

Simultaneous removal of particulates and NO by the catalytic bag filter containing MnOx catalysts

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Abstract—A catalytic bag filter which can remove particulates and NOx simultaneously was prepared and tested in a laboratory and a pilot plant. Manganese oxides (MnOx), active for the selective NO reduction with NH₃ at low temperatures, was utilized as the catalyst. This MnOx-coated bag filter showed 92.6% NOx conversion at 423 K with a space velocity of 400,000 h⁻¹.

Key words: Catalytic Bag Filter, Particulates Removal, NO Reduction, NH₃-SCR

INTRODUCTION

The emission control of nitric oxides (NO, NO₂ and N₂O) from various combustion processes is a major environmental concern. These gases affect air quality because they cause photochemical smog, acid rain, ozone depletion, and green house effects [1-3]. To date, more than 900 stationary sources world-wide have been equipped with post-combustion NOx control technology involving selective catalytic reduction (SCR) and selective non-catalytic reduction (SNCR). The major products of both processes are harmless nitrogen and water vapor. Both technologies were first applied in Japan in the 1970s and have since been installed in the US and several European countries. This technology is currently experiencing unprecedented growth and development. SCR has been more widely used than SNCR despite its higher cost because many countries now require very low NOx emissions (200 mg/m³) from combustion plants, and this can be achieved only with reduction efficiencies up to 80% [4].

In the tail end system, the SCR reactor is placed at the end of the gas cleaning chain directly following the desulfurization plant. Since only small amounts of SOx and particulates reach the catalyst, very little catalyst deactivation occurs. However, the flue gas temperature after normal reheating (383-423 K) is too low for most catalyst types [5,6]. Accordingly, reheating beyond that normally required is necessary to reach the effective operating temperature of the catalyst (up to 473 K); the cost of reheating only 50 K can be as much as 2-3% of the boiler capacity, unless a regenerative heat exchanger is used. The greatest disadvantage is the necessity of reheating the flue gas beyond that normally required after scrubbing.

Catalytic filters are special multifunctional reactors capable of carrying out two operations at the same time: particulates removal and catalytic abatement of noxious gases such as VOCs [7], dioxin and furan [8,9], nitrogen oxides [10,11]. This might entail a poten-

tial reduction in space requirements as well as energy and investment costs, as pointed out in a recent review [12].

A bag filter, placed downstream of the desulfurizer and electrostatic precipitator, is a widely used industrial separator for particles from SCR process and the temperature at this point is 423-433 K. If catalysts that operate efficiently at 423-433 K or lower can be developed, the catalytic processes will become economically attractive and probably find much greater use. Recently, Fino et al. reported that the incorporation of MnOx-CeO₂ catalyst, which is active at low-temperature NH₃-SCR into the catalytic filter system, could improve the De-NOx performance especially at low temperatures [13-15]. In a previous study [16], it has been shown that the MnOx catalyst exhibited high activity at the reaction temperatures in the range of 373-473 K at high space velocity of 400,000 h⁻¹. Furthermore, this catalyst was confirmed to be superior to MnOx-CeO₂ catalyst [16].

In this study, the catalytic bag filter containing MnOx catalysts was examined in a laboratory and also tested in a pilot plant to determine its feasibility in a real system.

EXPERIMENTAL

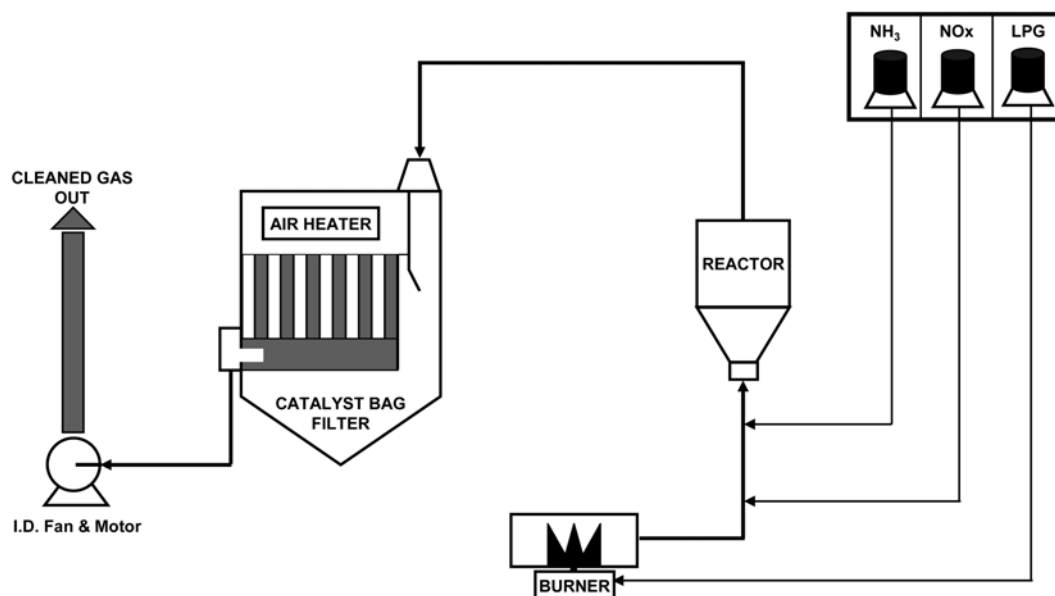
1. Preparation of Catalyst

Manganese oxides were prepared by a precipitation method with sodium carbonate as a precipitant. 0.5 M sodium carbonate (Na₂CO₃, SHINYO, 99.0%) aqueous solution was continuously added to 500 ml of 0.5 M manganese acetate ((CH₃COO)₂Mn·4H₂O, JUNSEI, 98.0+%) aqueous solution until the pH of the solution reached to 8. The resulting precipitate was aged at 298 K for 1 h, filtered, and washed several times with distilled water. The cake was dried in air at 393 K for 12 h and calcined at 623 K for 4 h in static air.

The bag filters employed in the laboratory and the pilot plant were prepared by a foam coating of MnOx. The calcined MnOx catalyst was scattered and then infiltrated with a vacuum suction method on the bag filter. And then polytetrafluoroethylene (PTFE) foam coating for lamination was conducted by a series of form gen-

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Scheme 1. A schematic flow diagram of the de-NO_x pilot plant.

erator, foam coater and pre-dryer using conventional winding equipment. They were dried again at 473 K for 12 h. The different amounts of catalyst coated on a bag filter from 100 to 750 g/m² can be controlled by repeating this procedure.

2. Reaction Test in a Laboratory

A reaction temperature such as 373 and 423 K was chosen for the comparison of laboratory results with those of a pilot plant. In the laboratory measurement, the catalytic performance was tested by MnO_x powder (as denoted by LAB-P) and MnO_x-coated bag filters (as denoted by LAB-C) in a tubular flow reactor of 8-mm i.d. or 20-mm i.d., respectively. The reactant gas typically consisted of 500 ppm NO, 500 ppm NH₃, 5 vol% O₂ in N₂ flow. In these experiments, MnO_x weights of about 600 mg were used, corresponding to gas hourly space velocity (GHSV) of 400,000 h⁻¹ for the MnO_x powder (LAB-P), and 100,000 h⁻¹ for MnO_x coated on bag filter (LAB-C). The NO_x concentration in the inlet and outlet gas was analyzed by means of an NO/NO₂ combustion gas analyzer (Euroton).

3. Reaction Test in a Pilot Plant

The flue gas was introduced into a reactor by I.D. fan & motor

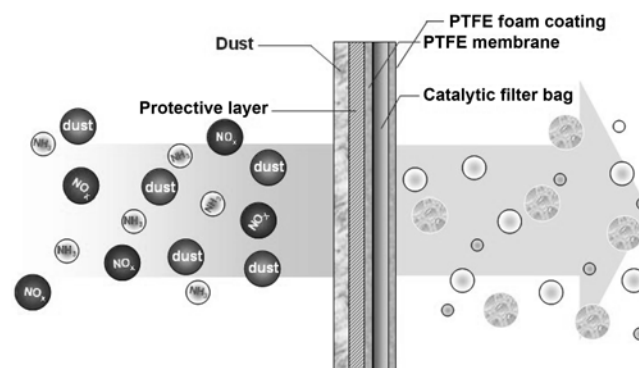
from the down stream of the bag filter in the pilot plant as shown in Scheme 1. The flue gas, which is produced from power plants, normally contains NO_x of 500 ppm in air flow. The NH₃ required for the NO removal was supplied by an NH₃ cylinder. The quantity of necessary NH₃ was determined on the basis of the NO_x concentration at the inlet of the reactor. The ratio of NH₃ to NO_x was normally about 1.0. The reactant gas typically consisted of 500 ppm NO and 500 ppm NH₃ in air flow. The NO_x concentration in the inlet and outlet gas was analyzed by means of a NO/NO₂ combustion gas analyzer (Euroton). All operating parameters of the reaction system were controlled by a computer-interfaced automatic system. The specification of the pilot plant is listed in Table 1.

RESULTS AND DISCUSSION

Scheme 2 shows the schematic diagram of de-NO_x process utilizing a catalytic bag filter. During the operation, particles in a flue gas were collected in the upstream of a filter membrane; whereas NO_x was transformed by a reaction with an injected NH₃ into N₂

Table 1. The specification of a pilot plant and a bag filter

	Specification
Capacity (m ³ /min)	10-30
Filtering velocity (m/min)	0.5-0.9
Filter materials	P-84, PTFE
Filtering area (m ²)	37.50
Temperature (K)	373-473
Pressure (mmH ₂ O)	400
Permeability (cm ³ /cm ² /sec)	2.0-3.0
I.D. Fan & Motor	380 V×5.5 KW×2P
Bag size	φ150×2200L×6
Bag quantity (SET)	6



Scheme 2. A schematic diagram of the de-NO_x process via the catalyst-coated bag filter.

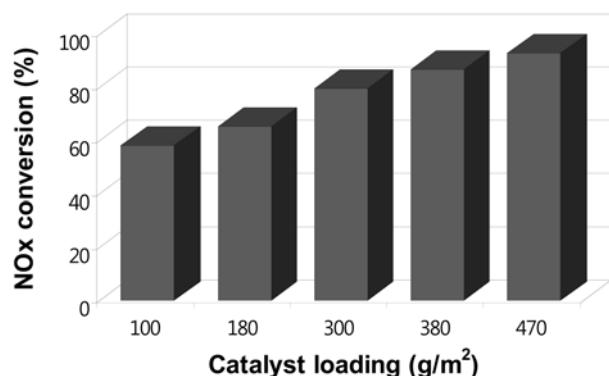


Fig. 1. NOx conversions over bag filters containing different amounts of MnOx at 423 K. Reactants; 500 ppm NO, 500 ppm NH₃ and 5 vol% O₂ in N₂. The gas hourly space velocity (GHSV) was 100,000 h⁻¹.

and H₂O over the catalytic filter, simultaneously. Another advantage of this system is that the upstream filter membrane collecting particles with a very high efficiency can protect the catalyst from being plugged with this potential flue gas poison.

The MnOx catalysts were coated on a bag filter (LAB-C) with a different loading from 100 to 470 g/m² and their corresponding catalytic activities are shown in Fig. 1 at a space velocity of 400,000 h⁻¹. The NOx conversion increased continuously with increasing catalyst loading up to 470 g/m². Consequently, the MnOx coated on a bag filter with a loading of 470 g/m² showed the highest activity and the NOx conversion was 93% at 423 K. The sudden decrease in the NOx conversion was observed at a catalyst loading of 750 g/m², which can be related to the rapid increase in the pressure drop. Because the pressure drop of the bag filter is the most important variable in the removal of particulate as well as NOx in this system, the higher loading above 470 g/m² was not examined further.

In the laboratory, the performance of MnOx powder (LAB-P) and MnOx-coated bag filters (LAB-C) for the low-temperature SCR at

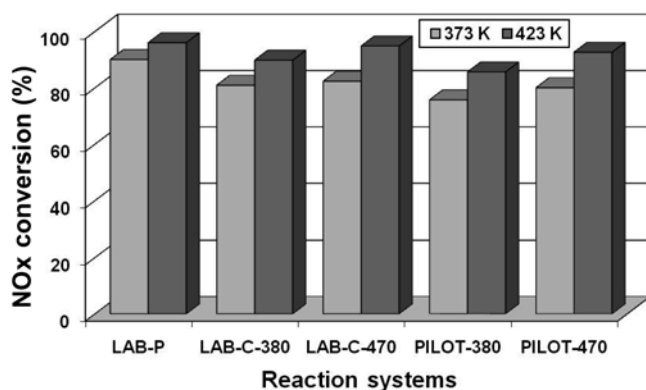


Fig. 2. NOx conversions over MnOx powder and MnOx-coated bag filter in a laboratory test and in a pilot plant test. Reactants: 500 ppm NO, 500 ppm NH₃ and 5% O₂ in N₂ for a laboratory test and 500 ppm NO and 500 ppm NH₃ in air for a pilot plant test. The gas hourly space velocities (GHSV) were 400,000 h⁻¹ for LAB-P, 100,000 h⁻¹ for LAB-C and 400,000 h⁻¹ for PILOT.

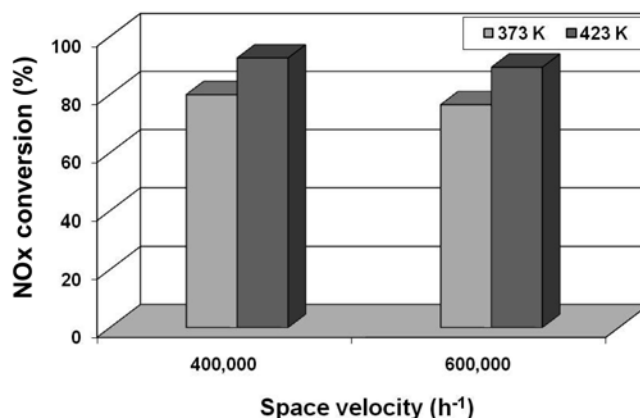


Fig. 3. NOx conversions over a MnOx-coated bag filters with a MnOx loading of 470 g/m² at 373 and 423 K. Reactants: 500 ppm NO and 500 ppm NH₃ in air.

reaction temperatures such as 373 and 423 K operating under the identical experimental condition is presented in Fig. 2. MnOx powder (LAB-P) was tested at 373 and 423 K with a high space velocity of 400,000 h⁻¹. The NOx conversion was 90.0 and 96.0%, respectively. The catalytic bag filters coated with 380 and 470 g/m² of MnOx catalyst (LAB-C) were also tested under the identical experimental condition for catalytic activity. The NOx conversions at 373 and 423 K were slightly lower than those of MnOx powder, and the MnOx-coated bag filter with a loading of 470 g/m² appeared to be slightly superior to that with a loading of 380 g/m². A similar result was also observed in a pilot test.

Fig. 3 shows the catalytic activity of the MnOx-coated bag filter at different space velocities. The space velocity was defined as the ratio of the flow rate of the flue gas to the volume of the bag filter. At a space velocity of 400,000 h⁻¹, the NOx conversion was 80.0 and 92.6% at 373 and 423 K, respectively. This high NOx conversion was maintained even at a much higher space velocity, which is very important for a practical application.

Fig. 4 exhibits the catalytic activities and particulate removal efficiency of the MnOx-coated bag filter in a pilot plant. The high NOx

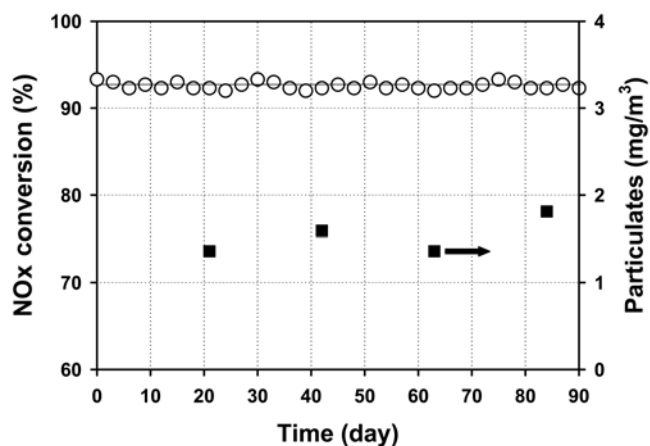


Fig. 4. NOx conversion and the particulate removal efficiency over a MnOx-coated bag filter at 423 K with time on stream. Reactants: 500 ppm NO and 500 ppm NH₃ in air.

conversion such as 92.6% was achieved and no deactivation was observed up to 90 days of operation in the presence of particulates. The particulates were introduced to the pilot plant at a concentration of 5,000 mg/m³. After passing through the catalytic bag filter, the particulate concentration decreased below 2.0 mg/m³. Therefore, the particulate removal efficiency was maintained at 99.9% for 80 days. Based on these results, this MnOx-coated bag filter developed in this work can be considered as a promising candidate for the multifunctional reactor to remove particulates and NOx, simultaneously. For instance, Saracco [10] reported that 95% of NOx conversion was obtained via NH₃-SCR at high temperatures ranging from 523 to 623 K by utilizing the catalytically modified fly-ash filters containing V₂O₅-TiO₂ catalyst. Therefore, additional reheating is required. However, in our case, even at a low temperature of 423 K, the high NOx conversion such as 90% can be achieved at a space velocity of 600,000 h⁻¹. This catalytic bag filter also showed very stable de-NOx activity at low temperatures and high removal efficiency of particulate over a long operation time, which is very important for a practical application.

CONCLUSIONS

A catalytic bag filter containing 480 g/m² of MnOx catalysts showed 92.6% conversions of NOx at 423 K and 99.9% of particulate removal efficiency. This MnOx-coated bag filter seems to be a promising unit for the selective catalytic reduction of NOx with NH₃ at low temperatures and particulate removal in the industrial application.

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