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Pores in various kinds of zeolites by positron annihilation spectroscopy

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Zeolite is a porous material with regular tubular pores (0.4-0.8 nm diameter) and cavities with the basic unit of tetrahedral (SiO4)4- and (AlO4)5-. There are various crystal structures, and it is known that chemical properties such as catalytic activity and ion exchange species changewith different Si/Al ratios and crystal structures. In this study, we have measured the Ps behavior, such as lifetime and formation rate, and correlated them with the crystal structure and the Si/Al ratio.

Positron annihilation lifetime spectroscopy (PALS) was carried out on ZSM-5 and ferrierite, both having a 10-membered ring, as wel as beta and mordenite, both of which possess a 12-membered ring. Zeolite powder was molded into pellets having a diameter of 10 mm and a thickness of 2 mm. Subsequently, all samples underwent vacuum heating at 458 K for 2 hours in order to dehydrate them. Finally, a dehydrating agent and an oxygen scavenger were enclosed in a thermally sealed polyethylene bag together with each sample and the positron lifetimes were measured under these controlled atmosphere conditions. Each measured spectrum was fitted using four lifetime components. The 3rd component (of the order of ns) and 4th component (of the order of tens of ns) are due to the annihilation of o-Ps.

The PALS results for the zeolites except ZSM-5 showed that the pick-off lifetime of o-Ps without dehydration was well correlated with the diameter of the tubular pores (Fig.1). On the other hand, the dehydration process led to a longer o-Ps lifetime. The adsorbed water in the tubular pores hindered the diffusion of Ps, so that the o-Ps lifetime actually reflected the diameter of the pores with adsorbed water. We concluded that Ps can more easily diffuse in the cavities by dehydration and thus the o-Ps lifetime becomes longer.

An increase in Al3+ ions gives rise to an addition of polar groups in the tubular pores. The plot of the sum of I3 and I4 vs Si/Al ratio is shown in Fig. 2. The higher the Si/Al ratio, the larger is the Ps formation component. Furthermore, the dehydration led to an increase of the Ps formation intensity. This result was expected given that electrons and positrons tend to be captured by the adsorbed water, leading to a decrease of the Ps formation probability.

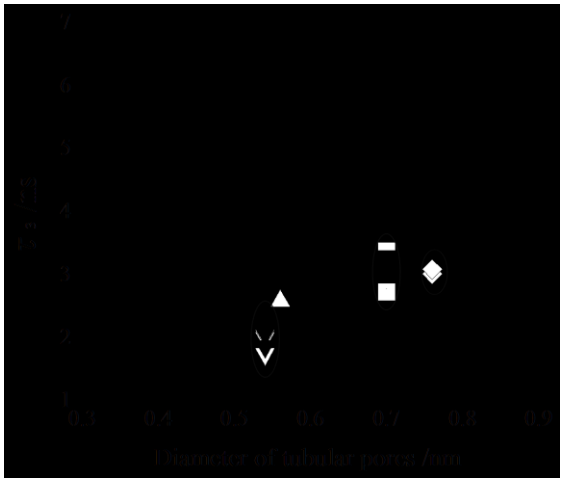

Fig. 1 Plot of τ_3 vs diameter of tubular pores.

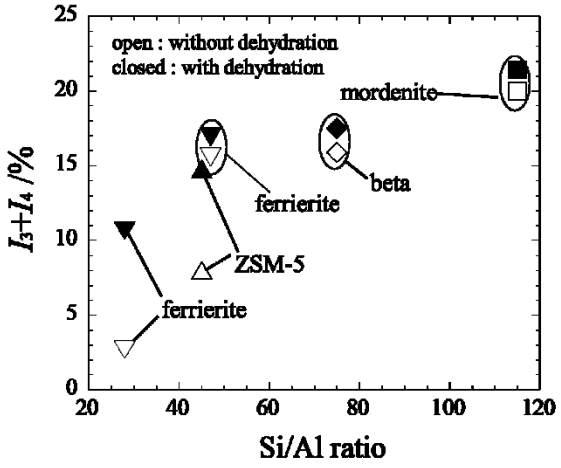

Fig. 2 Plot of I_3+I_4 vs Si/Al ratio.