DEVELOPMENT OF COMBINED NO AND SO_2 REMOVAL PROCESSES

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Introduction

Activated carbons are of interest for the removal of SO_x and NO_x through adsorption from flue gases generated by fossil flue combusion.[1,2] In removal processes of SO_x and NO_x, combining the required purification functions into a single gas processing unit, therefore, represents a major potential advantage with regard to capital cost and process simplicity. [3,4]

In the present study, the catalytic activity for SO_2 and reduction of NO with NH_3 and the reactivity of pitch- and PAN-based activated carbon fibers prepaed with steam could be controlled by preparation conditions. The effects of oxygen functional groups and surface area on catalytic activity using activated carbon fiber catalysts are investigated. The difference in catalytic activities with precusor species and preparation conditions, especially activation temperature and flow rate of steam, was explained with the aid of activated carbon fiber characterization.

Experimental

The preparing pitch- and PAN-based carbon fibers were used as raw material. The activation of carbon fibers (5g of sample in each experiment) was made at 800, 850, 900℃ with different flow rate of steam/N_2 mixture(10～100 ml/hr) in a horizental furnance.

The surface area of the fiber before and after various treatments was measured by adsorption of nitrogen at 77K using a Quantachrome Autosorb. Elemental analysis was carried out with a YANACO CHN CODER MT-3 elemental analyser. Pore size distributions were determined using the Dubinin-Raduskevich equation

SO_2 and NO removal over activated carbon fibers in fixed reactor were measure using ECOM A^+ analyser. The properties of PAN-ACFs and pitch-ACFs are listed in table 1. The conditions used for SO_2 removal: 1000ppm SO_2 / 10vol% H_2O / 10vol% O_2 /balance N_2; several reaction temperatures(20℃～100℃); flow rate=300 ml/g · min. The conditions for NO were: 500ppm NO / 500ppm NH_3 / 10% O_2 /balance N_2 at the same reaction temperatures and flow rate mentioned above.

The effects of preparation conditions on sulfur capture capacity and the catalytic activity for reduction of NO with NH_3, reaction temperature, oxygen, steam concentration and NH_3/NO mole ratio over the activated carbon fibers were investigated in a fixed bed integral reactor.

Result and Discussion

The breakthrough profiles of SO_2 and NO over PAN-ACF and pitch-ACF at 20℃ were shown Fig. 1. The gas carried 10 vol% water and 5 vol% O_2 in all case. Previous work has shown that the catalytic activity was related to the different characteristics of activated carbon fibers and PAN- and pitch-based ACF showed high activity for SO_2 removal and reduction of NO, respectively. Over PAN-ACF which exhibited SO_2 was adsorbed completely until 4.5 hr, no SO_2 being detected at all at the outlet of the reactor. SO_2 removed of about 92% from the flowing gas continuously.

Since NO is a supercritical gas at the ambient temperature, very little NO adsorbs on porous adsorbents. However, the breakthrough curves of pitch-based activated carbon fibers show that there was neither NO nor NO_2 detected in the exit gas during the first 3hr. The conversion of NO with NH_3 over activated carbon fibers was 100% at 20℃. After 3hr, dereasing gradually to 60%, after which the activity was maintained for at least 10hr, while reducing the NH_3 concentration reduced conversion value under the same conditions.

It is assumed that nitric oxide reduction at low temperatures also strongly inhibited by humidity, because the adsorption of water vapor prevents NO adsorption on the catalyst.

However, the present pitch-based ACF exhibits significant activity for the reduction of low concentrations of NO with NH_3 at ambient temperature, even at humidity. The pitch-based ACF appear to possess unique properties for this reaction, in comparison to PAN. The breakthrough time of NO increase with an increase in the concentration of oxygen. When moisture is present, the amout of NO adsorbed as HNO_3 increases

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In NO_x and SO_2 removal simultaneously, the activated carbon fibers exhibited high catalytic activity for reduction of NO with NH_3 and the reactivity of SO_2. The pitch-based ACF exhibits significant activity for the reduction of NO with NH_3 at ambient temperature, even at humidity

However, the reactivity of NO_x with commercial active carbon does not correspond to the rate of desulfurization. In order for the active carbon process to be more effective for the simultaneous SO_2 and NO_x system, our efforts should be made to raise the activity of activated carbon fibers catalysts.

REFERENCE

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Table 1. Proprties of PAN-ACFs and pitch-ACFs used.

	Elemental Analysis C	H	N	O	Surface Area m^2/g	Pore Volume cc/g
PAN-1	82.7	1.6	3.3	12.4	453	0.32
2	83.3	1.4	1.2	14.1	614	0.36
3	83.8	1.3	1.4	13.5	805	0.41
Pitch-1	90.6	0.9	0.6	7.9	1135	0.57
2	91.8	0.8	0.4	7.0	1432	0.71
3	92.4	0.6	0.3	6.7	1760	0.84

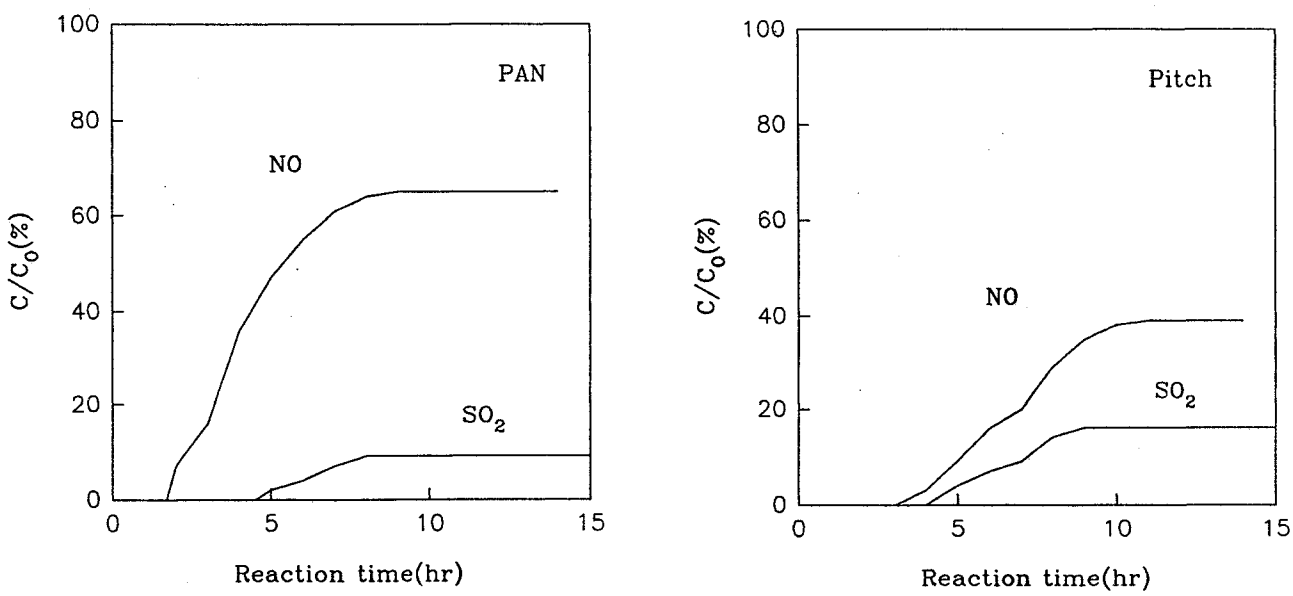

Fig. 1. The breakthrough profiles of SO_2 and NO over PAN-ACF and pitch-ACF at 20℃.