Novel LaMnNi Mixed Metal Oxides catalysts for Selective Catalytic Oxidation of NO

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Introduction

Selective catalytic oxidation of NO (SCO) is a significant step for fast SCR process and denitrification technology combined with liquid phase absorption. It obtained more and more attention in recent decades. Due to its excellent catalytic properties, some noble metals (such as Pt catalysts) were applied to oxidation and remove NO from automobile exhausted gas with SCR progress. However, the high price limits its promotion and use [1]. In recent studies, some non-noble metal oxides such as Mn, Co, Fe, and Cu supported on TiO_2 were investigated extensively in NO oxidation because of their low cost, high catalytic activity, and stability [2]. Whereas, these catalysts might be poisoned easily by compositions existing in flue gas, such as water vapor and SO_2.

In this work, a series of $LaMnNiO_x$ composite oxides catalysts were synthesized to improve the low temperature catalytic oxidation of NO. Acid treatment were used in the process of sample preparation to control the crystal structure of catalysts. Means of XRD, BET, H_2-TPR and O_2-TPD were applied to characterize these catalysts.

Materials and Methods

The different $LaMn_{0.8}B_{0.2}O_3$ (denoted as LaMnB, B= Ni, Cu and Fe), $LaMnO_3$ and Mn–Ni–O_x catalysts were prepared by a traditional co-precipitation method. A modification method was used after the previous catalysts were selected. To investigate the performance of the catalysts, the samples were characterized by means of SEM, TEM, BET, H_2-TPR, XPS and O_2-TPD.

Results and Discussion

The NO oxidation activity was tested over the prepared metal oxides catalysts. $LaMn_{0.8}Ni_{0.2}O_3$ treated by selective-removal method exhibited higher activity for NO oxidation to NO_2 and 83% efficiency could be achieved at 250 °C with a high GHSV of 170,000 h^{-1}. In addition, the NO conversion was also increased over acid treated catalysts (denoted as LMN (H)). In addition, it can keep a superior performance in the presence of H_2O on NO oxidation. X-ray diffraction (XRD) showed that new crystal structure of γ-MnO_2 appeared on the surface of the sample after treatment. In addition, the bigger BET surface area and the better redox ability showed in H_2-temperature programmed desorption (H_2-TPR) lead to a superior NO conversion in the test window. There are other characterization methods were used to explain the higher performance of the sample, such as oxygen temperature desorption (O_2-TPD), X-ray photoelectron spectroscopy (XPS) and NO + O_2-temperature programed desorption (TPD), etc.

The catalytic activity of the sample were also related to the ratio of Mn^{4+}/Mn^{3+} and Ni^{3+}/Ni^{2+}, oxygen species and the NO adsorption-desorption.

To investigate the effect of H_2O on NO adsorption, NO and O_2 were also adsorbed in the presence of H_2O. The adsorption amount of NO_2 is larger than that of NO over LMN-H sample in NO+O_2-TPD results. It indicated that LMN-H performed a superior NO adsorption activity corresponding to the results of NO + O_2-TPD and FTIR results.

Significance

By treating in acid solution, the NO oxidation activity and H_2O resistance was enhanced over LaMnNi composite oxides catalysts. It showed that the increased surface area, enhanced redox property and NO adsorption capacity might contribute to the better catalytic performance. The catalytic activity of the sample were also related to the ratio of Mn^{4+}/Mn^{3+} and Ni^{3+}/Ni^{2+}, oxygen species and the NO adsorption-desorption.

References

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