Influence of silica source on the crystallization of KFI zeolite

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KFI zeolites are of significant interest for efficient gas-mixture separation due it is synthetic microporous zeolite with a main channel size of 3.9 × 3.9 Å that is comparable to the gas molecule diameters. In addition, KFI zeolite is promising to overcome the "trade-off" effect of working capacity and gas-mixture selectivity due to its suitable and confined pore spaces [1-3]. For example, KFI zeolite (Si/Al = 4.59) adsorbent prepared to investigate the separation of the $CO_2/CH_4/N_2$ gas mixtures revealed that KFI zeolite exhibits high CO_2/N_2 and CH_4/N_2 selectivity based on strong adsorption of CO_2 and CH_4. Furthermore, under the same conditions, the separation performance of KFI zeolite was superior to that of commercial adsorbent zeolites 5A and 13x [4]. Therefore, this study aims to evaluate KFI zeolite crystals prepared with different silica sources to produce thin membranes of KFI zeolite with thermal and mechanical stability, uniform, and high permselectivity for gas separation.

Conventional hydrothermal synthesis was employed for KFI zeolite preparation. A precursor gel with a molar ratio of $2.3K_2O$: $0.1Sr(NO_3)_2$: $1Al_2O_3$: $10SiO_2$: $160H_2O$ was transferred to a Teflon-lined stainless steel autoclave and statically heated in an oven at 150 °C for 230 h. Aluminum wire was used as a source of Al, colloidal silica (Ludox® AS-40, 40 wt.% or Ludox® AM-30, 30 wt.% suspension in H_2O), TEOS or silicic acid (SA) was the silica source. KOH (90%) and $Sr(NO_3)_2$ (99%) provided the cations Sr^{2+} and K^+ which are the structure-directing agents. OH^- ions were the mineralizing agent that controls the solubility and mobility of the framework atom sources in the synthesis gel to maintain the availability of the useful species at the level needed for nucleation and crystal growth of zeolite [5]. All reagents were obtained from Sigma-Aldrich.

Fig. 1 shows the crystallographic pattern of the samples. The synthesis using Ludox® AS-40 and Ludox® AM-30 present peaks referring to the KFI zeolite according to IZA online database in the region 2θ region equal to 6.96, 9.64, 15.02, 20.34, 21.52, and 26.06 corresponding to the (110), (200), (310), (411), (420), and (521) planes, respectively. However, for the sample with Ludox® AS-40 some peaks in the 2θ region equal to 11.0, 11.76, 20.08, 20.64, and 32.12 that are not present in the KFI crystallographic pattern indicating a mixture of other zeolites (L or W) in the synthesized material. Thus, the use of Ludox® AM-30 as a source of silica produced a KFI zeolite with fewer impurities. Regarding the sample prepared with TEOS and aluminum wire as a precursor, XRD patterns presented peak characteristics of zeolites KFI and L. Due to the difficulty in the dissolution of silicic acid, an amorphous phase was obtained for the use of this silica source.

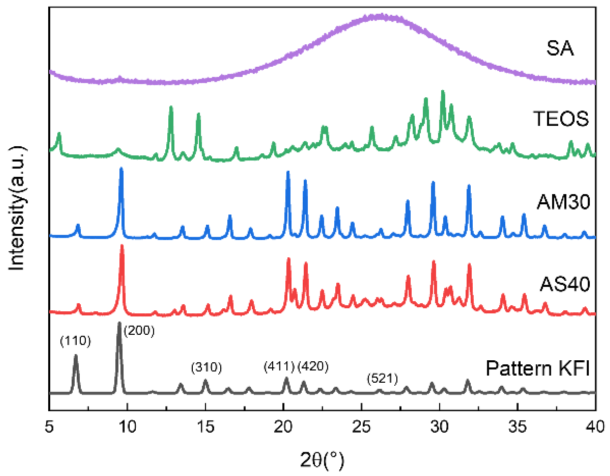

Fig. 1- XRD pattern of conventional KFI and samples prepared with different silica sources.

SEM micrographs of syntheses of KFI zeolite using Ludox® AS-40 and Ludox® AM-30 as silica sources are shown in Fig. 2(a) and 2(b), respectively. All images present a considerable amount of integrated growth of cubic-shaped particles characteristic of KFI zeolite. For zeolite prepared with Ludox® AS-40, small risks of impurities characteristic of zeolite W can be observed, while zeolite prepared with Ludox® AM-30 presents only cubic crystals, indicating that, as observed from the XRD analyses, the use of Ludox® AM-30 in the synthesis of KFI zeolite increase its purity. Moreover, SEM micrographs show that this zeolite crystals are smaller. The BET-specific surface area was verified by N_2 adsorption–

desorption isotherms. The results showed that with Ludox® AS-40 the zeolite exhibits a lower surface area (123.19 m^2 g^{-1}) than zeolite synthesized with Ludox® AM-30 (523.823 m^2 g^{-1}). It confirms that Ludox® AM-30 provided zeolite was characterized by a more complete pore structure, and a higher level of crystallinity and purity, consistent with the previous XRD and SEM analysis. Therefore, it is demonstrated that the silica source influences the purity and formation of KFI zeolite crystals. The KFI zeolite prepared can be used as seeds for the fabrication of uniform and thin membranes for gas separation.

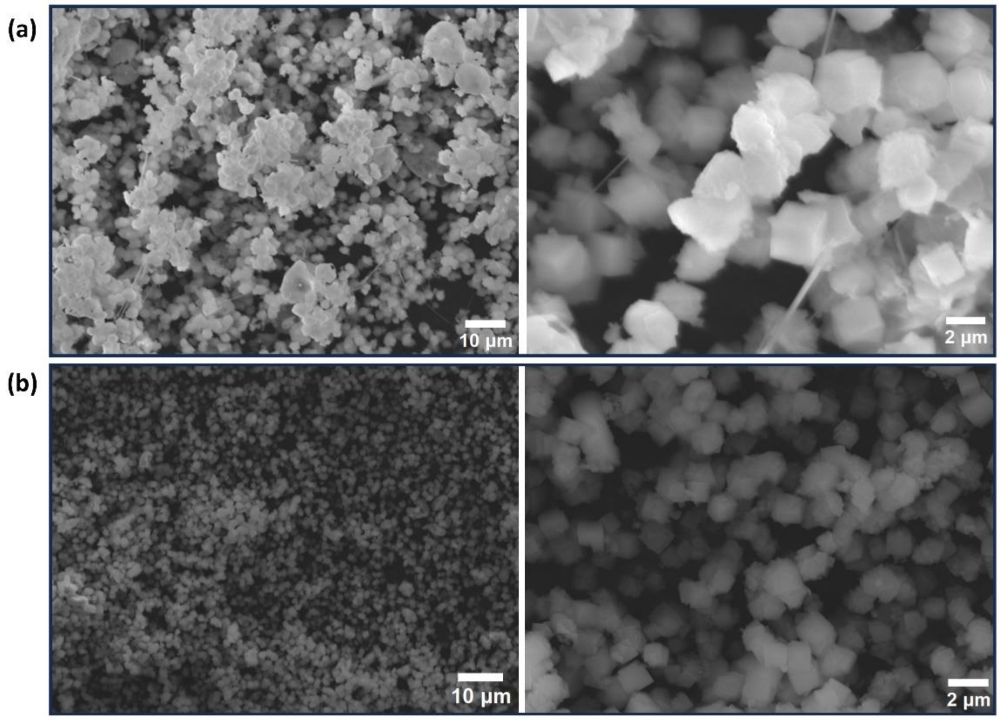

Fig.2- SEM micrographs of KFI zeolite synthesized using (a) Ludox® AS-40 and (b) Ludox® AM-30 as silica source.

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