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Pt-V_2O_5-WO_3/TiO_2 Catalysts Supported on SiC Filter for NO Reduction at Low Temperature

Joo-Hong Choi†, Jin-Hyun Kim, Young-Cheoul Bak, Rose Amal* and Jason Scott*

Department of Chemical and Biological Engineering/ERI, Gyeongsang National University,
900 Gazwa-Dong, Jinju 660-701, Korea
*School of Chemical Engineering and Industrial Chemistry, The University of New South Wales,
Sydney, NSW 2052, Australia

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Abstract−The catalytic filter, V_2O_5-WO_3-TiO_2 supported on a ceramic filter, is known as a promising material for treating particulates and NO_x simultaneously at optimum temperatures around 320 °C. In order to improve its catalytic activity at low temperatures, the effect of Pt addition on the catalytic filter has been investigated. Catalytic filters, Pt-V_2O_5-WO_3-TiO_2 /SiC, were prepared by co-impregnation of Pt, V, and W precursors on TiO_2 coated-SiC filter by vacuum aided-dip coating. The Pt-added catalytic filter shifted the optimum working temperature from 280-330 °C (for the non Pt-impregnated filter) to 180-230 °C, providing N_x slip concentration less than 20 ppm for the treatment of 700 ppm NO at a face velocity of 2 cm/s with the same value over the non Pt-added catalytic filters. The promotional effect following the addition of Pt is believed to result from electrical modification of the catalyst maintaining a high electron transfer state. Ammonia oxidation was also observed to be dominant above the optimal temperature for SCR.

Key words: Catalytic Filter, NO Reduction, Ammonia, Platinum Effect, Low Temperature, Ceramic Filter

INTRODUCTION

Catalysts based on vanadia-titania have been widely developed and are most successfully commercialized for the reduction of NO_x with ammonia [Alemany et al., 1995, 1996; Bosch and Janssen, 1988; Choi et al., 1994; Nam et al., 1995, 2000]. One of the critical issues regarding the commercialization of selective catalytic reduction (SCR) catalysts is related to ammonia slip in the system using ammonia as the reductant. NO reduction processes by hydrocarbon have been developed to overcome this problem. However, the NO reduction activity by the hydrocarbon is not satisfactory as the limited-value about 50% for the treatment of the considerable NO concentration using several catalysts [Kim et al., 1999; Lee and Rhee, 2002; Praserthdam et al., 2003]. Plugging the gaps of a monolith catalyst is also one of the serious operational problems that leads to the limited-application of SCR catalyst working at a dust-free stream only.

To overcome these problems, a highly effective catalytic filter in the form of rigid filter elements (such as ceramic filter candles) is one solution to prevent the plugging problem as it provides a low pressure drop system while maintaining control over particulate and nitric oxide emissions simultaneously [Saracco and Montanaro, 1995]. Advanced ceramic filter elements for particulate removal at high temperatures are typically comprised of two layers: an interior support and an exterior membrane layer. Most of the particles in the dusty stream are removed on the membrane layer whose thickness is about 100 μm. The interior layer is less useful for filtration purposes but acts mainly as a rigid support. The catalytic filter utilizes the large room afforded by the pores of the support layer, which generally possess a pore size of 100-200 μm. So the performance of a catalytic filter depends on how to load the catalyst effectively such that sufficient reaction sites are available. Recently, technologies have been successfully developed for coating the pore surface of ceramic filters with TiO_2 by using a centrifugal coating method [Choi et al., 2001a]. V_2O_5-WO_3-TiO_2/PRD catalytic filters prepared by this method provided high NO conversion greater than 99.9% (initial NO concentration=700 ppm) at gas face velocities of 1.2 cm/s over the temperature range 280 to 350 °C [Choi et al., 2001b].

Extending the function of the catalytic filter to the systems that operate at lower temperature is attractive for the treatment of ordinary flue gas streams. Supported transition metal oxides such as MnO_x/TiO_2 [Peña et al., 2004] and V_2O_5/AC [Zhu et al., 1999] showed high SCR activity at low temperatures (150-250 °C). Mn-Ce mixed-oxide catalysts showed superior SCR activity in the low-temperature region of 80-150 °C, and were substantially more active than all other catalyst reported in the literature and mildly affected by SO_2 and H_2O at high concentrations [Qi and Yang, 2003]. Ceria (CeO_2), which is considered one of the most important components of three-way catalysts, is known to act as an oxygen reservoir, storing and releasing oxygen via the redox shift between Ce^{4+} and Ce^{3+} under oxidizing and reducing conditions, respectively. Several inspiring reports illustrate the potential for adopting transition metals as the catalysts for NO reduction at low temperature. However, an alternative approach involves improvement of the current V_2O_5-WO_3-TiO_2-based catalyst, as its performance is already well proven in industry, by adding a promotional element for low temperature operation. This idea is derived from the example of ceria addition in three-way catalysts, which accelerates electron transfer in either direction in reactions involving a redox process. Pt was chosen in this study, as it is known to promote photocatalytic reactions involving TiO_2 by providing elevated redox properties [Siemon, 2002].

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†To whom correspondence should be addressed.
E-mail: jhchoi@gsnu.ac.kr

Electron transfer properties of TiO_2 increase with the loading of transition metal oxides such as V_2O_5 and especially with noble metals such as Pt and Pd [Linsebigler et al., 1995]. We believe the electron transfer properties of the metals are closely related to the activity of the SCR reaction with ammonia. The aim of this work is to investigate the promotion effect of Pt over the V_2O_5-WO_3-TiO_2-based catalytic filters for NO reduction.

EXPERIMENTAL

1. Preparation of the Catalytic Filters

TiO_2 powder (in the form of $TiO(OH)_2$) was purchased from Grand Chemical Ind. The TiO_2 powder was dispersed as the received form in 0.05 M HCl aqueous solution and stabilized for 1 hr under agitation. The cylindrical ceramic filter element, Dia Shumalith 10-20 (SiC) from Schumacher Umwelt und Trenntechnik, Germany, was used as the received. SiC is fabricated by the sintering of SiC powders using a special ceramic binder [Schulz and Durst, 1994]. It has a porosity of 38% and the pore size of the support layer is larger than 100 μm. It also has a membrane layer with a median pore size of around 10 μm. The candle-style filter elements possessed original dimensions of 1.5 m length and 4/6 cm inner/outer diameter. They were cut into 1 or 2 cm tubular slices in order to fit into in a small experimental reactor. The slices were cleaned in an ultrasonic water bath, treated in 0.05 M NaOH aqueous solution and dried at 110 °C for 2 hrs prior to loading with TiO_2.

TiO_2 particles as the catalyst support were coated on the pore surface of the ceramic filter elements by the vacuum aided-dip coating method. The filter element was first evacuated for 30 min at a vacuum of approximately 10^{-2} Pa and then soaked in an aqueous solution containing TiO_2 particles following introduction of the solution from the bottom of the vacuum chamber as shown in Fig. 1. The filter element was stabilized overnight under room conditions after being taken out from the solution and followed by drying for 1 hr at 110 °C and then treated in an air stream at 450 °C for 2 hrs. The TiO_2 loading on the filter elements was controlled by changing the TiO_2 concentration of the aqueous solution and measured by weighing the filter element pre- and post-coating. The TiO_2 loading on SiC filter is shown in Table 1.

To prepare the catalytic filter, the TiO_2 loaded-ceramic filter elements were impregnated in an oxalic acid solution of ammonium meta-vanadate (NH_4VO_3) and ammonium para-tungstate ($(NH_4)_{10}$

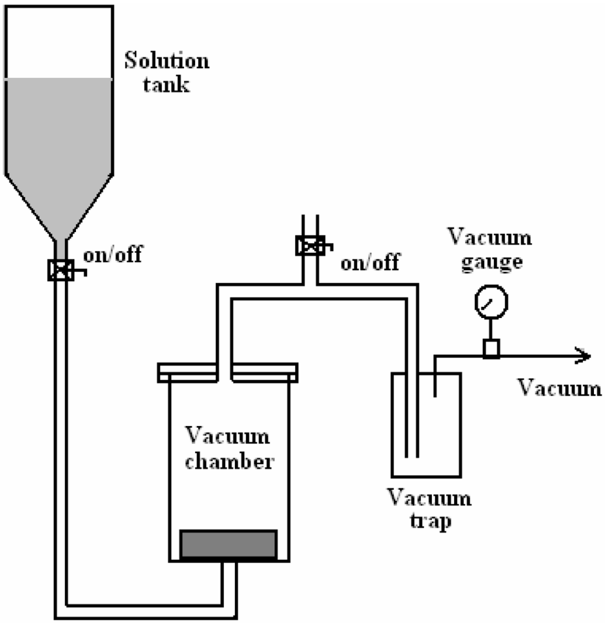


Fig. 1. Vacuum coating unit for coating of TiO_2 on the pore surfaces of ceramic filter.

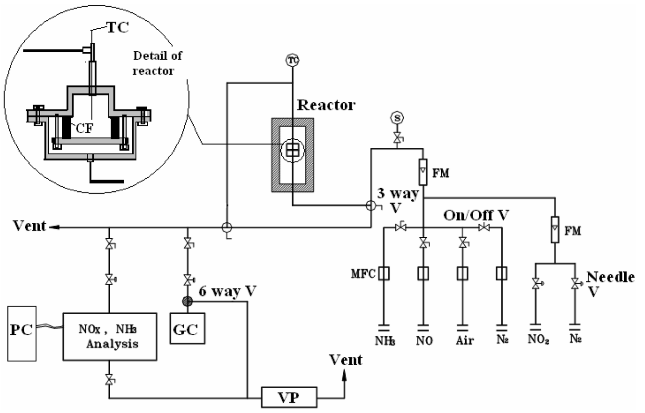


Fig. 2. Experimental unit for assessment of the catalytic filter.

CF: Ceramic Filter
PC: Personal Computer
FM: Flow Meter
TC: Thermocouple
GC: Gas Chromatograph
V: Valve
MFC: Mass Flow Controller
VP: Vacuum Pump

Table 1. Chemical composition and BET area of the catalytic filters prepared at different concentrations of TiO_2 and Pt

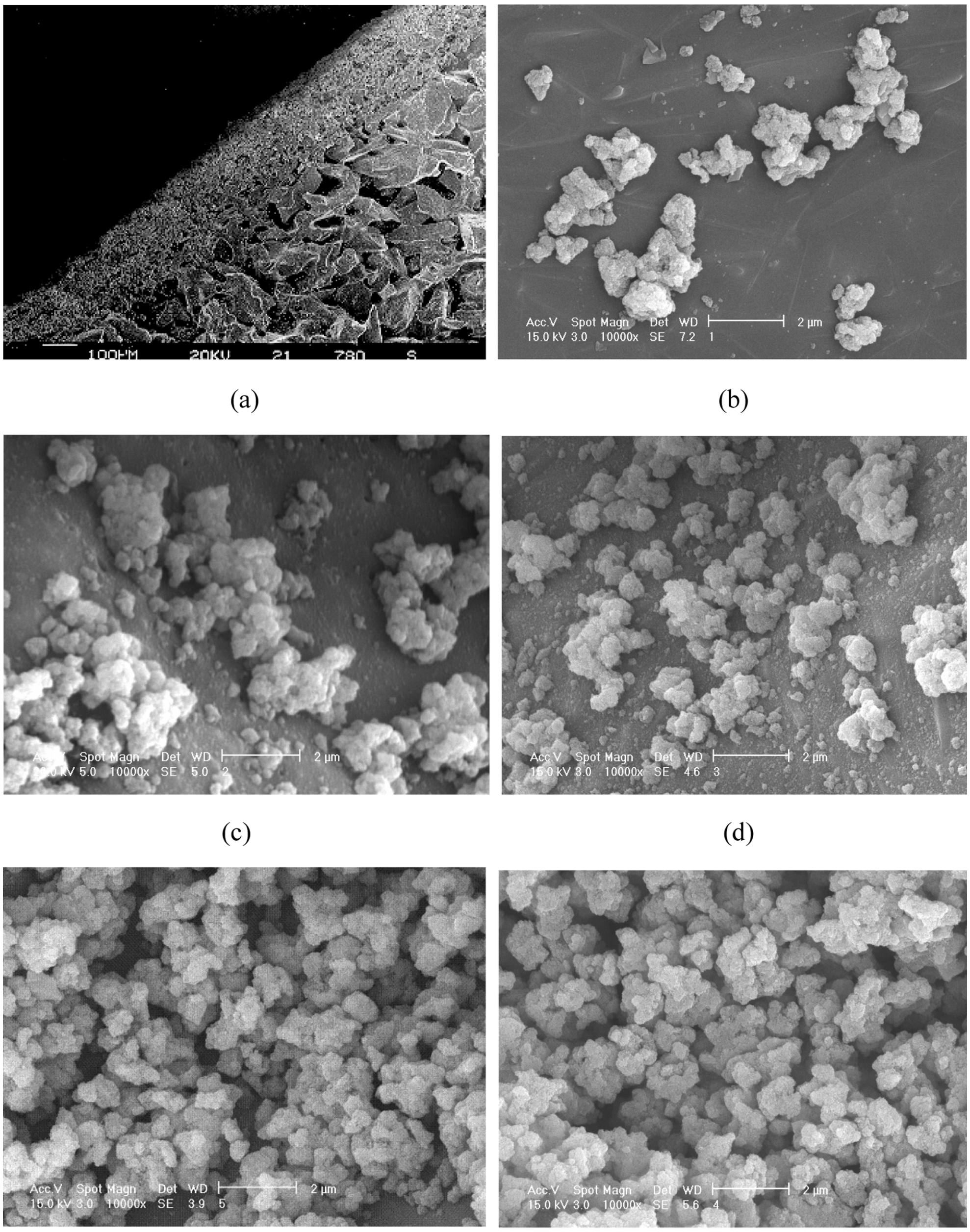
Sample	TiO_2 concentration (wt%)	TiO_2 load on SiC (wt%)	BET area of TiO_2 coated-SiC (m^2/g)	Pt load over V_2O_5 (wt%)
SiCT4	10	2.2	0.65	-
SiC1Pt	4	0.6	0.2	15
SiC2Pt	6	1.1	0.26	15
SiC3Pt	8	1.5	0.41	15
SiC4Pt (or SiCPt3)	10	2.2	0.65	15
SiC5Pt	12	2.5	0.67	15
SiCPt1	10	2.2	0.65	5
SiCPt2	10	2.2	0.65	10
SiCPt4	10	2.2	0.65	30

$W_{12}O_{41}$) in their desired-compositions. Platinum, in the precursor form of ammonium tetra-chloroplatinate(II) ($(NH_4)_2PtCl_4$), was added to the impregnation solution when required. The solution was slowly evaporated at 40 °C until the filter element absorbed all of the solution, followed by drying overnight at room temperature, 1 hr at 110 °C and then treated in an air stream at 450 °C for 2 hrs. The compositions of the fabricated catalytic filters are provided in Table 1. The TiO_2 composition of the catalytic filters in Table 1 denotes the weight percent based on the mass of the filter element. All the catalytic filters contain the same compositions of V_2O_5 and WO_3 (1 wt% and 8 wt% based on TiO_2, respectively), as this is known as the optimal composition for commercial SCR catalysts in the powder and honeycomb forms [Alemany et al., 1995, 1996].

The morphology and elemental compositions of catalyst supported on the filter elements were examined by using scanning electron microscopy (SEM) (accelerating voltage 20 KV) and energy-dispersive X-ray analysis (EDX) (JEOL JSM-6300), respectively, after the samples were sputtered with a thin layer of gold. The BET surface area was measured on the ASAP2000 utilizing sample masses of up to 5 g.

2. The Catalytic Activity of the Catalytic Filter for NO SCR

The catalytic activity was measured by the experimental unit shown in Fig. 2 by using a gas mixture of NO, NH_3, O_2, and N_2. In a standard test run, the concentrations of NO, NH_3, and O_2 were 700 ppm,



(a) (b)

(c) (d)

(e) (f)

Fig. 3. SEM images (multiplied by 10,000 except (a)) of: (a) filter cross-section showing titania particles supported on SiC: TiO_2 pore surface coverage following coating by aqueous solutions with TiO_2 loadings of (b) 4%; (c), 6%; (d) 8%; (e) 10% and; (f) 12%.

700 ppm, and 2 vol%, respectively, the remainder being N_2. The total gas flow rate was controlled with a mass flow controller to settle the actual face velocity at 2 cm/s. The catalytic filter was mounted in a reactor cell by using the assembled-flanges as shown by the detailed reactor schematic in Fig. 2. The reaction gases were introduced from an inlet at the base of the reactor and passed through the catalytic filter such that the direction of gas flow across the filter (inward from the outside surface) was the same as the normal industrial filtration mode for hot gas filtration using a ceramic filter element. The catalytic activity of the filter was measured at a series of steady state temperatures. The concentrations of NO, N_2O+NO_2, and NH_3 were continuously analyzed by a chemiluminescence method (TEI model 17, Thermo Environmental Instruments Inc.). With this instrument, NO concentration is measured first with the chemiluminescence of NO_2 formed from NO conversion. NO_x (NO+NO_2+N_2O) concentration is secondly determined with the conversion of these components into NO by a Mo converter working at 325 °C. Finally, N_x (NO_x+NH_3) is determined by the conversion of these components into NO by a stainless steel converter working at abot 825 °C. So NO_2+N_2O concentration is calculated by subtracting NO concentration from that of NO_x. The NH_3 concentration is calculated by subtracting NO_x concentration from the N_x concentration. Oxygen concentration was analyzed by using an off-line GC with TCD using a GS-Q capillary column.

RESULTS AND DISCUSSION

1. TiO_2 Coating on the SiC Filter

The pores of the SiC filter are formed by the sintering of irregular SiC particles with the assistance of a special binder with the resulting pore shapes irregular and possessing a wide size distribution, as shown in Fig. 3a. The upper-left layer in Fig. 3a is the membrane layer, having small pores of about 10 μm in diameter, with most of the filtration occurring in this layer. It appears that TiO_2 particles are rarely deposited in this layer, as the original TiO_2 particles tend to aggregate into large sizes mostly larger than 0.4 μm and subsequently find it very difficult to penetrate the small pores. Based on earlier gas-solid filtration studies [Shulz and Durst, 1994], the SiC filter element completely hindered the peneatration of particles larger than 0.4 μm on the membrane surface. However, the supporting layer enjoys the carrying of the TiO_2 aggregates as the pore size (mean pore size ~150 μm) is sufficiently large. The series of SEM images from Fig. 3b to Fig. 3f represent the morphologies of deposited TiO_2 particles on the surfaces of SiC pores, prepared from aqueous solutions containing different TiO_2 concentrations. The TiO_2 coverage of the sample prepared by using the dilute 4 wt% TiO_2 solution was very low as shown in Fig. 3b, possessing a TiO_2 loading of 0.6%, as shown in Table 1 (SiC1Pt). Fig. 3e shows the SiC surface is almost covered with TiO_2 particles when the TiO_2 load reaches 2.2 wt%. All the particles shown on the surfaces of the SEM images were confirmed as the TiO_2 particles by the EDX analysis (not shown the results here). At loadings greater than this, such as in the 2.5 wt% sample (Fig. 3f), the TiO_2 particles tend to aggregate with each other, forming multi-layers of particles. An increase in particle and pore sizes occurs, leading to a reduction in the surface area of the catalyst support. This detrimental aspect of the coating procedure was not addressed further in this paper, and developing procedures for preparing catalytic filters displaying higher surface areas is our next approach for further study.

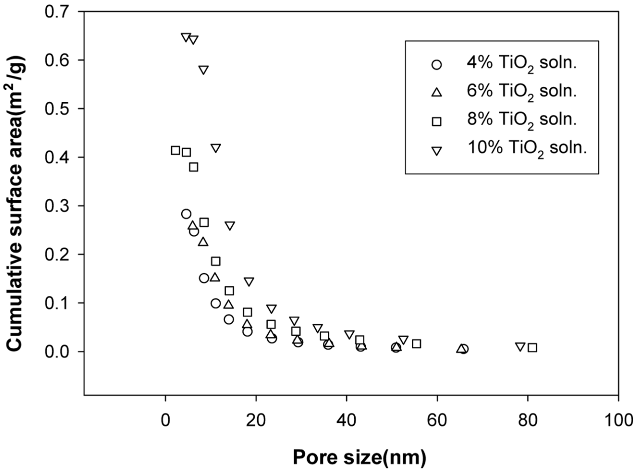


Fig. 4. Cumulative surface area of TiO_2 coated-SiC filter according to the change of pore size as the variation of TiO_2 concentration.

The BET surface area of the fresh SiC filter was 0.06 m^2/g. Fig. 4 presents the pore size distribution measured by BET adsorption by the denotation of cumulative surface area with the corresponding value of pore size. In respect of catalyst support, the pore size of supported-TiO_2 is important to allow room for space of supported-catalyst. It is uncertain, at the present time, what the proper size of pores as the catalyst carrier is. Considering the previous report [Perez et al., 2001] mentioned that platinum crystal grew up to 5 nm on membrane filter media, large pores of titania supported on SiC of more than 15 nm will be helpful to carry the SCR catalyst. So the cumulative surface area corresponding to the pore size of 15 nm is meaningful and increases considerably for the catalytic filter prepared with TiO_2 solution concentration of 10 wt% as we can see the value of 0.12 and 0.36 m^2/g for the cases of 8% and 10% TiO_2 solution, respectively.

2. Catalytic Activity of Pt-V_2O_5-WO_3-TiO_2/SiC Catalytic Filter

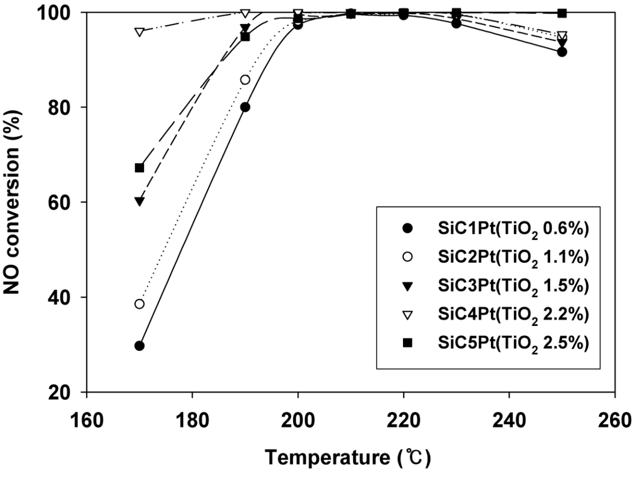


Fig. 5. NO conversion over Pt-V_2O_5-WO_3/TiO_2/SiC catalytic filters containing different TiO_2 loadings.

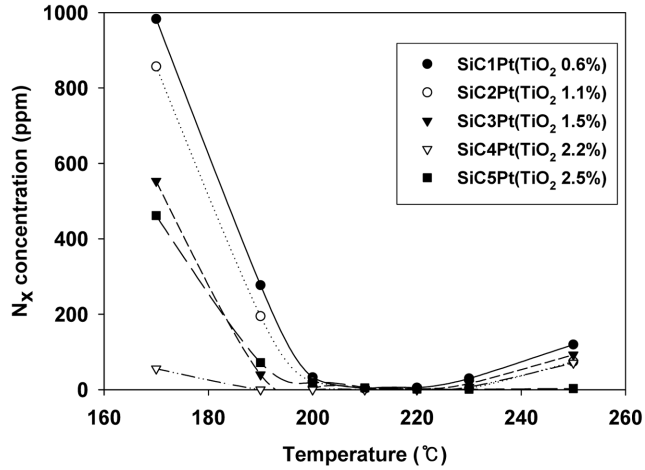


Fig. 6. N_x slip concentration over Pt-V_2O_5-WO_3/TiO_2/SiC catalytic filters containing different TiO_2 loadings.

Fig. 5 shows NO conversion over the catalytic filters V_2O_5-WO_3/TiO_2/SiC prepared with the different concentrations of TiO_2 for the coating solution (TiO_2 loadings of 0.6 to 2.5 wt% based on SiC) for the same Pt compositions of 15 wt% (based on V_2O_5). The result shows that SiC4Pt, prepared by the 10 wt% TiO_2 solution, exhibits the best catalytic activity showing the highest NO conversion and the broadest temperature window for conversion greater than 99.5%. SiC4Pt is also the most selective towards N_2 as shown in Fig. 6. Fig. 6 presents the N_x slip concentration in the discharge gas from the reactor for the same catalytic filters corresponding to the same reaction conditions in Fig. 5. N_x concentration includes the summation of NH_3, NO, N_2O, and NO_2 concentrations and represents the total pollution concentration sourced by N-components in the SCR reaction. N_2 selectivity by the N_x slip concentration serves to highlight a major pollution problem prevalent in the SCR system.

Generally, there is no universally set criterion for an environmentally acceptable N_x slip concentration value as it differs from system to system and is dependent on the national policy of individual nations. As one example, from 2008 onwards, the National Ambient Air Quality Standards (NAAQS) of the United States of America will require source emission reductions of NO_x to levels below 0.15 lb/10^6 Btu, which is equivalent to NO_x emissions less than 108 ppm from a coal power plant using Pittsburgh #8 coal [DOE of US, 2003a]. Furthermore, the New Source Performance Standards require NO_x emission rates to be 0.072 lbs/10^6 Btu, which is equivalent to NO_x emission less than 50 ppm for a coal power plant when using Pittsburgh #8 coal [DOE of US, 2003b]. Therefore, the limit value of 10 ppm for N_x slip concentration in the experimental unit is strictly acceptable to mention the SCR activity of developing catalytic filter regarding to the commercialization.

Fig. 5 also denotes the activation temperatures were shifted towards lower temperatures as TiO_2 loading increased, with SiC4Pt displaying the best activity and the lowest N_x slip concentration (Fig. 6). SiC5Pt exhibited a lower activity than SiC4Pt in spite of higher TiO_2 loading. This outcome highlights a limitation of the vacuum coating method, and indicates an alternative method is required in order to obtain a high TiO_2 surface area in the pores of the ceramic filter. By vacuum aided-coating, the initial vacuum effect will be dominant until the pores of the ceramic filter are occupied with the solution. However, after that time, there will be no more effect to keep the driving force for the particle migration into the inner sites of the ceramic filter because the pore cavities are already full with solution. So the extra activity of coating time is consumed to concentrate the coating development on the outer surface of ceramic filter only. Of the samples considered, SiC4Pt was the most effective catalyst for NO_x reduction. So the TiO_2 coating condition for SiC4Pt was used for preparation of catalytic filters containing platinum for the further studies. Subsequently, as the optimum coating condition, TiO_2 loading of 2.2 wt% was selected for the study of Pt promotion effect.

Figs. 7 and 8 compare NO conversions and N_x slip concentrations, respectively, over Pt-V_2O_5-WO_3-TiO_2/SiC catalytic filters (containing the Pt loadings of 5 to 30 wt% based on V_2O_5) with the same composition of SiCT4 for other components. The best catalytic filter, SiCPt3 (containing 15 wt% Pt), exhibited the best NO conversion greater than 99.8% and the lowest N_x slip concentration less than 10 ppm over the temperature range 180 to 230 °C. SiCPt4 (con-

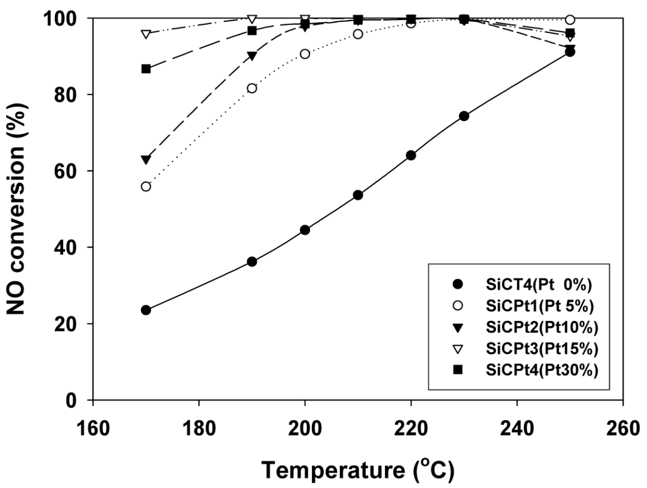


Fig. 7. NO conversion over Pt-V_2O_5-WO_3/TiO_2/SiC catalytic filters containing different Pt loadings.

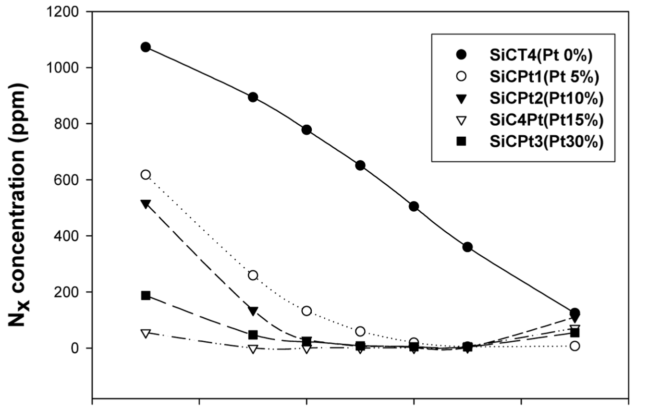


Fig. 8. N_x slip concentration for Pt-V_2O_5-WO_3/TiO_2/SiC catalytic filters containing different Pt loadings.

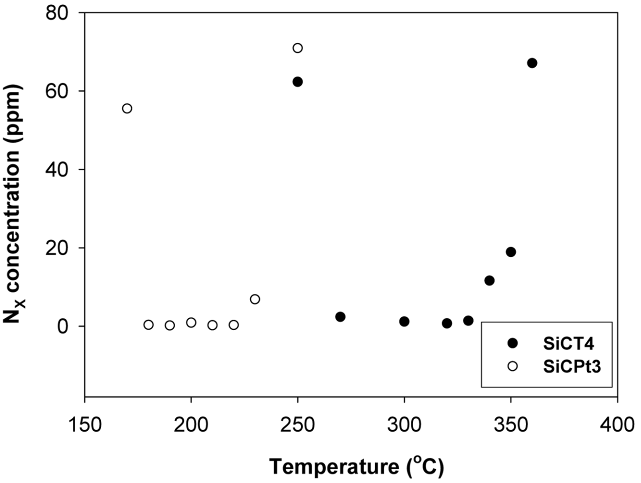


Fig. 9. Temperature effect on the N_x slip concentration over the catalytic filters with Pt (for SiCPt3) and without Pt (for SiCT4).

taining 30 wt% Pt) revealed a slightly lower NO conversion and higher N_x slip concentration at both low and high temperatures, compared to SiCPt3, which implies the existence of optimum Pt particle size for the best SCR performance. When the metal size becomes large, it is the general case that the reducibility of the metal increases as well as with the diminution of the active sites. The oxidation occurs easily on the metal surface [van den Broek et al., 1999], which leads the decrease of NO reduction in SCR.

Fig. 9 shows the N_x slip concentration over the catalytic filters with Pt (SiCPt3) and without Pt (SiCT4) in the extended-temperature range and presents the general trend of Pt addition effect to V_2O_5-WO_3-TiO_2/SiC catalytic filters for NO reduction. Pt addition consistently shifts the active temperature towards a lower temperature range (180-230 °C) with respect to catalytic filter without Pt (280-330 °C). This result indicates Pt reduces the activation energy of the SCR reaction and transfers the working temperature toward a lower value. The 15 wt% Pt loading of the catalytic filters in this study is based on the V_2O_5 loading and correspondent to 0.15 wt% based on TiO_2 because the V_2O_5 loading is 1 wt% based on TiO_2. It is apparent that such a small loading of Pt will cover only a minute percentage of the TiO_2 surface when previous report [Zhu et al., 1999] guessed the result that Pt dose of 10 wt% covers 6% of the TiO_2 surface. However, catalytic activity of the V_2O_5-WO_3-TiO_2/SiC catalytic filters is considerably promoted by the addition of such small amounts of Pt. In the context of electronic modification, the effect of tungsten oxide on the system is relatively small because the energy band gap and band position are similar for TiO_2 and WO_3 [Linsebigler, 1995]. Paganini et al. [1997] reported that WO_3/TiO_2 (for a WO_3 dose up to 9 wt%) showed similar electronic properties to TiO_2 alone. However, the inclusion of V_2O_5 (even at 1 wt% based on TiO_2) has enough impact to reduce the electron transfer energy with respect to TiO_2. Linsebigler et al. [1995] reported that a small amount (0.3-1.3 wt%) of vanadium present in V_2O_5/TiO_2 systems was preferentially reduced with respect to TiO_2. In case of Pt dosing, the TiO_2 surface is strongly modified by the formation of a Schottky junction between Pt and TiO_2 [Sanchez and Lopez, 1995; Simon et al., 2002], where the electrons are trapped in the Pt particle, resulting in the reduction of the band gap energy and the promotion of electron transfer between the reactants and surface sites. This kind of Pt promotion effect on Pt/TiO_2 catalysts has been well described for photo catalytic reaction systems that have been understood as being carried out by the redox process [Bruck et al., 1993; Simon et al., 2002].

Regarding the NO reduction mechanism with NH_3, there are a number of mechanistic proposals [Busca et al., 1998], which in some instances contradict each other. Almost all authors agree that the SCR involves a strongly adsorbed activated NH_3 species and a gas-phase or weakly adsorbed NO species. Moreover, there is little doubt the reaction proceeds via a "redox" process and requires "acid" sites where the adsorbed ammonia is activated. There remains debate on whether the acid sites are Brönsted and/or Lewis types or single or multiple sites. Irrespective of the type of active sites, it is evident that NO reduction with ammonia is deeply related to electron transfer or oxygen mobility. The electron transfer property seems to be strongly promoted by the presence of Pt on the catalysts.

In terms of SCR selectivity, the primary reason for the decrease in NO conversion and increase in N_x slip concentration (in Fig. 8 and Fig. 9) at high temperatures with Pt addition is believed to be that Pt stimulates the oxidation properties of the catalytic filter, in turn increasing NH_3 oxidation to N_2O, NO or NO_2 and NO oxidation to NO_2. Ammonia oxidation competes with SCR during NO reduction in the presence of ammonia. In the case of V_2O_5 crystallites, ammonia oxidation occurs above 400 °C when the vanadium reoxidation rate increases [Jung and Grange, 2002] in the normal SCR gas composition. Jung and Grange [2002] reported that V-OH groups (which are present below 300 °C and are responsible for SCR) were completely reoxidized to V=O and the reaction between structural oxygen and dissociated ammonia species was the main reason for ammonia oxidation to NO. Blanco et al. [2000] reported that, over a V_2O_5/TiO_2 monolith, N_2O did not form in the SCR reaction until NO_x conversion reached 80% in the temperature range 180 to 230 °C. They explained the reason to be due to the impedance of N_2O formation when NO is present in the reaction gas as NO effectively competes with the NO_2 reaction path (responsible for the formation of N_2O). Suárez et al. [2002] reported that NO oxidation over V_2O_5/TiO_2 catalyst did not appear until a temperature of 450 °C but occurred over CuO-NiO/TiO_2 catalyst from 200 °C upwards. They observed a similar effect during the SCR reaction whereby the threshold temperature of N_2O formation was 180 °C for the CuO-NiO/TiO_2 catalyst and 300 °C for the V_2O_5/TiO_2 catalyst. They also found ammonia oxidation activity linearly increased with a decrease in the initial reduction temperature of the catalyst. The initial reduction temperature of CuO-NiO/TiO_2 catalyst was 80 °C, while that of V_2O_5/TiO_2 catalyst was 280 °C. Li and Armor [1997] reported that N_2O formation over V_2O_5/TiO_2 catalyst appeared at 350 °C, while it appeared at 200 °C for PtO/Al_2O_3 catalyst.

At low temperature less than 200 °C, platinum has been used as an effective catalyst for the selective oxidation of ammonia to N_2 [van den Broek et al., 1999]. On Pt sponge [Sobezyk et al., 2004], N_2O was the dominant product during ammonia oxidation between 50 and 100 °C, without NO formation at these temperatures. NO was only observed to form at temperatures greater than 300 °C. Pt/TiO_2 (P-25) decomposes ammonia at 170 °C to N_2 in an environment containing water and oxygen with slight formation of NO_2^- and NO_3^- (3-4 ppm for an initial 1,000 ppm concentration and 95%

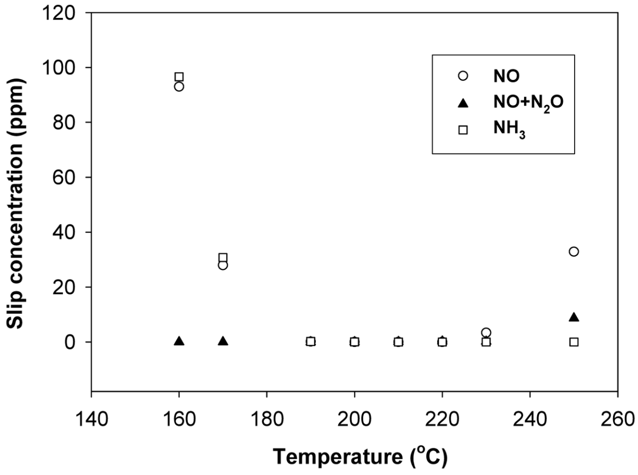


Fig. 10. Slip concentration of hazard components during NO SCR over SiCPt3 catalytic filter.

conversion) [Taguchi and Okuhara, 2004]. Pt/Al$_2$O$_3$ (1.2 wt%) showed 100% conversion with 87% selectivity to N$_2$ at 200 °C while V/Al$_2$O$_3$ (10 wt%) showed 45% ammonia conversion and 20 ppm N$_x$ slip concentration at 400 °C (for 1.14 vol% ammonia oxidation) at the same reaction conditions [Gang et al., 2000]. Long and Yang [2002] reported that Pt on ZSM-5 promoted the selective catalytic oxidation of ammonia to N$_2$ with the trend of selectivity decreasing as temperature rose above 200 °C.

Considering the results mentioned above, for SCR catalyst acting at low temperatures, Pt in particular displays a higher activity towards oxidation than V$_2$O$_5$. Similarly, for a catalytic filter containing Pt, NO conversion and N$_x$ slip concentration dramatically decrease after 250 °C as the increase of NO$_2$ formation as illustrated in Fig. 10. This occurs as oxygen mobility increases by the presence of metals on the catalyst, resulting in a shift of ammonia oxidation temperatures towards lower values.

CONCLUSIONS

The experimental investigation on the promotion effect of Pt addition over V$_2$O$_5$-WO$_3$-TiO$_2$/SiC catalytic filters for NO reduction with ammonia was carried out in an experimental unit. The catalytic filters were prepared by the impregnation of a TiO$_2$ coated-SiC ceramic filter with SCR catalysts using the solution of V, W, and Pt precursors.

TiO$_2$ was effectively coated on the pore surfaces of the ceramic filter (SiC) using the vacuum aided-dip coating method. Optimal TiO$_2$ loadings were identified as 2.2 wt% (based on filter mass) when the concentration of TiO$_2$ solution is 10%. The Pt optimum loading was found to be 15 wt% based on V$_2$O$_5$ which corresponded to a composition of 1.0 wt% based on TiO$_2$ mass.

The presence of Pt in the catalytic filter acted to shift the optimum working temperature (giving an N$_x$ slip concentration below 10 ppm at a face velocity of 2 cm/s) towards lower temperatures: 180-230 °C for V$_2$O$_5$-WO$_3$-TiO$_2$/SiC catalytic filters with Pt compared to 280-330 °C for the catalytic filter without Pt. Pt addition promoted catalytic activity at low temperatures but increased ammonia oxidation properties. This effect of Pt over the V$_2$O$_5$-WO$_3$-TiO$_2$/SiC catalytic filters is believed to be the reason that Pt increases the electron transfer properties of a catalyst system.

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