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Peculiar Diffusion Mechanisms within Micropores of Zeolite Catalysts

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Diffusion mechanisms of hydrocarbons within zeolites, especially MFI-type zeolite, were investigated by discriminating intracrystalline diffusivity from effective diffusivity. Intracrystalline diffusivity directly represents the mobility of molecules within pores. Effective diffusivity is obtained by multiplying the intracrystalline diffusivity by a partition factor given by the ration of the concentrations of molecules in zeolite crystals to that in gas phase. Intracrystalline diffusivity was the main subject of this study. Diffusion within MFI-type zeolite is dominated by the following mechanisms: (1) configurational diffusion, (2) resistance to mass transfer at pore mouths, (3) adsorption-controlled diffusion and (4) co-existing molecules with slow diffusing molecules. Intracrystalline diffusivity in the adsorption process is lower than in desorption process for (1) and (2) if the minimum molecular size is larger than the pore diameter, such as *ortho*- and *meta*-xylenes. The resistance to mass transfer at pore mouths becomes dominant in the adsorption process. This tendency is also observed for paraffins. Model equations were proposed for evaluating intracrystalline diffusivity based on the molecular size, molecular weight and pore diameter. Overall intracrystalline diffusivity for (3) is correlated with the configurational diffusion and the trapping effect on acid sites. This effect disturbs the mass transfer, especially at temperatures below 573 K, for aromatics. Similar effects are observed for lighter paraffins and olefins within MFI-type zeolites with metal cations. Intracrystalline diffusivities for (4) for silicalite-1 in a multicomponent system were measured using a new desorption under reduced pressure method. The diffusivity of the

slow component in the multicomponent system agreed well for that of a single component. However, the diffusivity of the fast component was largely decreased by co-existing slow molecules. A random walk simulation and an empirical equation could explain this tendency.

Effective diffusivity was calculated from the intracrystalline diffusivity and the partition factor, which was obtained from the adsorption isotherm. The partition factor suggested a marked condensation effect for MFI-type zeolites.

Intracrystalline and effective diffusivities for beta- and Y-type zeolites, and mordenite were also investigated.

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