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Direct conversion of ethylene to propylene (ETP) with SSZ-13 zeolites, converted from beta zeolites

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Highlights

- SSZ-13s, converted from BEA, were very competitive in ETP reaction.
- BEA zeolites were firstly converted into CHA (SSZ-13).
- Extra silica was essential for the conversion into SSZ when BEA was Al-rich
- Conversion of BEA into CHA showed the role of pore size rather than framework density.

1. Introduction

The direct conversion of ethylene to propylene (ETP) is interesting reaction considering the possible shortage of propylene (since ethane cracking and shale gas produce mainly ethylene rather than propylene) in near future. A few zeolites such as SAPO-34, ZSM-5 and UZM-35 were applied in ETP, and recently it has been suggested that SSZ-13s (with CHA structure) were very effective in the interesting reaction [1-3]. So far, the SSZ-13 has been synthesized mainly by two ways: direct synthesis (from silica and alumina) and conversion of zeolites (mainly Y). However, the methods should be improved not only for adequate performances but also for cost-effectiveness. In this work, the conversion of beta (BEA) zeolite into SSZ-13 has been studied for the first time. The obtained SSZ-13 was characterized thoroughly and applied in ETP to check the catalytic performances of the SSZ-13

2. Methods

The hydrothermal conversion of BEA-zeolites having different SARs was accomplished using a procedure similar to a reported method [3]. The obtained SSZ-13 zeolites were characterized with XRD, nitrogen adsorption, ^{27}Al NMR, NH_3-TPD, ICP and SEM. The ETP reaction was carried out at atmospheric pressure at 300-450 ºC with a conventional fixed-bed reactor made of a stainless-steel tube. The products and fed ethylene were analyzed by using GC.

3. Results and discussion

Fig. 1 shows the XRD patterns of obtained SSZ-13 zeolites. Fig. 1a shows that some sodium silicates are essential to produce SSZ-13 from BEA (with silica/alumina ratio or SAR of 15). Moreover, Fig. 1b illustrates that SSZ-13 can be synthesized from various BEA zeolites with wide range of SAR (10-80). Moreover, the syntheses of phase-pure SSZ-13s were confirmed by various techniques such as N_2-adsorption, ^{27}Al NMR and SEM etc. ETP results are summarized in Fig. 2. As shown in the figure, the performances of the SSZ-13 obtained in this study were very competitive against conventional SAPO-34.

4. Conclusions

Following conclusions can be drawn from the current study. First, the BEA zeolites can be successfully converted into CHA (SSZ-13) zeolite, and additional silica was essential for the conversion when aluminous BEA was applied. Second, based on the conversion, it could be confirmed that pore size rather than framework density is important in the synthesis of the zeolites. Third, the prepared SSZ-13 showed very competitive performances in the ETP, one of the important catalytic reactions.

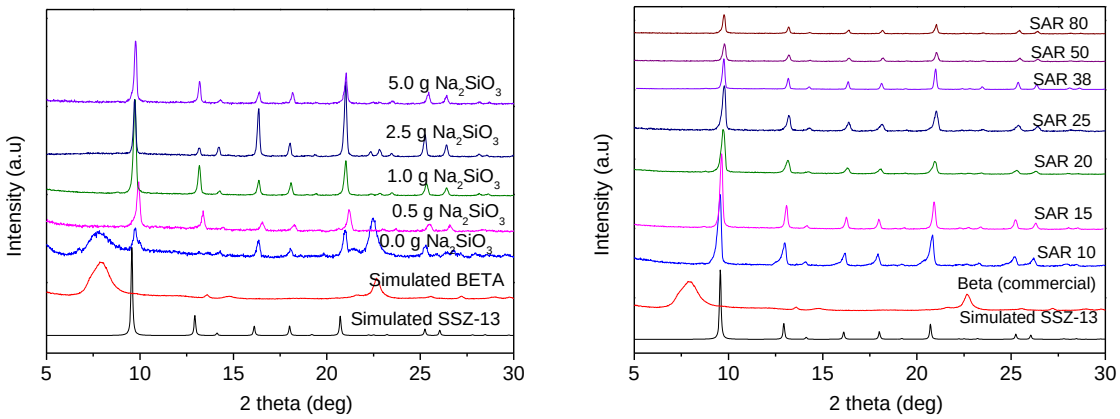

Figure 1. XRD patterns of synthesized SSZ-13 zeolites: (a) SSZ-13s were converted from BEA (SAR 15) with different amount of sodium silicate, and (b) SSZ-13s were obtained from various BEA (with different SARs).

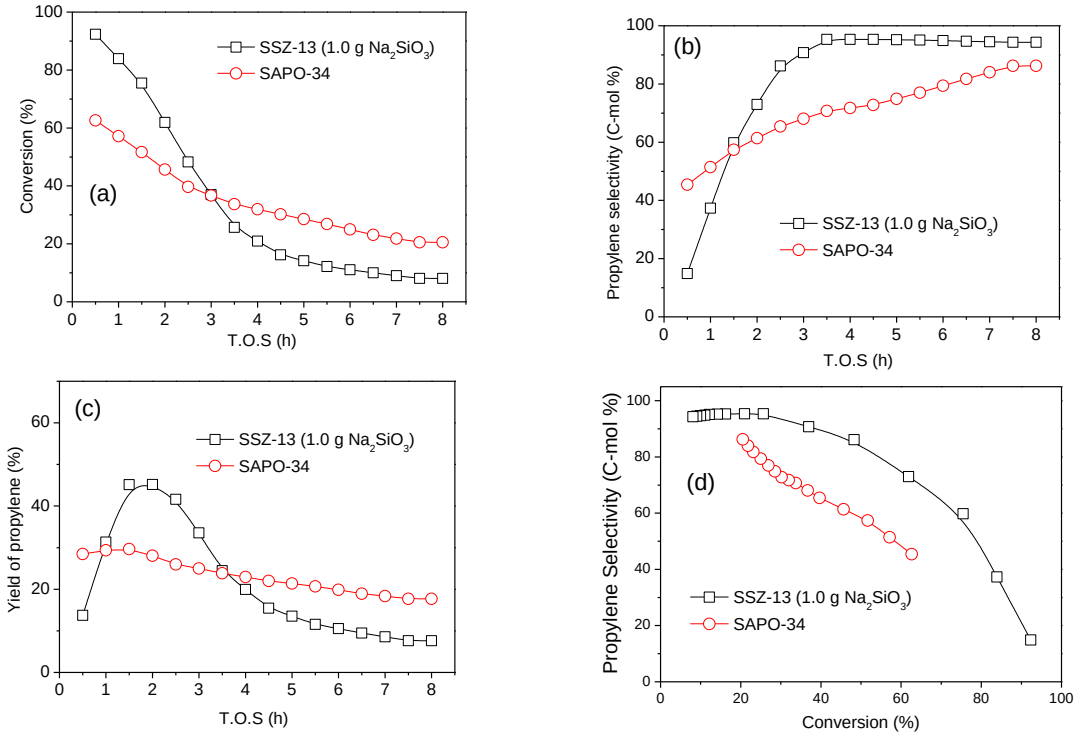

Figure 2. Effect of time-on-stream on the (a) ethylene conversion, (b) propylene selectivity, and (c) propylene yield; and (d) propylene selectivity with conversion from ETP reactions over SSZ-13 zeolite converted from zeolite BETA (SAR=15) in the presence of Na_2SiO_3. ETP results with conventional SAPO-34 were added for comparison.

References

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Keywords
Ethylene-to-propylene; Beta; SSZ-13; Zeolite conversion