

Wire-mesh honeycomb catalysts for selective catalytic reduction of NO with NH₃

Jie Yao*, Jin Seong Choi**, Kyung Shik Yang**, Dezhi Sun* and Jong Shik Chung*^{*,**,†}

*School of Municipal and Environmental Engineering, Harbin Institute of Technology, Harbin 150090, P. R. China

**Department of Chemical Engineering and School of Environmental Engineering,
Pohang University of Science and Technology, Pohang 790-784, Korea

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Abstract—Both flat and corrugated wire mesh sheets were coated with aluminum powder by using electrophoretic deposition (EPD) method. Controlled thermal sintering of coated samples yielded uniform porous aluminum layer with a thickness of 100 μm that was attached firmly on the wire meshes. Subsequent controlled calcination formed a finite thickness of Al₂O₃ layer on the outer surface of each deposited aluminum particles, which resulted in the formation of Al₂O₃/Al double-layered composite particles that were attached firmly on the wire surface to form a certain thickness of porous layer. A rectangular-shaped wire-mesh honeycomb (WMH) module with triangular-shaped channels was manufactured by packing alternately the flat sheet and corrugated sheet of the Al₂O₃/Al-coated wire meshes. This WMH was further coated with V₂O₅-MoO₃-WO₃ catalyst by wash-coating method to be applied for the selective catalytic reduction (SCR) of NO with NH₃. With an optimized catalyst loading of 16 wt%, WMH catalyst module shows more than 90% NO conversion at 240 °C and almost complete NO conversion at temperatures higher than 300 °C at GHSV 5,000 h⁻¹. When compared with conventional ceramic honeycomb catalyst, WMH catalyst gives NO conversion higher by 20% due to reduced mass transfer resistance by the existence of three dimensional opening holes in WMH.

Key words: EPD Method, Al/Al₂O₃ Layer, WMH, SCR of NO, NH₃, V₂O₅-WO₃/TiO₂ Catalyst

INTRODUCTION

NO_x emitted from automobiles and stationary sources such as oil- and coal-fired power plants, waste incinerators, industrial ovens, and chemical process plants is a major cause for acid rain, photochemical smog and ozone depletion [Rucas and Brown, 1982]. Intensive regulations of nitrogen oxide emissions have prompted development of many control technologies [Kim et al., 1987; Choi et al., 1996; Park et al., 2002; Yoo et al., 2003; Chung et al., 2003; Lee et al., 2001]. So far the most attractive technology is selective catalytic reduction (SCR) of NO_x with NH₃, based upon a reaction sequence involving the combination of equimolecular amounts of NO and NH₃ in the presence of oxygen to produce water and nitrogen.

A great variety of catalyst formulations have been claimed to be active for this process [Lee et al., 2002; Kim et al., 2001; Bosch et al., 1986; Went et al., 1992; Lintz and Turek, 1992], including supported noble metals, mixed oxides, and zeolite-based materials. Vanadia is known to be most active but also active for undesired oxidation of SO₂ to SO₃, and accordingly its content must be kept low [Rajadhyaksha et al., 1989; Chen and Yang, 1990; Handy et al., 1992; Weng and Lee, 1993; Topsoe et al., 1995; Dutoit et al., 1997]. The addition of WO₃ or MoO₃ acting as chemical and structure promoters is expected to improve mechanical, structural, thermal and morphological properties of the catalysts and retard the oxidation of SO₂ to SO₃, enlarging the temperature window of the SCR reaction [Amiridis et al., 1999; Piehl et al., 1999; Long and Yang, 2000; Lee et al., 2002; Chae et al., 2004]. Considerable progress has been made for the understanding of the SCR with ammonia over V/W(Mo)/

Ti/O catalysts, including catalyst preparation, reactor design and the elucidation of the reaction mechanism [de Boer et al., 1993; Liuqing et al., 2003].

Because of high-pressure drop during the gas passage through the catalytic bed, it is necessary to use structured catalyst beds for practical environmental applications [Cybulski and Moulijn, 1998]. It is usually shaped as monolithic honeycombs that are commonly composed of ceramic materials or as parallel passage reactors that are manufactured by packing flat sheet and corrugated sheet of a substrate alternatively. Catalytic component is introduced onto the structured substrate surface either by wash-coating or by being incorporated into the channel walls during extrusion of the monolith structure. Main drawbacks of these channeled structures are their low inter-phase mass transfer rates, suppressed radial mixing due to impermeable channel walls and the accumulation of wash-coating in the corners of the channels. They are especially weak for dust-laden gases such as emission gases from coal boilers because blocking of channel opening at the front part by dust can make its real part totally ineffective.

Metal wire mesh honeycombs (WMH) [Kim et al., 2002] manufactured by packing flat sheet and corrugated sheet of wire meshes could be alternative choices for dust-laden gases. This kind of WMH design permits free radial flow because the channel walls have three-dimensionally connected openings, providing a non-blocking network. This will permit a rapid mixing of the gas flow and enhance inter-phase mass transfer rate to reduce reactor size [Jiang et al., 2003]. WMH is especially suitable for treating dust-laden exhausts, since even if the front channel is blocked by dust its rear part will remain effective, which will extend the operational life of the monolith. Also, during wash-coating of catalyst powder, the catalyst powder can have more uniform coating. In this case, its effectiveness

[†]To whom correspondence should be addressed.

E-mail: jsc@postech.ac.kr

factor can be kept higher than that of conventional ceramic honeycomb in which catalyst is accumulated on the corner of channel wall [Jiang et al., 2003].

The main drawback of this type of WMH is the difficulty to coat catalyst powder to the smooth and round surface of thin wire mesh. Conventional wash-coating might not be effective and catalyst may flake away upon mechanical or thermal shock. Plasma spray method was applied to deposit catalyst powder on the metal surface to improve mechanical strength and thermal stability of the coated layer [Ahlstrom-Silversand and Odenbrand, 1997]. By employing electrophoretic deposition method, Yang et al. succeeded in depositing aluminum powder as catalyst support layer that can be firmly attached on thin wire-mesh substrates [Yang et al., 2003]. They then formed thin Al₂O₃ layer on the outer surface of each Al particles to make double-layered Al₂O₃/Al composite particles by means of controlled calcination process. This coating layer was very stable upon thermal or mechanical, yet porous and rough on the surface so that catalyst powder could be attached via conventional wash-coating. WMH coated with V₂O₅/TiO₂ catalyst was successfully applied to catalytic combustion of ethyl acetate and catalytic oxidation of 1,2-dichlorobenzene (*o*-DCB) [Chung et al., 2002; Yang et al., 2004]. The choice of mesh material depends on the reaction conditions, and can range from the cheapest carbon steel to more expensive stainless steel, or to refractory alloys for catalytic combustion at high temperatures.

The aim of this work was to investigate application possibility of the WMH catalyst module for the selective catalytic reduction of NO_x with NH₃. Stainless steel meshes that were pre-coated with aluminum powder were used to make WMH modules. Commercial V₂O₅-WO₃/TiO₂ catalyst was coated via wash-coating method. Catalytic activities were tested at various catalyst loading and reaction conditions. The results were compared with those obtained with ceramic honeycomb module with similar geometric surface area (after coating) and catalyst loading.

EXPERIMENTAL

1. Preparation of the WMH Catalyst

A commercial stainless steel wire mesh (SUS 316L, 20 Taylor mesh screen) was used as substrate. It weights approximately 0.0556 g/cm² and has a hole opening size of approximately 0.9 mm and a wire diameter of 0.27 mm. Wire mesh was pretreated with 10 wt% H₂SO₄ solution for 2 min at 80-90 °C to remove dirt and grease. Aluminum powder (99% purity, spherical, Johnson Co.) material that was used for deposition has a mean particle size of 3 μm and a specific surface area of 1.83 m²/g. Polyacrylic acid and aluminum isopropoxide (Aldrich) were used as additives to improve the adhesion of aluminum particles and to control the suspension conductivity, respectively. First, a suitable amount of additives (1.0-5.0 × 10³ M) was added to ethanol (99.9%, Aldrich) under stirring, and then aluminum powder (about 0.5 wt%) polished with 1 wt% NH₄OH solution was mixed to the suspension under magnetic stirring.

The set-up used for the electrophoretic deposition is shown in Fig. 1. For particle deposition at the cathode, the wire mesh was cut into pieces (3.5 cm × 10 cm). The stainless steel plate was used as an anode and the size was the same as that of the cathode. The distance between the two electrodes was kept at 10-20 mm. The

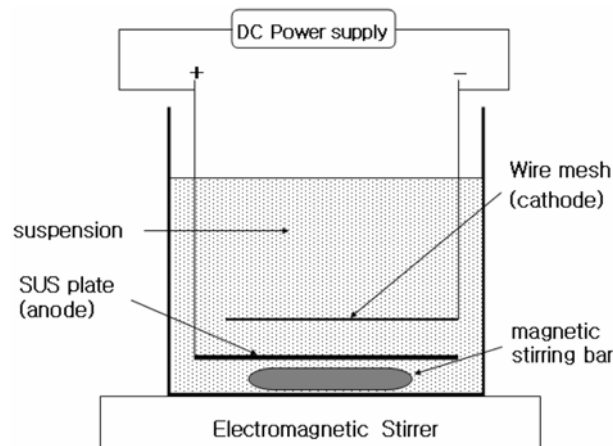


Fig. 1. Schematic diagram of EPD set-up.

DC voltage was kept at 100 V. The deposition time was kept for 1-2 min for flat sheets and for 3-4 minutes for the corrugated sheets. The aluminum-coated samples were dried at room temperature for 12 h, sintered at 800 °C for 10 h with He, and finally calcined at 500 °C for 5 h.

For making honeycomb-type catalyst module, two types of Al₂O₃/Al-coated wire mesh sheets (flat and corrugated type) were packed alternately to a final thickness of approximately 3.5 cm. The honeycomb module manufactured this way has a rectangular shape, 3.5 × 3.5 cm in width and 5.0 cm in length. It, therefore, has parallel passage channels along the longitudinal direction, shaped as an equilateral triangle with a side length of approximately 4 mm. From inherent properties of wire meshes, it also has opening holes (0.8 mm in size) perpendicular to longitudinal direction. This WMH module weighs 26.8 g, and the number of cells per square inch (cps) is 30 with a geometric surface area of 830 m² m⁻³. For comparison of catalytic activity, a conventional honeycomb catalyst module made of ceramic (cordierite) was also prepared. A ceramic honeycomb (CH in abbreviation) having channels of square cross-section (3 × 3 mm in channel size, 0.5 mm in wall thickness) was cut to a size of 3.5 × 3.5 cm in width and 5.0 cm in length. The CH contains 50 cells per square inch (cps) with a geometric surface area of 980 m² m⁻³, both of which were slightly higher than those of WMH.

V₂O₅-WO₃/TiO₂ catalyst was prepared by impregnating commercial TiO₂ support (Junsei Co., anatase form) with an aqueous solution of oxalic acid and ammonium metavanadate (NH₄VO₃) (KN, Aldrich Korea) to the incipient wetness. The samples were dried at 100 °C for 12 hours, then calcined at 500 °C for 5 h. The obtained powder was then impregnated again with an aqueous solution of citric acid and ammonium metatungstatehydrate ((NH₄)₆H₂W₁₂O₄₀) (JU, Aldrich Korea). Impregnated TiO₂ was dried at 100 °C for 12 hours and calcined at 500 °C for 5 hours. The weight percentages of V₂O₅ and WO₃ in the obtained catalyst were 2% and 6%, respectively. V₂O₅-WO₃/TiO₂ catalyst was coated onto the Al-Al₂O₃-coated WMH and CH modules by dipping them into a slurry solution which contains V₂O₅-WO₃/TiO₂ catalyst, 12.5 wt% colloidal silica (Aldrich, LUDOX AS-30) and 1 wt% PVA. The wash-coating was repeated to increase the loading of catalyst to the surface of the honeycomb module to a desired level. They were then dried at 100 °C for 12 hours and calcined at 500 °C for 5 hours.

2. Characterization of the Catalyst

The specific surface area was determined by nitrogen adsorption in a constant volume adsorption apparatus (Micrometrics, ASAP 2021C). X-ray diffraction (XRD) patterns of coated-samples in the present study were observed by M18XHF (Mac Science Co.) diffractometer employing CuK α radiation ($\lambda=1.5405 \text{ \AA}$) as X-ray gun. The diffraction patterns in the regions of $2\theta=10\text{-}90^\circ$ were usually examined with the scanning speed of $4.0^\circ/\text{min}$, operating at 40 kV and 200 mA of X-ray gun. The compound identification of XRD patterns was accomplished through the comparison of measured spectra with JCPDS file data. The surface structure and the cross section of coated wire were studied by means of scanning electron microscopy (SEM, Hitachi, S-2460N). The morphology and elemental distribution of the coated-samples were investigated by Philips SEM 515 equipped with an EDS detector and Hitachi HSM-2B Electron Probe Micro-analyzer with about 5.3 eV beam energy. X-ray photoelectron spectroscopy (XPS, VG Scientific, ESCA Lab. 220XL) was used for determining the surface structure and the phase of the coated samples.

3. Activity Tests on SCR of NO with NH₃

SCR activity test was carried out in a continuous flow reaction system, which was equipped with a square-shaped reactor chamber (3.5 cm \times 3.5 cm in width) of length 100 cm as shown in Fig. 2. Honeycomb catalyst module was located at a position 20 cm above the bottom of the reactor chamber. The bed was heated in two sections, each having its own heating furnace and controlling point. The line section before the reactor was also heated at temperature around 20-50 °C higher than the reaction temperature. The internal temperature just above the module was monitored continuously and maintained by adjusting the temperature of the preheating zone. NO gas (9.98%, N₂ balanced, DeokYang, SeoSan, Korea), and NH₃ gas (4.09%, %, N₂ balanced, DeokYang, SeoSan, Korea), measured by mass flow controller (MFC), were mixed with air and introduced into the reactor with NH₃/NO ratio at 1 : 1. A specially designed syringe was used to control the moisture inside the reactor. The gas hourly space velocity (GHSV, [1/h]) was expressed by the ratio of the gas flow rate to the volume of the WMH catalyst. The

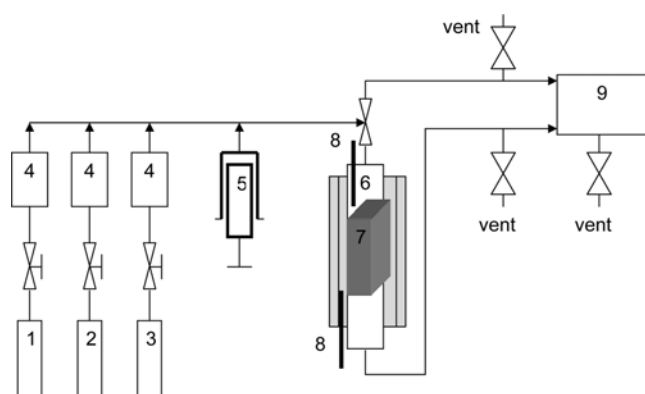


Fig. 2. Schematic diagram of SCR reactor system.

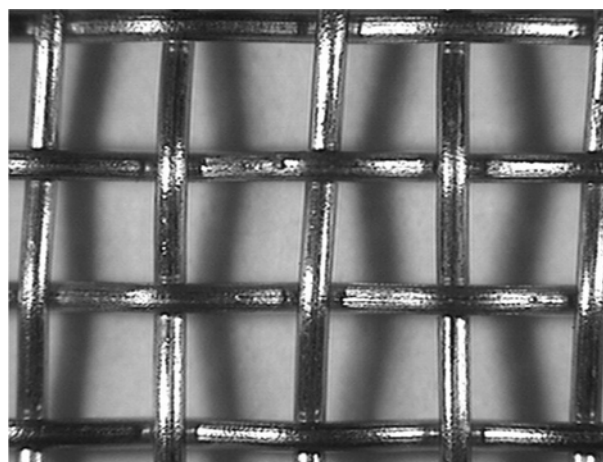
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|------------------------------------|------------------------------------|
| 1. NO+N ₂ | 6. Reaction chamber |
| 2. NH ₃ +N ₂ | 7. Catalyst module |
| 3. Gas cylinder (air) | 8. Thermocouple |
| 4. Mass flow controller | 9. NO and NO _x analyzer |
| 5. Moisture controller | |

inlet and outlet concentration of NO from reactor was analyzed by a chemiluminescent NO-NO_x analyzer (Thermo Environmental Instruments Inc., Model 42C). All of the stream lines were kept approximately 110 °C to prevent condensation of water and dissolution of NH₃ in water.

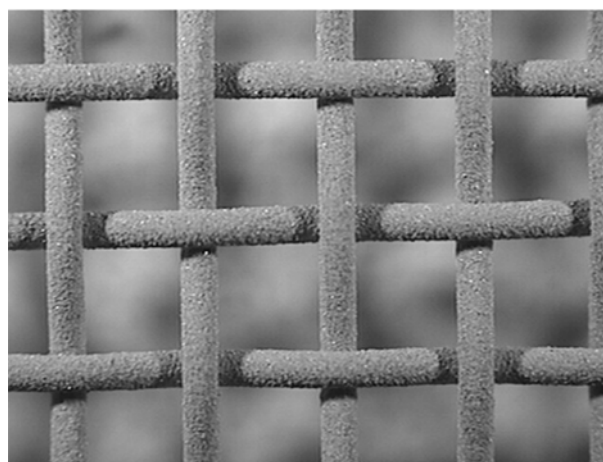
RESULTS AND DISCUSSION

1. Coating of Al₂O₃/Al and Catalyst Layer

Fig. 3 shows the surface structures of acid-cleaned bare wire mesh (3a) and aluminum-deposited wire-meshes (3b) that were measured by using a microscope. The coated layer is uniform in thickness regardless of position located in the wire meshes. In contrast to the smooth surface of bare wire meshes, the surface becomes highly rough and must be porous. The Al-coated wire-meshes were thermally treated with He gas purge for particle sintering. Al coatings are easily detached from the metallic substrate when the sintering temperature is less than 600 °C, since it is difficult for aluminum particles to be sintered at such low temperatures. It was shown that, upon ultrasonic vibration, the detached amount of the Al coatings is about 2 wt% at 750 °C. At 900 °C, coated Al particles were hardly



(a)



(b)

Fig. 3. Pictures of bare wire mesh (a) and Al-coated wire-meshes (b).

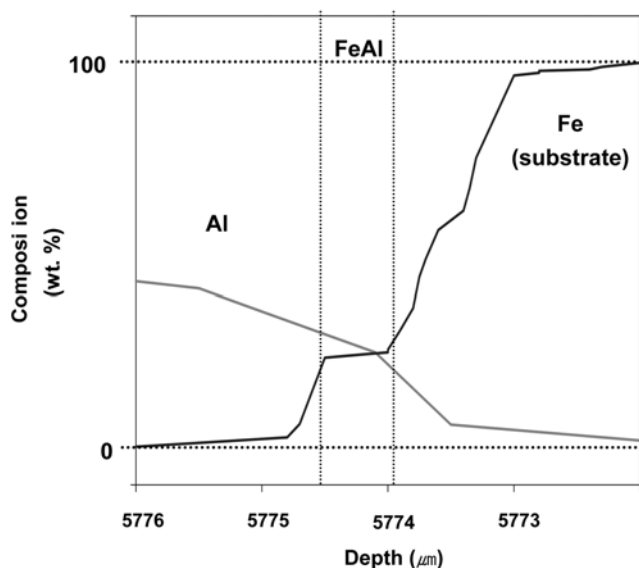


Fig. 4. Atomic depth profile at the interphase of Al coatings and wire-mesh.

detached from the metallic substrate even after ultrasound vibration. However, the Al coating layer might be highly sintered at such high temperature, so it had non-porous surface structure that was inadequate for catalyst coating. A suitable temperature range, therefore, was chosen around 800 °C for the thermal treatment. At this temperature, the coated layer had high stabilities against thermal and mechanical shock. In order to know the chemical composition at interphase for Al-coated layer and wire surface, atomic depth profiles of Al and Fe were performed by electron probe micro-analysis (EPMA) and the results are shown in Fig. 4. There exists an overlap zone of aluminum and iron species within a thickness around 0.5 μm. The atomic ratio of the aluminum and iron was 1 : 1 in the overlap zone, indicating the existence of an alloy structure of Al and Fe. Therefore, we have concluded that the formation of the Al-Fe alloy phase at the interface makes the Al coatings stick strongly to the metallic substrate of wire.

Above Al-coated wire-meshes were then calcined at 500 °C for a certain period of time in order to form a thin oxide layer of Al₂O₃ at the outer surface of each Al particle. EDS result confirms the existence of oxygen-containing layers as shown in the oxygen profile of Fig. 5. Detailed analysis of the oxidized Al₂O₃ layer by using XPS

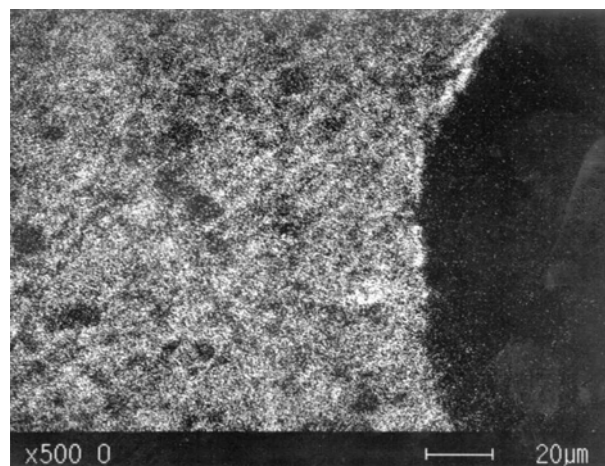


Fig. 5. Oxygen profile of the cross-section area of deposited aluminum layer.

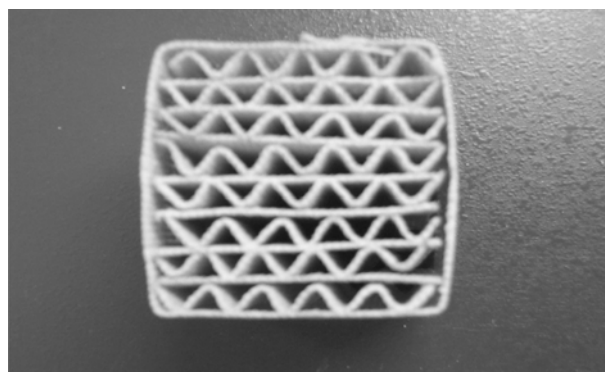


Fig. 6. Picture of catalyst-coated wire-mesh honeycomb.

depth profiling revealed that the thickness can be controlled at 10–100 nm, completely encapsulating each Al particles. V₂O₅-WO₃/TiO₂ catalyst powder was then coated on the above Al₂O₃/Al-coated wire-meshes via wash coating method. Fig. 6 shows the final wire mesh honeycomb with catalyst layer coated on wire meshes. Wire meshes have shown more or less uniform coating of catalyst since catalyst slurry was coated onto the outer surface of the wire rod. More coating was found at the cross section of wires, but non-uniformity was not so severe when compared to square shaped channels in a ceramic honeycomb where catalyst powder has more dense coating at the

Table 1. Physical Properties of Al/Al₂O₃-composite particles that were coated on wire-mesh and V₂O₅-WO₃/TiO₂ catalyst that was deposited on Al/Al₂O₃-coated wire-mesh

Item		Al/Al ₂ O ₃ -coated wire-mesh	V ₂ O ₅ -WO ₃ /TiO ₂ catalyst deposited on Al/Al ₂ O ₃ -coated WMH
Al/Al ₂ O ₃ layer	Thickness of Al ₂ O ₃ phase on individual Al particle	45 nm	-
	Thickness of coated layer	80 μm	-
	BET surface area	12.23 m ² /g	-
	Average pore diameter	110 Å	-
V ₂ O ₅ -WO ₃ /TiO ₂ layer	Thickness of coated layer	-	40 μm
	BET surface area	-	55 m ² /g
	Average pore diameter	-	100 Å

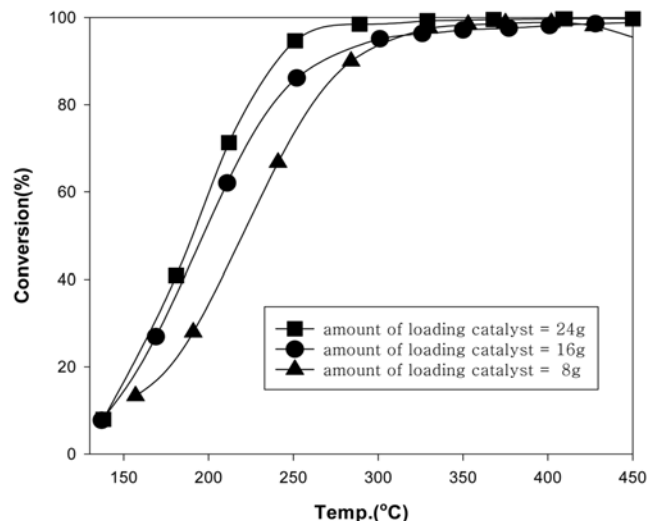


Fig. 7. Effect of loading amount of catalyst on the NO conversion. GHSV: 5,000/hr; initial concentration of NO: 500 ppm; Concentration of O₂: 21%; NH₃/NO ratio: 1.

corner of the channel. The physical properties of deposited layer of Al₂O₃/Al composite particles and that of V₂O₅-WO₃/TiO₂ catalyst powder are summarized in Table 1.

2. Influence of Catalyst Loading on NO Conversion

To find a proper loading amount of the catalyst on the WMH module, three different loadings from 8 to 24 wt% were tested for selective catalytic reduction of NO with NH₃ as shown in Fig. 7. With the increase of the catalyst loading, the SCR conversion of NO was also increased. However, the NO conversion is almost unaffected by changing the loading when temperature is above 300 °C. When catalyst loading is kept lower than 8 wt%, the surface of WMH cannot be fully covered with catalyst and uncovered surface will lead to the appearance of FeO_x which results in the ammonia oxidation. In this case the SCR activity is quite low while the ammonia oxidation becomes appreciable at higher temperatures.

It is revealed that, by visual inspection, the loading amount of approximately 16 wt% is enough for covering all the surface of wire mesh, as evidenced by a uniform yellow color of the module after calcination. As the loading increased above 16 wt%, the catalyst accumulated at the cross-points in the wire mesh and openings in the wire mesh prone to loss due to attrition or shock. With an optimized catalyst loading of 16 wt%, the WMH catalyst module shows more than 90% conversion at 240 °C and almost complete NO conversion at temperatures higher than 300 °C at GHSV 5,000 h⁻¹.

3. Effect of GHSV on NO Conversion

The effects of GHSV on the SCR conversion of NO tested at various reaction temperatures are shown in Fig. 8. Increased GHSV decreases the SCR conversion of NO. At a high space velocity, the decrease in NO conversion may result from insufficient contact time between the reactants and the catalyst or from insufficient amounts of active sites to accommodate the high flux of the reactants. Fig. 9 shows a comparison of the NO conversion between WMH catalyst and CH catalyst. When temperature is kept extremely low at 150 °C, there is no appreciable difference in the NO conversion between the two catalysts. However, WMH catalyst becomes much more effective than CH catalyst when temperature exceeds 150 °C.

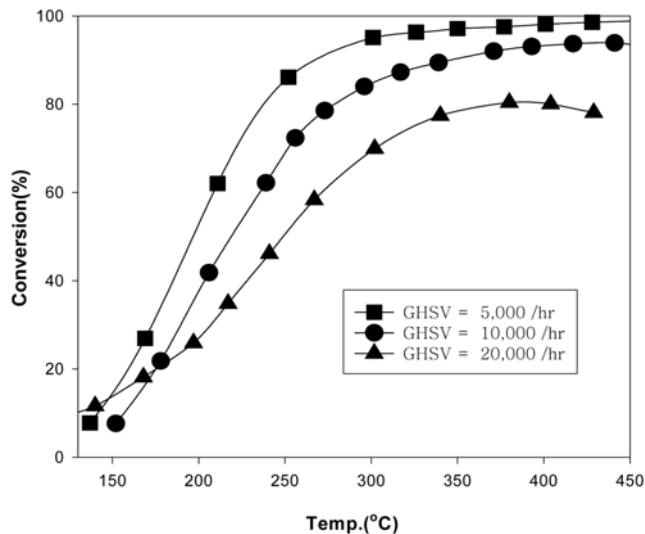


Fig. 8. Effect of GHSV on the NO conversion. Catalyst loading: 16 g; initial concentration of NO: 500 ppm; Concentration of O₂: 21%; NH₃/NO ratio: 1.

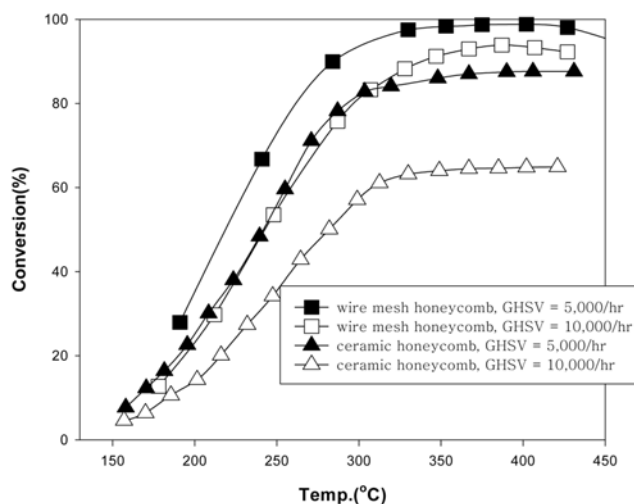


Fig. 9. Comparison of NO conversion between WMH catalyst and CH catalyst. Catalyst loading: 16 g; initial concentration of NO: 500 ppm; Concentration of O₂: 21%; NH₃/NO ratio: 1.

Activity difference becomes more appreciable as temperature increases especially at higher GHSV, indicating that the activity increase results from beneficial increase of radial mixing in the WMH due to the existence of additional openings in the wire mesh, which must result in three-dimensional flow to induce turbulence inside channels. Detailed analysis of flow patterns of WMH has shown that its mixing characteristics of gas flow inside channels of WMH are quite different from that of CH, and rather similar to the gas flow through a packed-bed [Jiang et al., 2003].

4. Influence of Water on SCR Conversion

To understand the effects of H₂O on the catalytic performance for SCR conversion of NO over WMH catalyst and CH catalyst, the amount of water in the feed was changed from 0-4 vol%. WMH catalyst was not affected by the water addition for the SCR conversion of NO, whereas CH catalyst showed a continuous decrease of

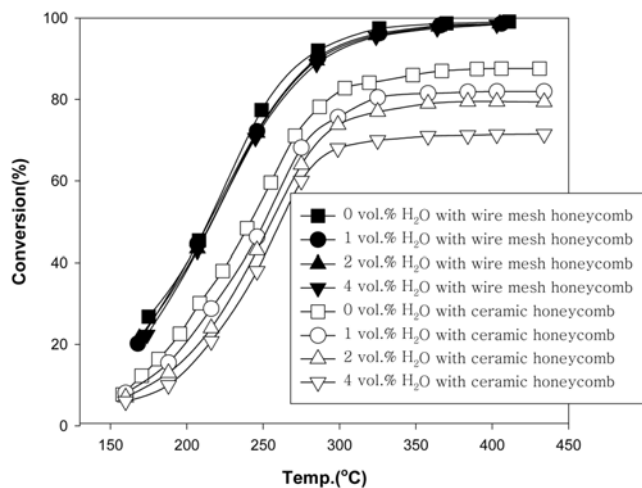


Fig. 10. Effect of water content in the feed on the NO conversion. Catalyst loading: 16 g; initial concentration of NO: 500 ppm; Concentration of O₂: 21%; NH₃/NO ratio: 1.

NO conversion upon increased addition of water in the feed. It is known that H₂O inhibits catalytic activity at low temperatures caused by water being adsorbed completely on the active site with ammonia [Topsoe et al., 1995]. We have coated the same kind of catalyst with the same amount for both WMH and CH modules. The only difference between the two is coating uniformity: WMH has more uniform coating profile of catalyst layer outside surface of wire meshes, whereas CH has a thicker layer on the corners inside rectangular-shaped channels. The water effect must be a catalytic factor; therefore it cannot be affected by any difference in the physical factor. At present, we do not know why there exists so much difference in the performance between WMH and CH, and must be studied further.

5. Transient Response

To get insight on the dynamics of the SCR process under both steady and unsteady state, the SCR reaction under unsteady conditions was investigated with the application of the transient response method. Perturbations are imposed on the reacting system by step changes in the inlet reactant concentration while being kept constant for the concentrations of the other reactants. Transient behavior is observed upon shutting off the feed of NH₃. When the NH₃ concentration was switched off to zero value, the outlet NO concentration was not immediately affected and several minutes of time period was required to reach the steady state value of inlet NO concentration. Upon turning the NH₃ feed back on, however, a relatively faster response in the NO concentration was observed. The results indicate the existence of a 'reservoir' of adsorbed ammonia species available for a reaction on the catalyst surface. The results are also in agreement with a general observation in adsorption theory: the desorption rate of adsorbed ammonia is much slower than the adsorption rate of gas phase ammonia to the surface. It may be speculated that NH₃ is adsorbed on V, W or Ti sites, since these sites are able to strongly coordinate ammonia. The observed transient responses are typical of a reaction involving a strongly adsorbed species (NH₃) and a gas-phase or weakly adsorbed species (NO) in line with the hypothesis of an Eley-Rideal mechanism for the SCR reaction. Fig. 11 shows that the gradient over WMH is steeper than that over CH. The results indicate that WMH has a more uniform

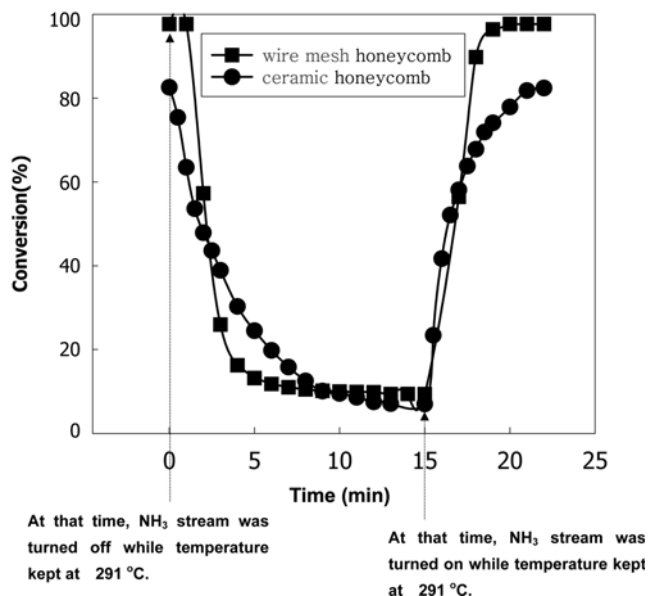


Fig. 11. Transient responses over WMH and CH catalyst. Reaction temperature: 291 °C; GHSV: 5,000/hr; initial concentration of NO: 500 ppm; Concentration of O₂: 21%; Catalyst loading: 16 g. NH₃ stream was turned off at time=0 min and later turned on at time=15 min while temperature kept on 291 °C.

concentration profile along axial directions of different channels and/or faster interphase mass transfer rate toward channel walls. This proves that WMH can reach higher activity in shorter time than CH at the initial starting stage of a reaction. WMH could be a promising catalyst for practical applications such as in controlling vehicle emission which requires achieving a higher activity to avoid the emission of exhaust gas at early start-up stage.

6. Long-Term Tests

The activity comparisons between WMH and CH catalyst for a long period of reaction are shown in Fig. 12. During the longevity test, we often shut off the feed and kept the reactor at room tem-

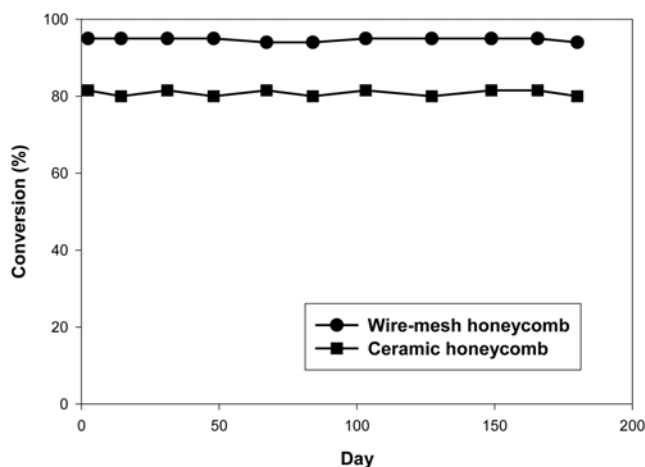


Fig. 12. Long-term tests over WMH and CH catalyst. Reaction temperature: 300 °C; GHSV: 5,000/hr; initial concentration of NO: 500 ppm; Concentration of O₂: 21%; Catalyst loading: 16 g.

perature overnight, at least 1-2 times every week. The SCR activities over WMH catalyst were always kept higher than those over CH catalyst. Six months of operational data with WMH shows that there is no appreciable decrease in the NO conversion with time on stream when compared to CH. The results confirm that the adherence of catalyst coatings on the Al/Al₂O₃-coated metal wire meshes is as good as that coated on the ceramic surface.

CONCLUSIONS

Electrophoretic deposition (EPD) method was used to coat a porous aluminum layer on wire meshes. After thermal sintering and calcining process, each deposited Al particles form a thin Al₂O₃ layer at the outer surface, resulting in double layered composite of Al/Al₂O₃ particles. The composite particles were firmly attached on the wire mesh surface as uniform and porous layers. WMH module with triangular channels was manufactured by packing alternately a flat sheet and corrugated sheet of the Al/Al₂O₃-coated wire mesh into a rectangular-shape. WMH and CH that were coated with V₂O₅-MoO₃-WO₃ catalyst powder were applied for the selective catalytic reduction of NO with NH₃. The results show that WMH catalyst module has higher SCR reactivity for NO conversion with NH₃, good endurance against mechanical stresses and vibrations. WMH catalyst shows many advantages over other SCR catalyst, such as excellent thermal response, excellent thermal shock resistance, geometric flexibility, axial dispersion and simplicity of recovering catalyst. WMH catalyst shows broader temperature window and higher NO conversion than conventional CH catalyst due to reduced mass transfer resistance by the existence of three dimensional holes in the WMH. Faster transient response makes it ideal to be used for treating vehicle emissions.

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REFERENCES

- Ahlstrom-Silversand, A. F. and Odenbrand, C. U. I., "Thermally sprayed wire-mesh catalysts for the purification of flue gases from small-scale combustion of bio-fuel Catalyst preparation and activity studies," *Applied Catalysis, A*, **153**(1-2), 177 (1997).
- Amiridis, M. D., Duevel, R. V. and Wachs, I. E., "The effect of metal oxide additives on the activity of V₂O₅/TiO₂ catalysts for the selective catalytic reduction of nitric oxide by ammonia," *Applied Catalysis B: Environmental*, **20**(2), 111 (1999).
- de Boer, M., Huisman, H. M., Mos, R. J. M., Leliveld, R. G., van Dillen, A. J. and Geus, J. W., "Selective oxidation of ammonia to nitrogen over SiO₂-supported MoO₃ catalysts," *Catalysis Today*, **17**(1-2), 189 (1993).
- Bosch, H., Janssen, F. J. J. G., van den Kerkhof, F. M. G., Oldenziel, J., van Ommen, J. G and Ross, J. R. H., "The activity of supported vanadium oxide catalysts for the selective reduction of NO with ammonia," *Applied Catalysis*, **25**, 239 (1986).
- Chae, H. J., Nam, I.-S., Ham, S.-W. and Hong, S. B., "Characteristics of vanadia on the surface of V₂O₅/Ti-PILC catalyst for the reduction of NO_x by NH₃," *Applied Catalysis B: Environmental*, **53**(2), 117 (2004).
- Chen, J. P. and Yang, R. T., "Mechanism of poisoning of the V₂O₅/TiO₂ catalyst for the reduction of NO by NH₃," *Journal of Catalysis*, **125**(2), 411 (1990).
- Choi, H., Ham, S. W., Nam, I. S., Kim, Y. G., Shim, J. H. and Ha, B. H., "NO reduction with ammonia using parallel passage reactor," *HWAHAK KONGHAK*, **34**, 91 (1996).
- Chung, J. H., Shon, B. H., Yoo, K. S., Kim, H. K. and Lee, H. K., "Simultaneous removal of SO₂ and NO_x by the absorbent from coal fly ash," *HWAHAK KONGHAK*, **41**, 403 (2003).
- Chung, K.-S., Jiang, Z., Gill, B.-S. and Chung, J. S., "Oxidative decomposition of o-dichlorobenzene over V₂O₅/TiO₂ catalyst washcoated onto wire-mesh honeycombs," *Applied Catalysis A: General*, **237**(1-2), 81 (2002).
- Cybulski, A. and Moulijn, J. A. (ed.), *Structured catalysts and reactors*, Marcel Dekker, Inc. (1998).
- Dutoit, D. C. M., Reiche, M. A. and Baiker, A., "Vanadia-silica aerogels. Structure and catalytic properties in selective reduction of NO by NH₃," *Applied Catalysis B: Environmental*, **13**(3-4), 275 (1997).
- Handy, B. E., Baiker, A., Schraml-Marth, M. and Wokaun, A., "Vanadia supported on TiO₂-SiO₂ mixed oxide gels: Structure of the dispersed phase and activity for the selective catalytic reduction of NO with NH₃," *Journal of Catalysis*, **133**(1), 1 (1992).
- Handy, B. E., Maciejewski, M. and Baiker, A., "Vanadia, vanadia-titania, and vanadia-titania-silica gels: Structural genesis and catalytic behavior in the reduction of nitric oxide with ammonia," *Journal of Catalysis*, **134**(1), 75 (1992).
- Jiang, Z., Chung, K.-S., Kim, G.-R. and Chung, J. S., "Mass transfer characteristics of wire-mesh honeycomb reactors," *Chemical Engineering Science*, **58**(7), 1103 (2003).
- Kim, B. S., Lee, S. H., Park, Y. T., Ham, S. W., Chae, H. J. and Nam, I. S., "Selective catalytic reduction of NO_x by propene over copper-exchanged pillared clays," *Korean J. Chem. Eng.*, **18**, 704 (2001).
- Kim, B. T., Lee, H. G., Chun, G. S., Lee, G. J. and Park, H. S., "NO_x absorption into aqueous solutions: II. HNO₃ and HNO₂ concentration produced by NO_x absorption for a sieve tray column," *HWAHAK KONGHAK*, **25**(2), 169 (1987).
- Kim, G. R., Jiang, Z. and Chung, J. S., Korean Patent 0336821 (2002).
- Lee, I.-Y., Kim, D.-W., Lee, J.-B. and Yoo, K.-O., "A practical scale evaluation of sulfated V₂O₅/TiO₂ catalyst from metatitanic acid for selective catalytic reduction of NO by NH₃," *Chemical Engineering Journal*, **90**(3), 267 (2002).
- Lee, H. T. and Rhee, H. K., "Steam tolerance of Fe/ZSM-5 catalyst for the selective catalytic reduction of NO_x," *Korean J. Chem. Eng.*, **19**, 574 (2002).
- Lee, S. H., Ahn, J. S. and Kim, J. H., "Selective catalytic reduction of NO_x catalyst," *News & Information for Chem. Eng.*, **19**(4), 468 (2001).
- Lintz, H.-G. and Turek, T., "Intrinsic kinetics of nitric oxide reduction by ammonia on a vanadia-titania catalyst," *Applied Catalysis A: General*, **85**(1), 13 (1992).
- Liuqing, T., Daiqi, Ye. and Hong, L., "Catalytic performance of a novel ceramic-supported vanadium oxide catalyst for NO reduction with NH₃," *Catalysis Today*, **78**(1-4), 159 (2003).
- Long, R. Q. and Yang, R. T., "Catalytic performance and characteriza-

- tion of VO²⁺-exchanged titania-pillared clays for selective catalytic reduction of nitric oxide with ammonia," *Journal of Catalysis*, **196**(1), 73 (2000).
- Lucas, D. and Brown, N. J., "Characterization of the selective reduction of NO by NH₃," *Combustion and Flame*, **47**, 219 (1982).
- Park, J. H., Kim, D. J. and Kim, G. S., "Analysis on NO_x conversion and particle characteristics in NO_x removal by corona discharge," *HWAHAK KONGHAK*, **40**, 351 (2002).
- Piehl, G., Liese, T. and Grünert, W., "Activity, selectivity and durability of VO-ZSM-5 catalysts for the selective catalytic reduction of NO by ammonia," *Catalysis Today*, **54**(4), 401 (1999).
- Rajadhyaksha, R. A., Hausinger, G., Ramstetter, H. Z. and Knözinger, H. S., "Vanadia supported on titania-silica: Physical characterization and activity for the selective reduction on nitric oxide," *Applied Catalysis*, **51**(1), 67 (1989).
- Topsoe, N. Y., Topsoe, H. and Dumesic, J. A., "Vanadia/titania catalysts for selective catalytic reduction (SCR) of nitric-oxide by ammonia: I. Combined temperature-programmed *in-situ* FTIR and on-line mass-spectroscopy studies," *Journal of Catalysis*, **151**(1), 226 (1995).
- Weng, R.-Y. and Lee, J.-F., "Catalytic performance and active sites determination of niobium oxide promoted vanadia/titania catalysts for selective catalytic reduction of nitric oxide," *Applied Catalysis A: General*, **105**(1), 41 (1993).
- Went, G. T., Leu, L.-J., Rosin, R. R. and Bell, A. T., "The effects of structure on the catalytic activity and selectivity of V₂O₅/TiO₂ for the reduction of NO by NH₃," *Journal of Catalysis*, **134**(2), 492 (1992).
- Yang, K. S., Choi, J. and Chung, J. S., "Evaluation of wire-mesh honeycomb containing porous Al/Al₂O₃ layer for catalytic combustion of ethyl acetate in air," *Catalysis Today*, **97**(2-3), 27-159 (2004).
- Yang, K. S., Jiang, Z. and Chung, J. S., "Electrophoretically Al-coated wire mesh and its application for catalytic oxidation of 1,2-dichlorobenzene," *Surface and Coatings Technology*, **168**(2-3), 103 (2003).
- Yoo, K. S., Lee, J. G., Park, D. G., Chung, M. J., Lee, C. and Shin, J. W., "The selective non-catalytic reduction reaction of NO_x using urea solution in a flow reactor," *HWAHAK KONGHAK*, **41**, 219 (2003).