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NOTES

Coke Shell Profiles in the Fouling of H-Mordenite

We have recently reported on the relationship between coke content, activity, and intraparticle diffusivity resulting from cumene cracking on a commercial H-mordenite (*1*). Effective diffusivity decreased by over a factor of 2 during coking, and the development of a heavily coked outer shell was observed from SEM images of particle cross sections. It was postulated that the change in catalyst activity subsequent to the initial shell formation was strongly influenced by the rate of transport through the shell itself. Herein are reported some additional experimental observations in support of this view.

Figure 1 shows scans taken across the diameter of four samples coked for differing periods of time at 260°C and 0.33 W/hr/W space velocity. Given are the relative values of induced current off the sample for scans at 20 KV incident energy using a JEOL 50A scanning microscope operated in the AEI mode. The induced current is a function of coke content, though perhaps not a linear one, and the curves shown are extracted from the baseline of a noisy signal (due to sample surface roughness), so they are not quantitative. However, there is clear indication of where the coke is, and is not, in the interior of the particle.

The striking thing about these profiles is their fixed position. The ultimate depth of penetration of coke into the particle is attained after a very short time on stream, and is undoubtedly determined by the initial effective diffusivity. Subsequent coke deposition occurs entirely within this outer shell and an uncoked central core, apparently never contacted by reactant remains intact for the duration of the experiment. No analysis of intraparticle diffusion and coking that has appeared in the literature (*2–5*) would predict the establishment of such a spatially fixed outer shell of coke deposit.

It is possible that the time scale in the experiments was too short to show this behavior; however, examination of particles on stream for as long as 6 hr still clearly indicates the uncoked central core. Weight change of coke on catalyst between 3 and 6 hr was only 0.2% of a total of 3.4%, so this possibility seems unlikely.

These results suggest some type of compensation between decreasing activity and decreasing effective diffusivity that limits utilization of the total intraparticle catalytic surface. Further investigation of this is under way.

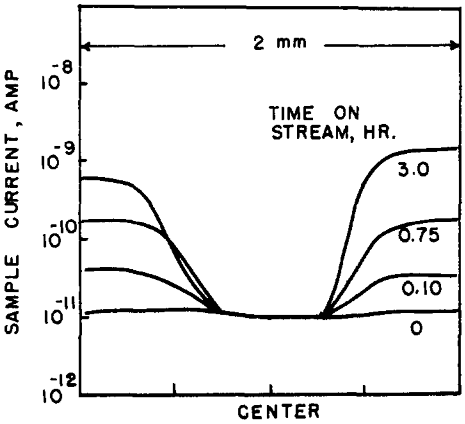

FIG. 1. Sample current vs radial position at differing extent of coke deposition. Norton Zeolon 900 H; 2 mm cylindrical cross section.

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