthesized without organic templates by the hydrothermal method at 175° C for 36 h. Quartz is evidenced as an impurity by the XRD diffraction pattern in case of a longer synthesis (72 h). The formation of ZSM-5 is also supported by the FT-IR spectra obtained. The formation of mesoporous ZSM-5 is proved by nitrogen absorption/ desorption and pore size distribution curves recorded.

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