ZSM5 catalyst onto stainless steel monolith for environmental applications

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Introduction

The use of structured zeolitic materials for environmental applications has increased considerably. The advantage of the *in-situ* synthesis [1] to structure zeolites is that a stronger adhesion to the support can be achieved. However, the accessibility to the active sites within the zeolite framework is also important. The secondary synthesis can help in obtaining preferential orientations which favor the catalytic performance [2].

For some applications, metallic substrates are preferred due to their higher mechanical resistance and thermal conductivity and the possibility of allow higher cell densities with low pressure drop [3].

For these reasons we have produced and characterized a ZSM5 structured catalysts in the form of a coatings onto FeCrAl alloy supports. The catalytic performance was evaluated in the selective reduction of NO_x with methane by exchanging In as active material.

Materials and Methods

Supports were calcined at 900 ºC for a superficial Al_2O_3 segregation. Metallic monoliths were prepared by rolling strips. After that, the samples were seeded with nanometric ZSM5 crystals and then subjected to a hydrothermal treatment for 24 h at 180 ºC [4].

A synthesized Na-ZSM5/monolith was first exchanged with ammonium and then was loaded with In with a solution of $In(NO_3)_3$ through an exchange procedure that to develop the $(InO)^+$ active species in the zeolite framework [5].

The samples were characterized by Scanning Electron Microscopy (SEM), Electron Probe Micro Analysis (EPMA), X-ray Diffraction (XRD) and NO Temperature-Programmed-Desorption (NO-TPD). The catalytic evaluation was conducted at $[NO] = [CH_4] = 1000$ ppm; $[O_2] = 10\%$; Q/W = 500 $cm^3.min^{-1}.g\ zeolita^{-1}$.

Results and Discussion

Catalyst characterization

After the secondary synthesis a weight increase was observed due to the ZSM5 crystals onto the support. The XRD analysis shows a highly crystalline ZSM5 coating and the crystals in the coating are highly oriented with the c axis perpendicular to the support surface.

The SEM images confirm a total coverage of crystals over the support. The film thickness was between 3 and 10 µm with highly intergrowth and columnar microstructure. The c faces planes point outward the support surface. The coating was enriched in Al due to its migration from the support surface to the zeolite growth. The EPMA analysis also shows some Cr and Fe in the zeolite.
The TPD-NO shows one NO_2 signal located in one peak at low temperature. The species associated could be the InO^+ interchanged in the zeolite framework.

Catalytic performance

The In-ZSM5/FeCrAl monolith was evaluated in the SCR of NOx with methane in oxygen excess. The NOx to N_2 conversion reach 75 % at 450 ºC and the methane conversion is not total at 600 ºC. Also in the exhaust gases was detected CO. The NOx to N_2 conversion is lower compared with a powder evaluated in similar conditions (86%). This characteristic is opposite at the performance that we found in an In-Mor/FeCrAl monolith [2].

Significance

The obtained structured catalyst is constituted by a continuous and intergrowth In-ZSM5 coating onto FeCrAl substrate with c axis orientation. It is a very active catalyst in NOx SCR with methane and the lower conversion compared with the powder could be due to a disadvantageous spatial orientation of the crystals. Despite the In-ZSM5/FeCrAl monolith is a good structured catalyst with a highly adherent coating over a support with high thermal conductivity and mechanical resistance that could has advantages over other structured catalyst in environmental applications.

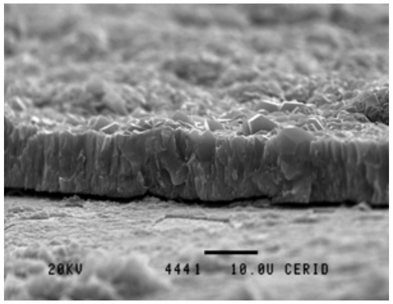

Figure 1. SEM image of a transversal cut of the ZSM5 coating onto metallic support after three hydrothermal treatments.

References

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