

The NiAl mixed oxides: The relation between basicity and SO₂ removal capacity

Sulphur dioxide (SO₂) is a pollutant gas produced by various human activities such as the operation of industrial boilers and the burning of fossil fuel at power plants. Stringent environmental regulations limiting atmospheric SO_x emissions encourage the research of more efficient ways to reduce them. Many new strategies have been proposed. Of the above, the method receiving most attention has been the sorption or reaction of SO₂ with dry regenerable solid sorbents. Moreover, due to the strong acid character of SO₂, basic oxides have been proposed to trap these molecules. In recent years, basic mixed oxides obtained from hydrotalcite-like (HT) compounds have shown high efficiency in reducing these SO_x emissions from several sources. HTs materials could be prepared by different methods. Among them, the urea hydrolysis method facilitates the catalyst that maintains high specific area, controllable pore system and good dispersion of active phase compared to the traditional coprecipitation method. Thus, in this work, a series of NiAI mixed oxides with different Ni/AI atomic ratios derived from the hydrotalcite-like compounds were prepared by the urea hydrolysis method. The prepared samples were characterized by means of TGA, XRD, SEM and N₂ adsorption/desorption isotherm techniques and evaluated as potential SO₂ sorbent using a volumetric method. Their adsorption performance was found to be closely correlated with their basicity. Furthermore, we focus on the relationship between the basicity of NiAI mixed oxides and their SO₂ removal catalytic ability.

The use of the urea hydrolysis method for the preparation of NiAl mixed oxides derived from hydrotalcite compounds leads to smaller homogeneous particle aggregates with flower-like morphology. These materials all exhibit certain adsorptive ability of SO_2 . The adsorption equilibrium isotherm of SO_2 was analyzed using a volumetric method at 298 K. The method is based on the mole balance of SO_2 gas in a closed system (Figure 1).

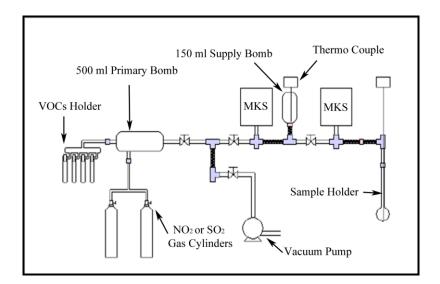


Figure 1. Schematic of SO₂ adsorption isotherm system

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Their adsorption performance has been found to be closely correlated with their basicity and the active oxygen species on the surface (Figure 2). The NiAlO-3 sample with the Ni/Al molar ratio of 3 shows the best adsorption performance of SO_2 , owing to the strongest basicity and most active oxygen species on the surface. Furthermore, the small pore opening in the quasimicropore region (4.3 nm) also contributes to the high activity of the NiAlO-3 sample in the adsorption of SO_2 .

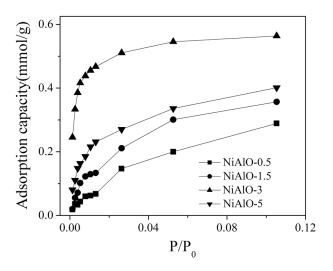


Figure 2. Adsorption equilibrium isotherms of SO₂ in NiAIO samples with different Ni/AI ratios calcined at 773 K.

The CO₂-TPD experiments were carried out on a sample of 0.2 g under Ar (45 cm³/min). Prior to CO₂ adsorption at 473 K, all samples were pre-treated at 773 K for 1 h in a flow of Ar. Once the physically adsorbed CO₂ was purged off, the CO₂-TPD experiments were started from 303 K to 1173 K with a heating rate of 10 K/min under Ar flow (50 ml/min). A mass spectrometer (Hiden HPR-20 QIC) was used for on-line monitoring of the CO₂-TPD effluent gas (Figure 3)

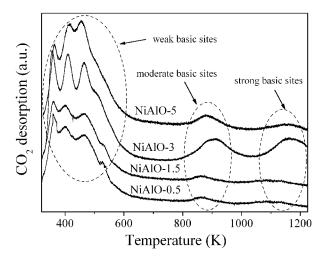


Figure 3. CO₂ -TPD profiles of the NiAIO samples with different Ni/AI ratios calcined at 773 K.

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Project Summary by:

Xinyong Li Key Laboratory of Industrial Ecology and Environmental Engineering and State Key Laboratory of Fine Chemical, School of Environmental Sciences and Technology Dalian University of Technology Dalian 116024 China



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