Effects of Mesoporosity and Crystallinity on Deactivation of ZSM-5 in Propanal Conversion [1]

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Zeolites are important solid acid catalysts and generally deactivate due to coke formation. It has been reported that 'hierarchical zeolites' with secondary mesoporosity can show a significantly increased catalyst lifetime because of retarded coke formation. Although the generation of secondary mesoporosity has been considered as one of the most effective strategies to retard deactivation, in certain cases, even faster deactivation after the mesopore generation has also been reported. We believed that such exceptions may be originated from other important structural factors of a zeolite, in particular, the crystallinity of a zeolite.

In the present work, we studied the effect between the secondary mesoporosity and zeolite crystallinity on the deactivation of ZSM-5 catalysts. Varied secondary mesoporosities were introduced into a commercial ZSM-5 by desilication using solutions with different NaOH concentrations. The results showed that the mesoporosity gradually increased with increasing concentration of NaOH, while the microporosity decreased. This means that the desilication for mesopore generation is a destructive technique that sacrifices the zeolite crystallinity. In propanal conversion which is known as a representative acid catalyzed reaction, ZSM-5 showed a longer catalyst lifetime as the external surface area increased (or as the zeolite framework thickness decreased) in the mild desilication regime. The enhanced lifetime could be attributed to the faster diffusion of coke precursors out of the zeolite structure. However, when the zeolite crystallinity of ZSM-5 was decreased too much from severe alkaline treatments, deactivation of the catalyst became again faster. This means that the unique 10-membered microporous channel of ZSM-5 is also important in suppressing the coke formation. This is in line with earlier reports showing that coke formation itself is a shape-selective reaction and thus inhibited in the constrained space of the 10-membered micropore channels of ZSM-5. The present results imply that the generation of large mesoporosity (enhancing the diffusion of coke precursors) while keeping the zeolite crystallinity intact (suppressing coke formation by shape-selectivity) is desirable for designing a zeolite catalyst with an enhanced catalyst lifetime.

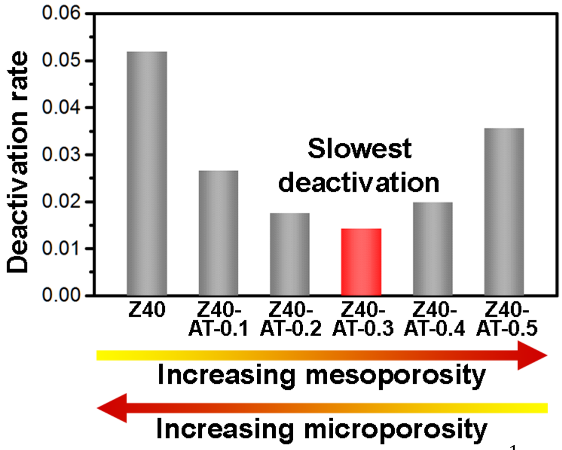

Fig. 1. Deactivation rate ($g_{cat.}$ g_{react}^{-1}) for alkaline treated ZSM-5 catalysts.

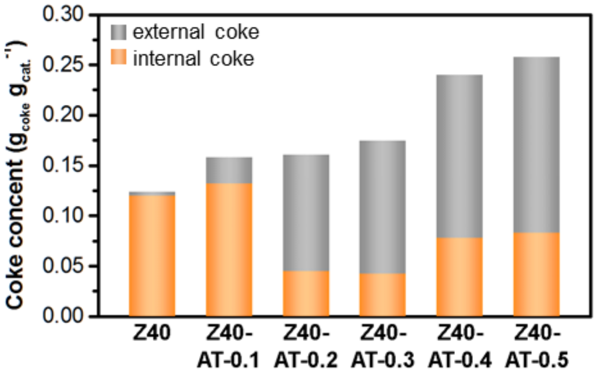

Fig. 2. Coke content for alkaline treated ZSM-5 catalysts

REFERENCES

[1] Manuscript under revision in *Microporous and Mesoporous Materials*