

## The Adsorption of NO<sub>x</sub> on Magnesium Aluminium Hydrotalcite

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**Abstract:** Magnesium aluminium hydrotalcite (Mg-Al-HT) with molar ratio of Mg-to-Al of 3 to 1 was prepared and characterized by X-ray diffraction (XRD) and infrared spectra (IR). The performances of Mg-Al-HT for the adsorption and desorption of NO<sub>x</sub> were studied. The results indicated that the adsorption capacity of the hydrotalcite for NO<sub>x</sub> was 1398.2 mg/g, and it was higher than the acticarbon's. The adsorption capacities depended on adsorption time and temperature. Mg-Al-HT could be regenerated by thermal decomposition, and the adsorption efficiency had not changed markedly after three cycles.

**Keywords:** Magnesium aluminium hydrotalcite, NO<sub>x</sub>, adsorption.

Hydrotalcite has been widely used in environmental protection, especially in the catalytic adsorption of nitric oxides<sup>1-4</sup>. Most of nitric plants adsorb nitric oxides in the liquid, such as low concentration solution of nitric acid or the alkali liquor, the efficiency was quite low. Molecular sieve and acticarbon have been used to adsorb nitric oxides successfully. By using molecular sieve to treat nitric acid tail gas, 2.5% yielding nitric acid could be reclaimed from the tail gas<sup>5</sup>, but the adsorption capacity of molecular sieve and acticarbon for high concentration of nitric oxides is limited, and the regeneration methodology of acticarbon is very complex. The acticarbon will autoignite at 573 K, which is very unfavorable to adsorption and reconstitution. In this paper, we used Mg-Al-HT to adsorb nitric oxides, and the adsorption and desorption of NO<sub>x</sub> on Mg-Al-HT were studied.

### Experimental

#### *Preparation of Mg-Al-HT*

A mixture of Mg(NO<sub>3</sub>)<sub>2</sub>•6H<sub>2</sub>O and Al(NO<sub>3</sub>)<sub>3</sub>•9H<sub>2</sub>O (molar ratio of Mg-to-Al was 3) was added slowly (1 droplet/s) into the solution of NaOH and Na<sub>2</sub>CO<sub>3</sub> at 303 K in a beaker under vigorous stirring, and the pH was kept between 9 and 10. The mixture was continuously stirred for about 30 min, and heated to 338 K, then kept at this temperature for 18 h. The precipitate was washed with deionized water until pH = 7, dried at 353 K for 12 h, calcined at 773 K for 5 h, and then cooled to room temperature.

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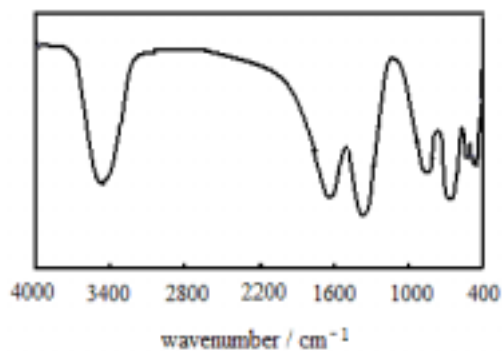
*Adsorption and desorption of Mg-Al-HT*

Mg-Al-HT was loaded in a vacuum container (0.29 L). Nitric oxides were injected into the container and shaken. The concentration of  $\text{NO}_x$  was measured by N-(1-naphthyl) ethylenediamine hydrochloric acid spectrophotometer. After saturation of adsorption, the hydrotalcite was reclaimed washing, drying at 353 K, calcination at 773 K for 5 h.

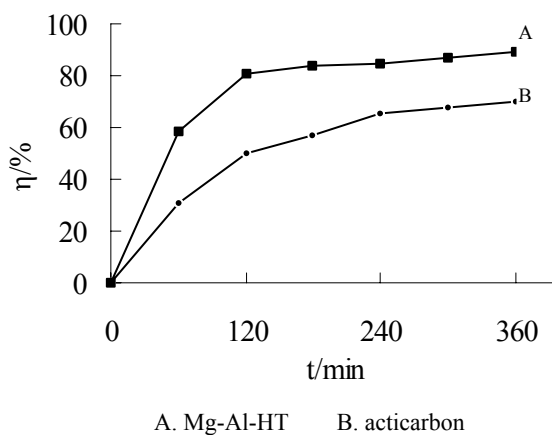
**Results and Discussion**

The result of X-ray diffraction pattern of Mg-Al-HT was shown that it was just qualitative hydrotalcite<sup>6</sup>. There were three characteristic diffracted peaks at  $2\theta = 11.22, 22.66, 34.74$ , respectively. The results indicated that the product was monophasic with good crystallization, and layer spacings 0.7756 nm, 0.3921 nm and 0.2617 nm, respectively.

**Figure 1** IR spectrum of the sample with the molar ratio of Mg-to-Al of 3 before calcination



**Figure 2** Effect of adsorption time on adsorption rate at 294 K



FT-IR spectra of the product was shown in **Figure 1**. The absorption peaks at 3462 cm<sup>-1</sup> and 1650 cm<sup>-1</sup> had been assigned to the bending and expanding vibrations of water, indicating some water on the surface of the particle or in the interlayer of crystal lattice. The peak at 1383 cm<sup>-1</sup> was assigned to C-O expanding vibration of CO<sub>3</sub><sup>2-</sup>, which was compared with the adsorption peak of CO<sub>3</sub><sup>2-</sup> (1430 cm<sup>-1</sup>), the peak of C-O shifted to lower wavenumber, this showed that CO<sub>3</sub><sup>2-</sup> were not really free in interlayer, hydrogen bonds must exist between CO<sub>3</sub><sup>2-</sup> and water. Two weaker absorption peaks at 840 cm<sup>-1</sup> and 653 cm<sup>-1</sup> were assigned to the face shifting angle vibration and non-plain face vibration of CO<sub>3</sub><sup>2-</sup>. The absorption peaks at 450 cm<sup>-1</sup> and 500 cm<sup>-1</sup> were assigned to crystal lattice vibration of Mg<sup>2+</sup> and Al<sup>3+</sup>.

The effect of adsorption time on the adsorption capacities of Mg-Al-HT and powder acticarbon was investigated. **Figure 2** showed that the adsorption rate of NO<sub>x</sub> by Mg-Al-HT was very rapid. In the initial 60 min, the adsorption capacity reached 58.7%, after 6 h, the adsorption capacity was 89.1%. Within the same adsorption time, the adsorption capacity of Mg-Al-HT was higher than the acticarbon's. Perhaps, due to that the surface of the hydrotalcite was alkaline. The OH<sup>-</sup> and CO<sub>3</sub><sup>2-</sup> in Mg-Al-HT were removed after calcination at 773 K, and the number of microporous increased significantly, the metal particles were dispersed, and many pore channels and bigger specific surface area appeared, but its surface (BET was less than 300 m<sup>2</sup>/g)<sup>7, 8</sup> was lower than that of the acticarbon. Therefore, the properties of Mg-Al-HT remain to need the further study.

The concentration of NO<sub>x</sub> was measured at different adsorption temperatures after adsorption of NO<sub>x</sub> for 6 h. The results showed that the adsorption capacity of Mg-Al-HT was highly dependent upon the adsorption temperature. The adsorption capacity decreased with the increase of temperature.

After adsorption of NO<sub>x</sub>, some of Mg-Al-HT converted to nitrate. The nitrate decomposed easily on heating. For example, the aluminium nitrate decomposed at 473 K, magnesium nitrate decomposed at about 673 K, so they can be regenerated by the thermal decomposition. (aqua destillata) The Mg-Al-HT with saturation adsorption was washed with then dried the at 353 K and calcined at 773 K for 5 h. The results showed that the adsorption efficiency of regenerated Mg-Al-HT had not changed markedly, the adsorption capacity being between 1217 mg/g and 1244 mg/g, and the adsorbent recovery reclaim rate was between 77-83% at three recycles. We supposed that the hydrotalcite possesses a special "memory effect"<sup>1, 9</sup>, in the particular condition, it can reconstitute the original Mg-Al hydrotalcite layered structure. It was likely that the hydrotalcite possessed many pore channels and higher surface area after removing the NO<sub>3</sub><sup>-</sup> in the Mg-Al-HT by calcination at 773 K.

## Conclusion

Mg-Al-HT was a suitable adsorbent for NO<sub>x</sub>. The adsorption capacity of Mg-Al-HT for NO<sub>x</sub> was quite high, the static adsorption capacity was 1398.2 mg/g. The adsorption capacity depended upon the adsorption time and temperature, and the adsorption rate was very rapid in the beginning, higher temperature is not favorable to the adsorption capacity. Mg-Al-HT could be recycled by thermal decomposition when adsorption reached saturation. Its adsorption capacity could be regenerated easily.

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