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The Reactivity of V_2O_5-WO_3-TiO_2 Catalyst Supported on a Ceramic Filter Candle for Selective Reduction of NO

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Abstract−For realizing the environmental issues and constituting an economical treatment system, a catalytic filter based on V_2O_5/TiO_2 supported on tubular filter elements has many advantages by removing NO_x and particulate simultaneously from flue gas. In order to improve the activity of a catalytic filter based on V_2O_5/TiO_2 supported on a commercial high temperature filter element (PRD-66), the promoting effects of WO_3 were investigated in an experimental unit. PRD-66 presented very good properties for SCR catalyst carrier since it contains much active material such as Al_2O_3, SiO_2, and MgO whose contributions were remarkable. For additional catalyst carrier, TiO_2 particles were coated in the pores of PRD-66 with relatively good distribution of the particle size less than 1 μm, by a coating process applying centrifugal force. WO_3, in the V_2O_5-WO_3-TiO_2/PRD-66 catalytic filter system, increased the SCR activity significantly and broadened the optimum temperature window. The catalytic filter shows the maximum NO conversion of more than 95% for NO concentration of 700 ppmv at face velocity of 0.02 m/sec, which is comparable to the current commercial catalytic filters of plate form.

Key words: Promotion Effect, WO_3, Catalytic Filter, NO Reduction, Ammonia, V_2O_5/TiO_2

INTRODUCTION

Catalysts for selective catalytic reduction (SCR) of NO_x have been widely developed in their forms and compositions [Busca et al., 1998; Che et al., 2000; Choi et al., 1999a, 1994; Nam et al, 1995; Pârvulescu et al., 1998; Ham et al., 2000]. Among them, catalysts based on vanadia-titania are effectively commercialized for the reduction of NO_x with ammonia in SO_2-containing flue gases [Amiridis et al., 1996; Bosch and Janssen, 1988; Forzatti, 1966; Terabe et al., 1994]. Vanadium oxide has been known as the active site not only for SCR but also for the undesired oxidation of SO_2. Thus, it is present in very small amounts (<1% w/w) in commercial catalysts. The catalysts contain a large amount (<10% w/w) of WO_3 or MoO_3 over the TiO_2-anatase carrier [Alemany et al., 1996]. WO_3 has been known as providing much less activity for SCR but presenting an inhibiting effect against the oxidation of SO_2 by vanadium oxide as well as stabilizing the structural and morphological characteristics of V_2O_5 anatase [Alemany et al., 1995]. WO_3 also increases the acidity of TiO_2 [Sohn and Bae, 2000]. One of the recent key issues on the commercial SCR catalyst is to limit the possible oxidation of SO_2 to SO_3 as much as because SO_3 forms alkali sulphates. Although sulphate species of monolayer on vanadia stabilizes the anatase phase of vanadia inhibiting the phase transition [Alemany et al., 1996], they inhibit the SCR activity by plugging the monolith gap. Such a plugging problem in the monolith catalyst also causes a serious operational problem of this catalyst, especially at high dust concentration. Therefore, the role of W atom in the V_2O_5/TiO_2 catalytic system is very important.

The advantage of the catalytic filter in the form of rigid filter elements like ceramic filter candle is solving the plugging problem while providing the lower pressure drop system as well as its dual function for the control of particulates and nitric oxides. The advanced ceramic filter elements for application at high temperature are composed of two layers: interior support and exterior membrane layer [Choi et al., 1999b]. Most of the filtration is carried out through the membrane layer whose thickness is about 100 μm. The support layer is useful not for the function of the filtration but for a rigid supporter [Choi et al., 1999c]. The catalytic filter utilizes these large pores of the support layer whose pore size is about 100 μm for the room of the catalyst support. However, the potential task for applying the catalytic filter in an advanced system is how to increase the catalytic activity by increasing the catalyst dispersion. Recently, the technology to support TiO_2 particles in the pores of PRD-66 has been successfully developed by using the sol-gel centrifugal coating process [Choi et al., 2000]. The V_2O_5-TiO_2 based-catalytic filter prepared by the method presents NO conversion more than 90% for the NO concentration of 500 ppmv at the face velocity of 0.02 m/sec. The result satisfactorily meets the regulation of NO from the stationary process. However, in order to meet the ammonia slip (less than 10 ppmv) recommended for the commercialization of SCR unit using ammonia, the catalytic activity still needs to improve. So the aim of this work is to improve the catalytic activity by using WO_3. The promoting effect of WO_3 was discussed from the experimental results obtained by using simulated gas.

EXPERIMENTAL

1. Preparation of Catalytic Filter

The catalytic filters were prepared with the methods of the centrifugal coating of TiO_2 sol-gel prepared using tetra isopropyl orthotitanate (TIPOT) [Choi et al., 2000]. The commercial ceramic filter element, PRD-66 (PRD), from AlliedSignal was utilized as received form and compositions. The PRD, which is prepared by spin-weav-

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ing the textile glass yarn, is a composite of corundum, cordierite, and mullite. It has high porosity of 60% with its pore size of the support layer larger than 100 μm. It has an effective membrane layer of median pore size about 10.5 μm. The original dimension of the filter candles is 1.5 m length, 0.04 and 0.06 m of inner and outer diameter, respectively. It was cut in the tubular form of 0.02 m length in order to fix in a small experimental reactor. The filter tube was washed in an ultrasonic water bath and treated in 0.05 M NaOH aqueous solution and dried for 2 hr at 110 °C before being used.

The TiO_2 particles were first supported on the PRD by letting TiO_2 sol-gel solution flow across the filter element by applying centrifugal force [Choi et al., 2000] in order to enhance for the TiO_2 particles to penetrate into the inner layer of the filter element. The rotation speed was controlled at 580 rpm and measured by a tachometer. Colloidal TiO_2 was prepared with a sol-gel method [Terabe et al., 1994; Choi et al., 1999a]. The TiO_2-loaded filter tube was dried overnight at room temperature and 1 hr at 110 °C and then calcined in the air stream at 450 °C for 2 hr. In order to impregnate the catalysts, TiO_2-loaded PRD was immersed in the oxalic acid solution of ammonium metavanadate and/or ammonium paratungstate. The solution was slowly evaporated at low temperature until the filter element absorbed all of the solution (called dry impregnation), followed by drying overnight and calcining at 450 °C for 2 hr. In the dip impregnation process, TiO_2-loaded PRD was immersed in the excess solution for 12 hr, followed by drying overnight and calcining at 450 °C for 2 hr. The catalytic filters fabricated in the several compositions and different procedures are described in Table 1. The composition of catalysts in Table 1 denotes the weight percent over the PRD. Catalytic filters of VWT series denote V_2O_5-WO_3-TiO_2/PRD. VWT2 and VWT4 represent the catalytic filters fabricated by the procedure that V atoms were impregnated on the previously prepared WO_3-TiO_2/PRD. VOT, VWO and OWT catalytic filters present V_2O_5-TiO_2/PRD, V_2O_5-WO_3/PRD, and WO_3-TiO_2/PRD, respectively. The morphology and elemental composition of the catalytic filter were analyzed with SEM and SEM-EDS, respectively.

2. The Performance Test

The activity of the catalytic filter was tested in an experimental unit using a gas mixture of NO, NH_3, O_2, and N_2 [Choi and Ahn, 1999a]. 0.2% NH_3 in N_2, 0.15% NO in N_2, and air were used as the source of the gas mixture. In a normal test run, the concentrations of NO, NH_3, and O_2 were 500, 500 ppm, and 2 vol% in N_2 balanced, respectively. The total gas flow rate was exactly controlled with a mass flow controller at the actual face velocity of 0.02 m/sec. The catalytic filter was mounted using flanges. It was first fixed between top and middle flanges with long bolts. The reaction part was sealed with the top and bottom flanges. The reaction gases were introduced from bottom inlet of the reactor and passed out across the catalytic filter. Thus, the direction of gas pass (inward from the outer surface) is the same as that of industrial filtration mode using ceramic filter element. After the impregnation step, the catalytic filter was dried and kept in a low humidity chamber. And it was calcined on-site at the reaction unit before being used for the test of catalytic performance. NO conversion over the catalytic filter was measured at steady state of each temperature.

The concentrations of NO, NO_2, and NH_3 were continuously analyzed by using a chemiluminescence method (TEI model 17, Thermo Environmental Instruments Inc.). Oxygen concentration was analyzed by an off-line GC with TCD using GS-Q capillary column.

RESULTS AND DISCUSSIONS

1. Characteristics of the Catalytic Filters

The particle size of TiO_2 sol-gel is effectively controlled by the change of HCl/TIPOT mole ratio (H/T) and increases with the increase of HCl concentration [Choi et al., 1999a]. H/T was adjusted to 3 in this study in order to control the particle size of TiO_2 about 400-500 nm. About 50 ml of the sol-gel solution was circulated 10 times across the filter element with a rotation speed of 580 rpm. Fig. 1a represents the SEM photograph of the fresh PRD showing that the external membrane layer has very fine pores and the interior supporting one has large pores of more than 100 μm. TiO_2 particles less than 1 μm were coated with good distribution in the interior pores of PRD as shown at Fig. 1b. Thus it is certain that the coating method applying a centrifugal force is very effective to load the TiO_2 particles into the inner pore of the support layer of the filter element. In case of VWT3 composed of V_2O_5-WO_3-TiO_2/PRD, V and W atoms were also distributed over the entire surfaces of both the PRD and TiO_2. It is certain then that the PRD has a remarkable surface area for the catalyst carrier. The elemental analysis using SEM-EDS shown in Table 2 presents the amount of surface element for catalytic filter. TiO_2 covers about 65% of total surface of catalytic filter. And total coverages of V and W atoms are 2.7 and 9.2% w/w, respectively, with respect to TiO_2. Parts of

Table 1. The description of catalytic filter prepared by sol-gel centrifugal method

Name	V_2O_5 loading (% w/w)	WO_3 loading (% w/w)	TiO_2 loading (% w/w)	Impregnation method	Solvent	BET surface area (m^2/g)
VWT1	0.04	0.3	3.2	Dip co-impregnation	Oxalic acid	1.67
VWT2	0.06	0.5	3.0	Dip V impreg. on WO_3/TiO_2/filter	Oxalic acid	2.42
VWT3	0.46	3.7	3.1	Dry co-impreg.	Oxalic acid	1.33
VWT4	0.47	3.8	3.1	Dry V impreg. on WO_3/TiO_2/filter	Oxalic acid	1.16
VOT	1.62	-	2.9	Dry impreg.	Oxalic acid	0.19
OWT	-	1.1	4.9	Dry impregnation	Oxalic acid	3.49
VWO	0.49	3.9	-	Dry impregnation	Oxalic acid	0.57
OOO	-	-	-	Fresh filter	-	0.10
OOT	-	-	3.1	TiO_2/filter	-	1.38

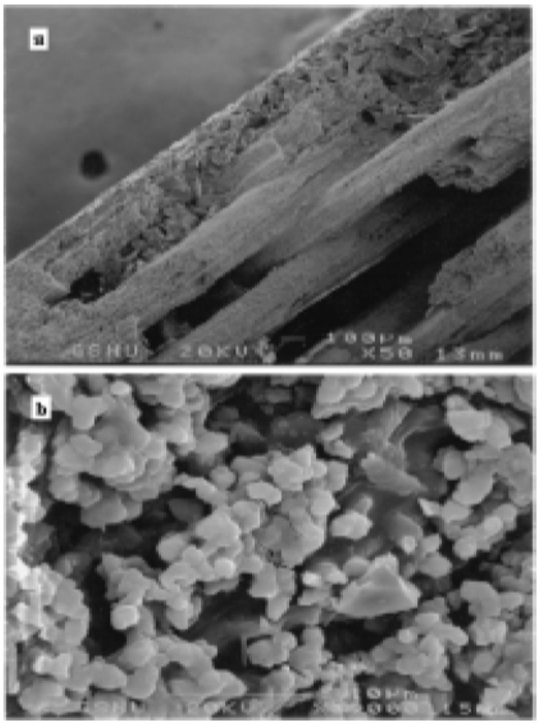

Fig. 1. SEM images of fresh PRD-66 (a) and TiO_2 supported on PRD-66 (b).

Table 2. Elemental analysis of catalytic filter by SEM-EDS

Element	Fresh PRD-66		VWT3	
	Atomic (%)	Elemental (%)	Atomic (%)	Elemental (%)
Mg(MgO)	3.68	3.28	-	-
Al(Al_2O_3)	61.03	60.38	24.00	12.59
Si(SiO_2)	35.29	36.35	-	-
Ti(TiO_2)			67.94	63.28
V(V_2O_5)			1.81	1.79
W(WO_3)			6.25	22.34

these atoms are also loaded on the TiO_2-free surface of PRD.

The BET surface area of the PRD is about 0.1 m^2/g, according to the data presented by maker [AlliedSignal, 1999]. And the value measured in this study was 0.96 m^2/g in the form of fragment about 1 mm. The different value in BET surface area for the fragment form is due to the increase of surface by reducing the particle size. The BET surface areas shown in Table 1 are corrected by eliminating the difference (0.86 m^2/g) from the measured one. The surface area of catalytic filter increased additionally 1.28 m^2/g by supporting TiO_2. However, it was markedly decreased from 1.38 (for OOT) to 0.19 m^2/g (for VOT) by the loading of V atoms. However, it increased to 3.49 (for OWT) from 1.38 m^2/g (for OOT) by the loading of W atoms. The surface area of VWT3, containing both V and W oxide was 1.33 m^2/g, which means that W atoms increase the surface area of V_2O_5-TiO_2/PRD. BET data in Table 1 reveal a general pattern that V loading reduces the surface area while W loading increases it. This result is similar to the case of V_2O_5-WO_3/TiO_2 ternary system reported by Alemany et al. [1995]. They reported that the surface area of V_2O_5(x)-WO_3(9)/TiO_2 gradually decreases as the V_2O_5 loading (x) increases, while WO_3 loading increases BET surface area at a given V_2O_5 loading or bare TiO_2.

2. Performance of the Catalytic Filter

In order to investigate the effect of WO_3 on NO reduction activity of SCR catalytic filter, NO conversion over catalytic filters, OWT (WO_3-TiO_2/PRD), VOT (V_2O_5-TiO_2/PRD) and VWT3 (V_2O_5-WO_3-TiO_2/PRD), were compared. Fig. 3 represents NO conversion when NO concentration in the reactant is 500 ppm at the face velocity of 0.02 m/sec. VWT3 shows the maximum NO conversion of 95%, while VOT and OWT show 68 and 75%, respectively. NO conversion over OWT is less than 40% in the optimum temperature range of 320-350 °C for VOT. And the temperature window presenting NO conversion of more than 95% of its maximum is broadened from 30 °C (320-350 °C) for VOT to 60 °C (290-350 °C) for VWT3. The results indicate that WO_3 significantly promotes catalytic activity at lower temperature and broadens the temperature window. However, OWT containing only WO_3 reveals low activity at high temperature (400 °C) with a narrow temperature window. This result is very similar with that over the ternary (V_2O_5-WO_3-TiO_2) catalyst reported by Alemany et al. [1995]. They reported that WO_3 in the catalytic system promoted SCR activity as well as increased the selectivity to N_2. In the presence of WO_3 (9% w/w), the temperature required for 50% conversion of NO was lowered from 300 to 270 °C for V_2O_5 (0.78% w/w), and to 210 °C for V_2O_5 (1.4% w/w). And the temperature window for maximum NO conversion was greatly broadened in the ternary catalyst from 15 to 70 °C for V_2O_5 (1.4)-WO_3 (9)/TiO_2 since the catalyst has high activity at lower temperature and high selectivity to N_2 at higher temperature. The role

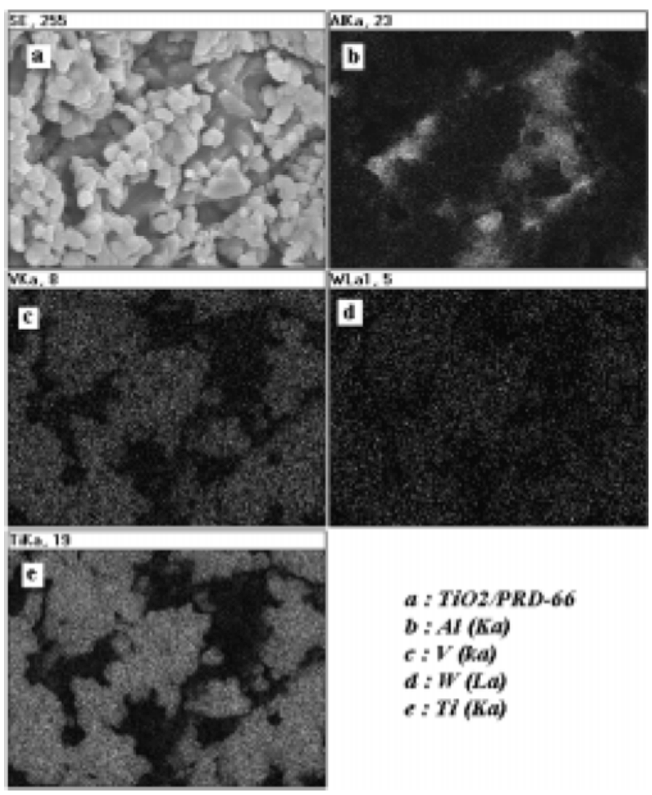

Fig. 2. EDS for elements mapping of VWT3 catalytic filter.

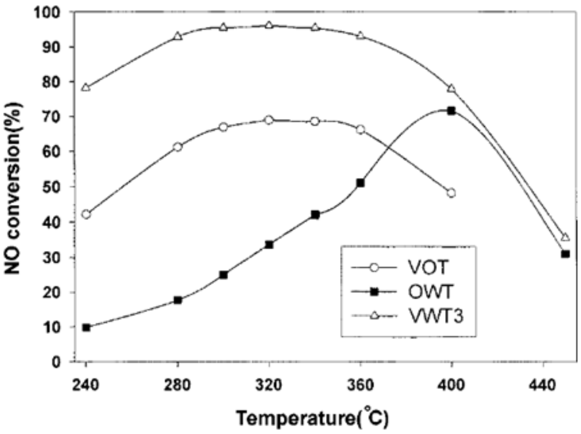

Fig. 3. The effect of WO_3 on the catalytic activity of V_2O_5-TiO_2/PRD.

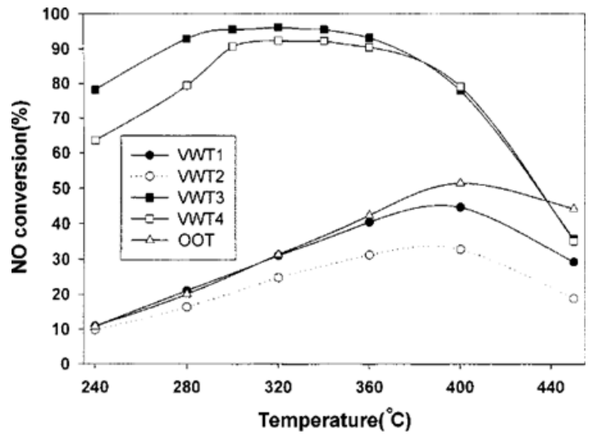

Fig. 5. The effect of V loading on NO conversion over VWT catalytic filters.

of WO_3 in the ternary catalyst has been understood as increasing the surface acidity of V_2O_5/TiO_2 [Shin et al., 1994]. And the higher reactivity is related to their superior redox properties owing to interaction between the monomeric V and W oxides over TiO_2 [Alemany et al., 1995]. In case of catalytic filter fabricated with PRD filter element, the PRD itself has several kinds of oxide support. According to the manufacturer's report [AlliedSignal, 1999], the PRD is composed of corundum (alumina), cordierite, and mullite after fabricating from the oxide mixture of Al_2O_3, SiO_2, and MgO. V_2O_5/Al_2O_3 is a very good SCR catalyst [Saracco et al., 1995]. SiO_2 and MgO show relevant properties for catalyst carrier [Pârvulescu et al., 1998]. Although vanadia/alumina and vanadia/silica have a lower activity for SCR than vanadia/titania, titania grafted on silica or alumina shows a remarkable activity. Thus, it is estimated that V_2O_5-WO_3-TiO_2/PRD has plenty of active surface composed of complicated active sites formed on TiO_2 particles as well as the PRD itself. This assumption is evident from the analysis of EDS of VWT3 shown in Fig. 2. According to the result, TiO_2 particles form the isolated particles in the pore of the PRD while V_2O_5 and WO_3 are distributed all over the surfaces (bare PRD and TiO_2 particles) exposed. The results shown at Fig. 4 also support the assumption since VWO containing no TiO_2 particles has a good SCR activity, which means PRD has remarkable surface areas for SCR catalyst of V_2O_5-WO_3. However, VWO catalytic filter has a limited activity that is not satisfactory when NO concentration is high as much as 700 ppmv. And the selectivity to N_2 is relatively low at the high temperature region. Consequently, the PRD-66 filter element additionally supplies the sites for the catalyst supporter.

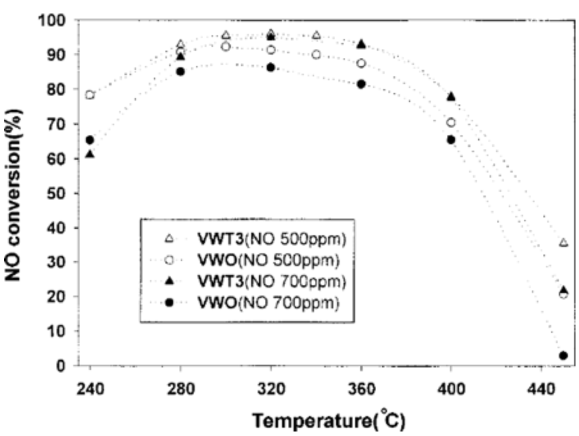

Fig. 4. NO conversion over VWT3 and VWO at different NO concentration.

As the same aspects reported previously about ternary catalyst [Alemany et al., 1995; Topsøe et al., 1995], the pattern of the reaction over the catalytic filter was considerably affected by the V loading. Fig. 5 shows that VWT1 and VWT2 with the low load of V and W atoms present very low NO conversion that is lower than that of OOT. The loadings of V and W atoms in these two catalysts are less than 1.5% w/w with respect to TiO_2 and less than 0.01% w/w with respect to the PRD. Topsøe et al. [1995] reported that more than 2% w/w of V_2O_5 over TiO_2 is required for the monolayer coverage of V_2O_5 on TiO_2 (BET surface area is 90 m^2/g). Alemany et al. [1995] also reported that V and W cover 23 and 69% only for the high load catalyst of $V_2O_5(2.56)$-$WO_3(9)/TiO_2$ (BET surface area is 90 m^2/g). They reported that pure TiO_2 represents the Lewis acid sites (Ti-OH) and V_2O_5 generates Brønsted acid sites (V-OH) known as the active sites for NO reduction. The reason why catalysts prepared with dry impregnation show higher NO reduction than those prepared with wet impregnation is high catalyst loading. Fig. 5 also shows that VWT2 and VWT4 have lower activity than VWT1 and VWT3 fabricated with dry co-impregnation method, containing a corresponding amount of V and W, respectively. VWT2 and VWT4 were prepared by V impregnation over the WO_3-TiO_2/PRD preliminary prepared by the impregnation with ammonium paratungstate oxalic solution. Several catalytic filters fabricated by this procedure, not mentioned in this paper, always showed lower activity than a co-impregnated one. This result implies that parts of active sites of WO_3 preliminarily exposed are lost by the coverage by post-loaded V_2O_5. So the assumption that active sites (V_2O_5) are formed on the W-modified TiO_2 surface is not reasonable. Otherwise, the experimental results support the assumption mentioned by Alemany et al. [1996] that the active sites are monomeric isolated V and W oxide centers that strongly interact with each other through the TiO_2 semiconductor support.

Fig. 6 presents the effect of face velocity on the catalytic activity of VWT3. The face velocity here denotes the total gas flow rate divided by the out layer surface area of the filter element. And it is

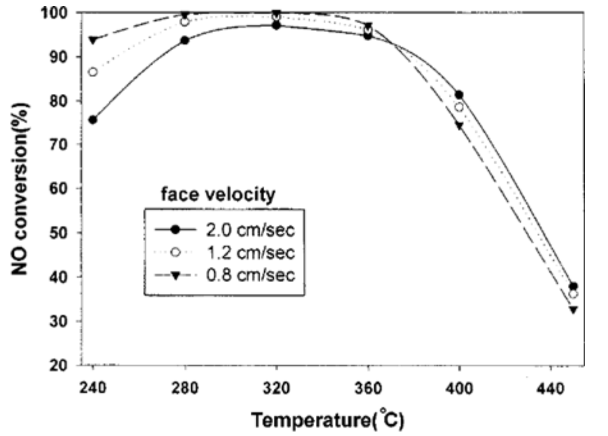

Fig. 6. The effect of face velocity on NO conversion over VWT3 catalytic filter.

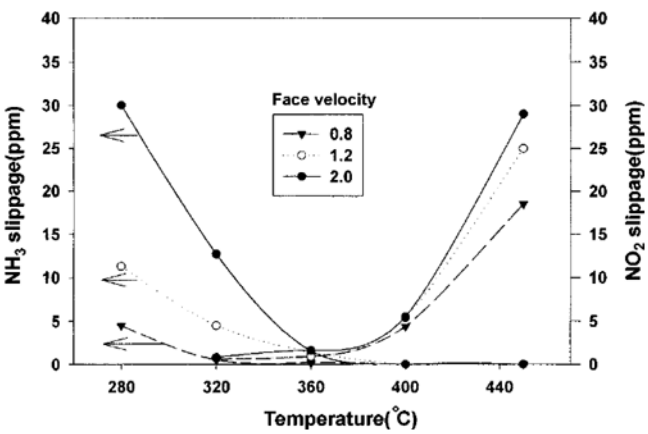

Fig. 7. The effect of face velocity on NH_3 and NO_2 slippage.

a very useful factor to evaluate the dimension of the filter unit. For general commercial filtration using a high temperature filter unit such as the integrated gasification combined cycle (IGCC), it is designed at the value of 0.01-0.03 m/sec, usually 0.012 m/sec. NO conversion over VWT3 reaches near to 100% when the face velocity is lower than 0.012 m/sec. And, in this condition, NH_3 and NO_2 slippages are less than 10 ppmv and 2 ppmv in the temperature range of 290-350 °C, respectively, as shown at Fig. 7. The catalytic filter also presents high activity of more than 95% of NO conversion when NO concentration is 700 ppm as shown at Fig. 4. Considering that NO conversion of currently operating SCR of plate-formed catalyst is about 80%, it is a very encouraging result that catalytic filters show such a high NO conversion.

CONCLUSIONS

In order to develop an effective catalytic filter based on V_2O_5/TiO_2 supported on a commercial high temperature filter element (PRD-66), the promoting effect by WO_3 was investigated.

PRD-66, known as a good filter element at high temperature, presented good properties for SCR catalyst carrier since it contains a great deal of active surface material such as Al_2O_3, SiO_2, and MgO whose contribution is remarkable. For additional catalyst supporter, TiO_2 particles were well distributed in the pores of PRD-66 with particle size of less than 1 μm and increased the surface areas to 1.38 from 0.1 m^2/g.

WO_3, in the V_2O_5-WO_3-TiO_2/PRD-66 catalytic filter system, increased the SCR activity significantly and broadened the optimum temperature window (from 30 to 60 °C) as the similar results reported for V_2O_5-WO_3-TiO_2 ternary catalytic system. The catalytic filter based on the PRD-66 shows the maximum NO conversion of more than 95% for NO concentration of 700 ppm at face velocity of 0.02 m/sec in the temperature range between 290 and 350 °C, which is very promising for the commercialization of this catalytic filter system.

REFERENCES

AlliedSignal, "Preliminary Engineering Data," AlliedSignal Composites Inc., P.O.Box 9559, Newark, DE 19714-9559 (1999).

Amiridis, M. D., Wachs, I. E. and Jehng, J. M., "Reactivity of the V_2O_5 Catalysts for the Selective Catalytic Reduction of NO by NH_3," *J. Catal.*, **161**, 247 (1996).

Alemany, L. J., Berti, F., Busca, G., Ramis, G., Robba, D., Toledo, G. P. and Trombetta, M., "Characterization and Composition of Commercial V_2O_5-WO_3-TiO_2 SCR Catalysts," *Applied Catalysis B: Environmental*, **10**, 299 (1996).

Alemany, L. J., Lietti, L., Ferlazzo, N., Forzatti, P., Busca, G., Giamello, E. and Bregani, F., "Reactivity and Physicochemical Characterization of V_2O_5-WO_3/TiO_2 De-NO_X Catalysts," *J. of Catalysis*, **155**, 117 (1995).

Bosch, H. and Janssen, F., "Catalytic Reduction of Nitrogen Oxides. A Review on the Fundamentals and Technology," *Catalysis Today*, **2**, 369 (1988).

Chae, H. J., Nam, I. S., Yang, H. S., Song, S. L. and Hur, I. D., "Selective Catalytic Reduction of NO_x by NH_3 over V_2O_5/Ti-PILC Catalyst," *HWAHAK KONGHAK*, **38**, 783 (2000).

Choi, E., Lee, J. K., Park, D. and Park, W. H., "Removal of SO_x and NO_x from Flue Gas with Ceria," *Korean J. Chem. Eng.*, **11**, 25 (1994).

Choi, J. H. and Ahn, G. H., "Preparation of Catalytic Filter for the Simultaneous Treatment of NO_x and Particulate," The 4th Intern. Symposium on Coal Combustion, Beijing, 197 (1999a).

Choi, J. H., Ahn, G. H. and Kim, S. S., "The Characteristics of NO Reduction over V_2O_5/TiO_2 Catalyst attached on Ceramic Filters," *J. of Korean Society of Environ. Eng.*, **21**(10), 1861 (1999b).

Choi, J. H., Keum, S. M. and Chung, J. D., "Operation of Ceramic Candle Filter at High Temperature for PFBC Application," *Korean J. Chem. Eng.*, **16**, 823 (1999c).

Choi, J. H., Kim, S. K., Ha, S. J. and Park, Y. O., "The Supporting of V_2O_5/TiO_2 Catalyst on the Ceramic Filter Candle for Selective Reduction of NO," International Symposium on Chemical Engineering, Feb. 8-10, Cheju Island, Korea, 234 (2001).

Choung, J. W., Choi, K. H., Seong, H. J., Chai, H. J. and Nam, I. S., "The Effect of HCl Gas on Selective Catalytic Reduction of Nitrogen Oxide," *J. of KSEE*, **22**(4), 609 (2000).

Forzatti, P. and Lietti, L., "Recent Advances in De-NO_x ing Catalysis for Stationary Applications," *Heterog. Chem. Rev.*, **3**(1), 33 (1996).

Gennrich, T. J., "Filter Bags Help Meet Particulate Control Standards," Power Engineering, 1 (1993).

Ham, S. W., Nam, I. S. and Kim, Y. G., "Activity and Durability of Iron-exchanged Mordenite-type Zeolite Catalyst for the Reduction of No by NH_3," *Korean J. Chem. Eng.*, **17**, 318 (2000).

Kudlac, G. A., Farthing, G. A., Szymanski, T. and Gortbett, R., "SNBR Catalytic Baghouse Laboratory Pilot Testing," *Environmental Progress*, **11**(1), 33 (1992).

Nam, I. S., "A Catalytic Process for the Reduction of NO_x from Stationary Sources," *Catalysis*, **11**(1), 5 (1995).

Parvulescu, V. I., Grange, P. and Delmon, B., "Catalytic Removal of NO," *Catalysis Today*, **46**, 233 (1998).

Saracco, G. and Montanaro, L., "Catalytic ceramic Filter for Flue Gas Cleaning. 1. Preparation and Characterization," *Ind. Eng. Chem. Res.*, **34**, 1471 (1995).

Shin, B. S., Lim, S. Y. and Choung, S. J., "WO_3 and MoO_3 Addition Effect on V_2O_5/TiO_2 as Promoters for Removal of NO_x and SO_x from Stationary Sources," *Korean J. Chem. Eng.*, **11**, 254 (1994).

Sohn, J. R. and Bae, J. H., "Characterization of Tungsten Oxide Supported on TiO_2 and Activity for Acid Catalysis," *Korean J. Chem. Eng.*, **17**, 86 (2000).

Terabe, K., Kato, K., Miyazaki, H., Yamaguchi, S. and Imai, A., *J. Mater. Sci.*, **29**, 1617 (1994).

Topsφe, N.-Y., Topsøe, H. and Dumesic, J. A., "Vanadia/Titania Catalysts for Selective Catalytic Reduction (SCR) of Nitric Oxide by Ammonia," *J. Catalysis*, **151**, 226 (1995).