

Effect of the calcination temperature on the performance of a CeMoOx catalyst in the selective catalytic reduction of NOx with ammonia

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Introduction

The selective catalytic reduction (SCR) of NOx with NH₃ is considered to be the most efficient technology for reducing NOx emission in the presence of excess oxygen [1]. Although V₂O₅-WO₃(MoO₃)/TiO₂ has been widely employed as a SCR catalyst commercially, some serious problems still remain, i.e. the high activity of SO₂ oxidation, the toxicity of V₂O₅ [2]. Recently, Peng *et al.* [3] reported a CeMoOx catalyst with excellent SCR performance. For the practical use, the sudden change in temperature of the catalyst brick may happen and may lead to the structural and performance change of the catalyst [4]. Therefore, the examination of the effect of the calcination temperature on the catalyst is worthwhile.

Materials and Methods

CeOx-MoOx catalysts were prepared by the urea co-precipitation method. The precursors were calcinated at different temperatures for 4 h in air. Finally, the samples were crushed and sieved to 40 - 60 mesh.

Activity measurement: 500 ppm NO, 500 ppm NH₃, 5% O₂ and N₂ as the balance gas. The total flow rate is 500 ml/min and gas hourly space velocity (GHSV) = 50000 h⁻¹.

Results and Discussion

As shown in Fig. 1, compared to the CeMoOx-400 sample, CeMoOx-500 shows an activity with a slightly lower NOx conversion below 200 °C, but a higher NOx conversion above 350 °C. However, further increase of the calcination temperature leads to the obvious drop of NOx conversion. High N₂ selectivity, higher than 90% at 400 °C, was achieved with the samples calcined at 400 and 500 °C. In contrast, much lower N₂ selectivity, 73% and 70% at 400 °C, was measured with the other two catalysts.

The spectrum of XPS of O1s is fitted into three overlapped peaks. The bands at 529.3-529.4 eV is attributed to the lattice oxygen (O_C). The two shoulder peaks are assigned to the surface chemisorbed oxygen (O_B), and the chemisorbed water as contaminants at the surface (O_A), respectively. The surface chemisorbed oxygen was reported to be more active in SCR reaction because its mobility is higher than that of the lattice oxygen [3]. As shown in Table. 1, with the increase of the calcination temperature, O_B ratio decreases. Furthermore, compared to the O_B peaks of CeMoOx-400 and CeMoOx-500, the O_B peaks of CeMoOx-600 and CeMoOx-700 samples shifts to a lower binding energy at 530.6 eV. This shift suggests a stronger interaction between the surface chemisorbed oxygen and metal atoms, lowering the surface oxygen mobility and SCR activity as a result. Furthermore, the increase of calcination temperature leads to the enrichment of MoOx on the surface of catalyst.

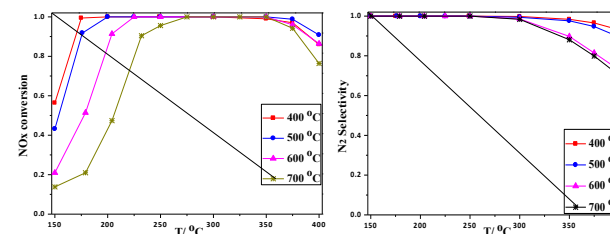


Fig.1 NH₃-SCR performance on Ce-Mo catalyst calcinated at different temperatures.

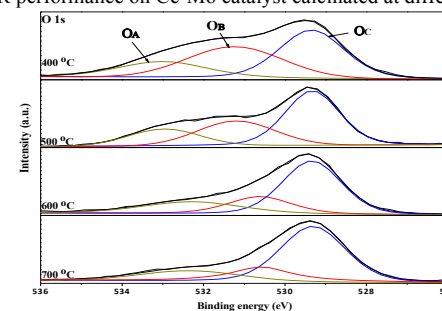


Fig. 2 XPS spectra of O 1s for the CeMoOx catalysts calcinated at different temperatures.

Table. 1 XPS results of the CeMoOx catalysts calcinated at different temperatures

Sample	Binding energy of oxygen (ev)			O _B ratio ^a (%)	Mo/Ce atomic ratio
	O _A	O _B	O _C		
400 °C	533.0	531.2	529.3	36.9	0.18
500 °C	533.0	531.2	529.3	30.0	0.23
600 °C	532.3	530.6	529.4	21.4	0.32
700 °C	532.3	530.6	529.3	19.0	0.42

^a O_B ratio = O_B/(O_A+O_B+O_C) ratio

Significance

CeMoOx-400 and CeMoOx-500 samples show excellent SCR performance, with more than 90% NOx conversion and N₂ selectivity between 175-400 °C. However, further increase of the calcination temperature to 600 and 700 °C leads to the drop of SCR performance. The low ratio and low mobility of the surface chemisorbed oxygen as well as the enrichment of MoOx contribute to the poor redox ability of the samples calcinated at high temperatures.

References

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