

## Structure investigation of mordenite induced by molecular sieve using the Rietveld method

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### [Introduction]

There are a large number of natural and synthesis zeolite minerals that are generally composed of Si/AlO<sub>4</sub> three-dimensionally framework structure. All of them have a porous structure that can adsorb water molecules in their micropores. In addition, zeolite minerals have the highly adsorption and preservation properties generally called molecular sieve that accommodate a variety of cations and molecules in the micropore due to the negative electric charge caused by the substitution of Al<sub>3</sub><sup>+</sup> for Si<sub>4</sub><sup>+</sup>. The high cation exchange ability has been received great attention in many fields.

Mordenite [(Na<sub>2</sub>, Cs, K<sub>2</sub>)<sub>4</sub>(Al<sub>18</sub>Si<sub>40</sub>)O<sub>96</sub>·28H<sub>2</sub>O] (Martucci et al., 2003) is well known for its high selectivity for radioactive Cs and Sr. However, there are only few studies that addressed the crystal structural change of mordenite by Cs<sup>+</sup>-ion exchange molecular sieve. Hence, this study is aimed to clarify the relationship between molecular sieve and structural change of mordenite.

### [Experimental Methods]

The structural change of mordenite were investigated by the Rietveld refinement analyses based on the powder X-ray diffraction. measurements; CuK $\alpha$  ( $\lambda = 1.54056 \text{ \AA}$ ),  $5^\circ \leq 2\theta \leq 120^\circ$ , scanning speed =  $4^\circ/\text{min}$ , scanning width =  $0.02^\circ$ . Cs-exchanged mordenite were prepared from H-mordenite by ion exchange in CsCl solutions with various Cs concentration (0.010, 0.10, 1.0, 2.0, 4.0, 8.0, 10.0, 30.0g/l) . Cs-exchange experiments were executed by the shaking (32rpm) for 48hrs. The temperatures of solutions were maintained around 20°C.

### [Results and Discussion]

The pH value of the solutions was initially  $3.44 \pm 0.02$ , but it was decreased to  $2.70-3.09 \pm 0.02$  with addition of mordenite. The decreasing of pH value with the Cs concentration indicated that Cs<sup>+</sup>-ion was substituted for H<sup>+</sup>-ion within the micropore of mordenite structure. Consequently, the lattice parameter of mordenite was isotropically decreased with the Cs<sup>+</sup>-ion concentration. The unit cell volume of mordenite was slightly decreased from 2880 (12)  $\text{\AA}^3$  to 2836 (12)  $\text{\AA}^3$  (at most 44 (17)  $\text{\AA}^3$ ).

The molecular sieve of mordenite was characterized by the contraction of 12-membered ring channel (12MRc) and 8-membered ring channel (8MRc) where the Cs<sup>+</sup>-ion can be accommodated within the mordenite structure. The 12MRc with the largest pore size was appropriately contracted with the Cs content due to the electrostatic attraction between Cs<sup>+</sup>-ion and framework atoms. The contraction of 12MRc (at most 0.31(8)  $\text{\AA}$ ) especially acted toward the direction of b-axis. The high absorption efficiency of the molecular sieve is caused by the site preference of Cs<sup>+</sup>-ion into the 12MRc. The 8MRc with the smaller pore size than the 12MRc directly connected with the 12MRc. It behaves in the opposite way to the 12MRc. The 8MRc is strongly affected with the structural modification of 12MRc. Since the Cs<sup>+</sup>-ion are hardly incorporated into the 8MRc, the site occupancy of Cs<sup>+</sup>-ion in the 8MRc is always lower than that in 12MRc, which leads to the small contraction values of 8MRc.

Keywords: Mordenite, Molecular sieve, Rietveld refinement