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AN EXPERIMENTAL INVESTIGATION ON THE DESIGN AND IMPLEMENTATION OF THE MIXING CHAMBERS FOR IMPROVING THE THERMAL-DECOMPOSITION OF THE UREA

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ABSTRACT

Urea-SCR has been reported to be the most promising method for meeting NO_X emissions regulation. In the urea-SCR process, NH_3 is generated by urea thermolysis and hydrolysis and used as a reductant of NO_X. Consequently, to improve NO_X conversion efficiency of urea-SCR, it is required to enhance the thermal-decomposition and hydrolysis rate upstream of SCR catalyst.

In the present work, experiments are carried out by varying inlet velocities (4~12 m/s) and temperatures (350~500℃) to analyze the characteristics of urea thermal-decomposition. Two types of the mixing chambers are designed and fabricated to maximize the thermal-decomposition of urea. The thermal-decomposition characteristics of urea to isocyanic acid (HNCO) and ammonia (NH_3) are experimentally examined without and with mixing chamber.

The thermal-decomposition of the urea is greatly enhanced at low velocities and high gas temperatures. At inlet velocity of 6 m/s and inlet temperature of 400℃, NH_3 concentration with A-type mixing chamber is maximally about 200% greater than that without mixing chamber. When the inlet velocity and temperature with B-type mixing chamber are 4 m/s and 400℃, respectively, NH_3 concentration is maximally about 187% greater compared to that without mixing chamber. It is noted that as the mixing chamber is implemented the conversion of urea to NH_3 is significantly enhanced because the residence time of urea in the exhaust pipe increases and the mixing between urea and exhaust gas is improved.

INTRODUCTION

The deNO_X technologies, such as SCR (selective catalytic reduction) using urea or HC (hydrocarbon), LNC (lean NO_X catalyst) and LNT (lean NO_X trap), are being presented for reduction of NO_X in diesel engines. Selective catalytic reduction of NO_X by NH_3 is a well-developed technique and the most common technology for the control of NO_X emitted from stationary sources. SCR systems can be classified into two categories: NH_3-SCR and urea-SCR. NH_3-SCR using NH_3 as reductant gas is very effective to control NH_3 injection for reduction of NO_X in exhaust from diesel and gasoline lean-burn engines, however, this technology is not suitable for automotive use because of difficulties in the storing and handling of NH_3. Urea is the best choice because it is not toxic and can be easily transported, and hence urea-SCR for the NO_X removal from diesel engine exhausts has been studied by many previous researches [1, 2].

When the urea-water solution (UWS) is atomized into an exhaust gas stream, the first step for the decomposition is the evaporation of water from the droplets of UWS, as shown in reaction (1). Urea is then thermally decomposed into NH_3 and HNCO. HNCO formed by reaction (2) is quite stable in the gas phase, but it easily hydrolyzes on the surface of metal oxides, producing NH_3 and CO_2 shown in reaction (3) [3].

$$H_2N\text{-}CO\text{-}NH_2 \rightarrow H_2N\text{-}CO\text{-}NH_2\,(s) + xH_2O\,(g) \quad (1)$$

$$H_2N\text{-}CO\text{-}NH_2 \rightarrow NH_3\,(g) + HNCO\,(g) \quad (2)$$

$$HNCO\,(g) + H_2O\,(g) \rightarrow NH_3\,(g) + CO_2\,(g) \quad (3)$$

In the urea-SCR process, NH_3 generated by urea thermolysis and hydrolysis is used as a reductant of NO_X. Consequently, in order to improve NO_X conversion efficiency of urea-SCR, it is required to enhance the thermal-decomposition and hydrolysis rate upstream of SCR catalyst. Nishioka et al. [4] reported NH_3-SCR showing the ideal performance in NO_X conversion efficiency, compared with urea-SCR. Shimizu and Satsuma [1] presented that the NO conversions over Ag/Al_2O_3 SCR catalyst in H_2 assisted urea-SCR (H_2-urea-SCR) condition are lower than those in the H_2-NH_3-SCR condition and are higher than those in the H_2-C_3H_8-SCR condition below 450℃. Sullivan and Doherty [5] showed that the NO_X conversions over Cu/AlO and Cu/TiO SCR catalysts in H_2O-urea-SCR condition are lower than those in the H_2O-NH_3-SCR condition. Koebel and Strutz [6] presented that urea-SCR technique suffers from short residence times, leading

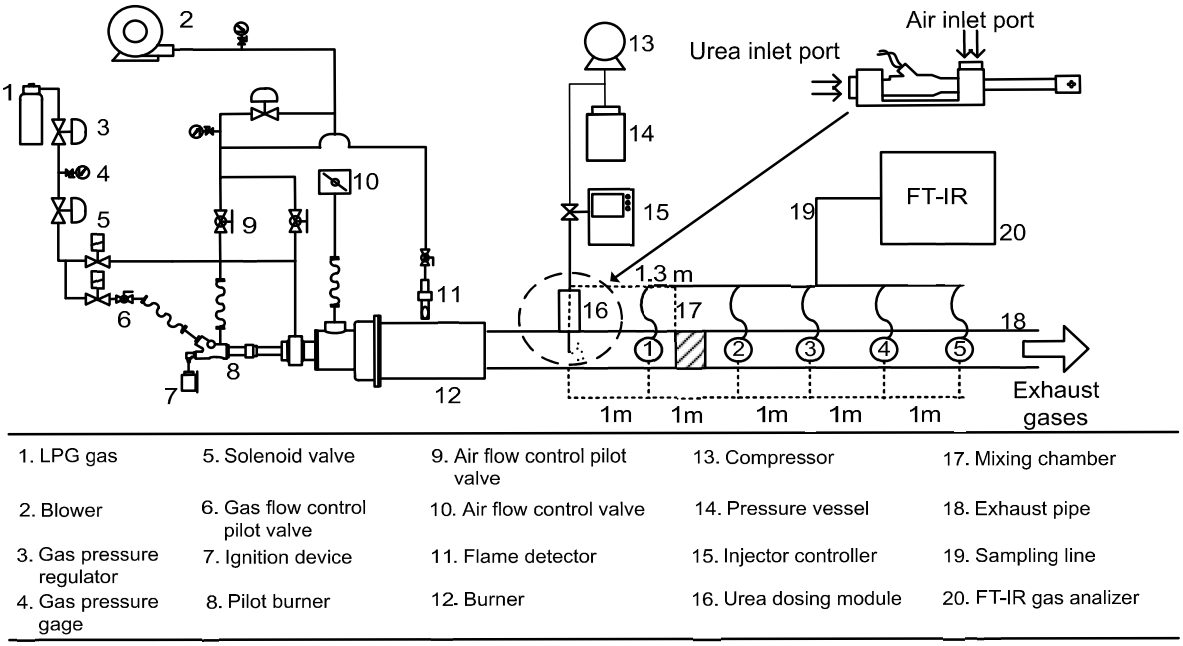

Figure 1 Schematic of the experimental setup

to incomplete decomposition into NH_3 and HNCO and causing a significant performance loss of the SCR catalyst. Accordingly, an increase in the residence time of urea in the exhaust pipe is essentially required to enhance the conversion of urea to NH_3.

In the present study, experiments are carried out by varying inlet velocities (4~12 m/s) and temperatures (350~500℃) to analyze the characteristics of urea thermal-decomposition in the exhaust pipe of 60 mm in diameter. Also, two types of the mixing chambers are designed and fabricated to maximize the thermolysis of urea. The thermal-decomposition characteristics of urea to HNCO and NH_3 are experimentally examined without and with mixing chambers.

EXPERIMENTAL SETUP

Experimental apparatus depicted in Figure 1 consists of three main parts: gas burner and blower, urea injection module, FT-IR (Fourier transform spectroscopy) gas analyzer. A desired amount of 1.56 g/min UWS (32.5%) is continuously fed into the exhaust pipe using an air-assist type urea injection module. The exhaust pipe is well insulated and maintained.

The gas burner and blower, which consist of gas pressure regulator, solenoid valve, gas and air flow control pilot valve, ignition device, pilot burner, flame detector, and burner, are used to generate inlet velocity and temperature conditions that range from 4 to 12 m/s and from 350 to 500℃, similar to typical operating condition in diesel engine. The various sets of inlet velocity and temperature conditions are kept constant by feedback control system.

The urea injection module consists of compressor, pressure vessel, controller, and injector. Pressure vessel filled with UWS is kept under constant pressure of 2 bar by using compressor, and its maximum allowable working pressure is 10 bar. The compressed UWS in the pressure vessel is supplied into the urea inlet port and the UWS through gasoline injector is mixed with compressed air of 1.8 bar in the mixing chamber of the urea injection module. Then, well-mixed urea and air through the four-hole nozzle are injected into the exhaust stream. The urea injection rate is controlled by adjusting the applied frequency and duty ratio in the controller. Urea injector and controller operate on 12V DC.

Gas sampling is conducted from the five sampling ports with the increments of 1 m from the urea injection. Mixing chamber is mounted in the exhaust stream 1.3 m from the injection nozzle. The NH_3 and HNCO concentrations are measured using a FT-IR (Fourier transform spectroscopy) gas analyzer.

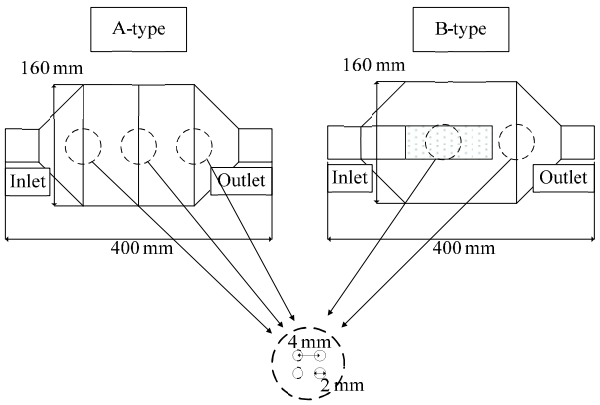

Figure 2 Schematic of the mixing chambers used in the experiment.

MIXING CHAMBER

The two types of mixing chambers are presented in Figure 2. A- and B-type mixing chambers are used to enhance the conversion efficiency of urea to NH_3 by increasing the residence time and the mixing of urea with the exhaust gas. The A-type mixing chamber consists of three plates having punching holes on the cross-section of plate and these plates are set up with an equal interval. A partial amount of exhaust gas passes through the punching holes, while the rest forms the vortex flow by the plate. The B-type mixing chamber consists of a cylindrical pipe with punching holes on periphery and one end blocked and a plate. The diameter of the cylindrical pipe is equal to the diameter of the main exhaust pipe and is smaller than that(160 mm) of mixing chamber. The exhaust gas flows into the cylindrical pipe and flows radially out through the peripheral holes of the cylindrical pipe. The diameter of the punching hole and space between holes are 2 mm and 4 mm, respectively.

EXPERIMENTAL RESULTS AND DISCUSSIONS.

EFFECT OF INLET VELOCITY AND TEMPERATURE ON THE UREA THERMOLYSIS WITHOUT MIXING CHAMBER.

In order to analyze the characteristics of urea thermolysis without mixing chamber, NH_3 concentration produced from urea thermal-decomposition is measured along the exhaust pipe at the inlet velocities in the range of 4~12 m/s and inlet gas temperatures in the range of 350~500℃.

Figure 3 shows the exhaust gas temperature and NH_3 concentration distributions along the pipe for various inlet gas temperatures at the inlet velocity of 6 m/s. With increasing inlet gas temperature and axial distance, the NH_3 concentration increases and the gas temperature decreases gradually. The maximum NH_3 concentration is about 145 ppm at the fifth measuring point with the inlet gas temperature of 500℃. The highest increasing rate of the NH_3 concentration, defined as the ratio of the increment of NH_3 concentration to the increment of length, is about 75 ppm/m between the first and the second measuring points. Upon increasing distance at the inlet temperature of 500℃, the increasing rate decreases due to a monotonic decrease in the exhaust gas temperature by the vaporization of UWS and a convection heat loss to atmosphere, and this tendency occurs similarly at the inlet temperatures lower than 500℃. Regardless of the measuring points, the NH_3 concentration increases with an increasing inlet temperature. When the inlet temperature increases from 350℃ to 400℃, the highest increasing rate of NH_3 concentration, the ratio of the increment of NH_3 concentration to the increment of inlet gas temperature, is about 1.12 ppm/℃ at the fifth measuring point,

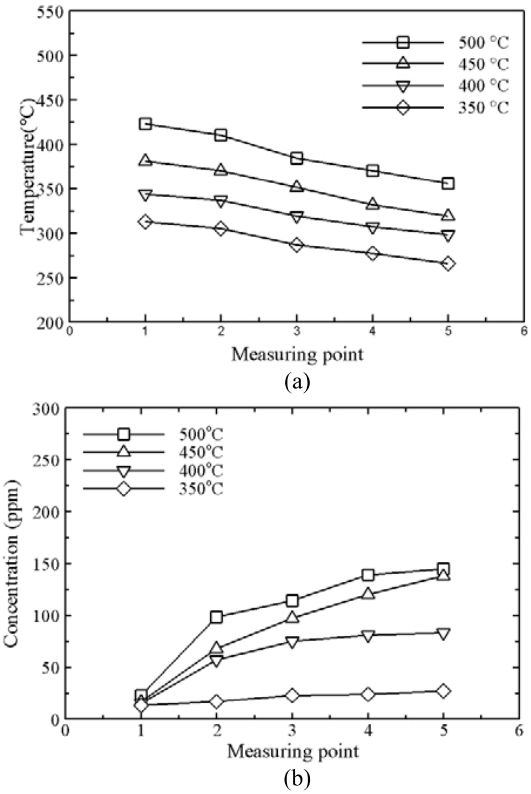

Figure 3 Gas temperature (a) and NH_3 concentration (b) distributions along the pipe with respect to inlet gas temperature at a fixed inlet velocity of 6 m/s without mixing chamber

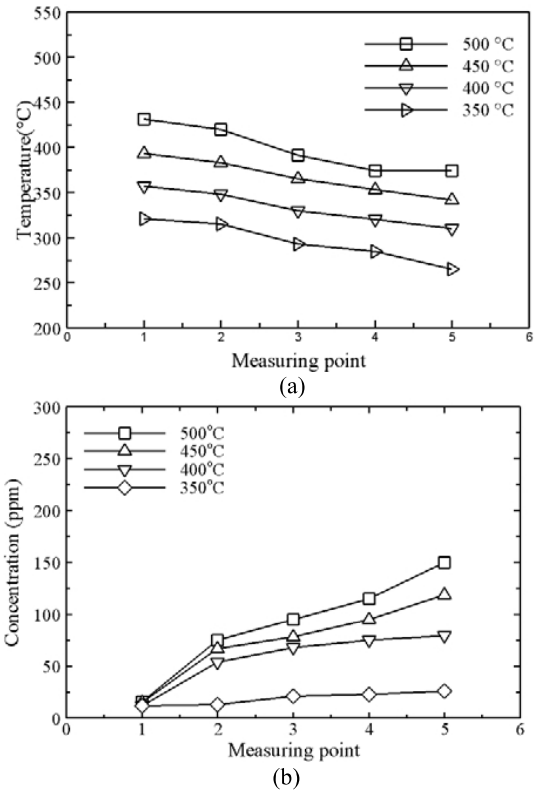

Figure 4 Gas temperature (a) and NH_3 concentration (b) distributions along the pipe with respect to inlet gas temperature at a fixed inlet velocity of 8 m/s without mixing chamber

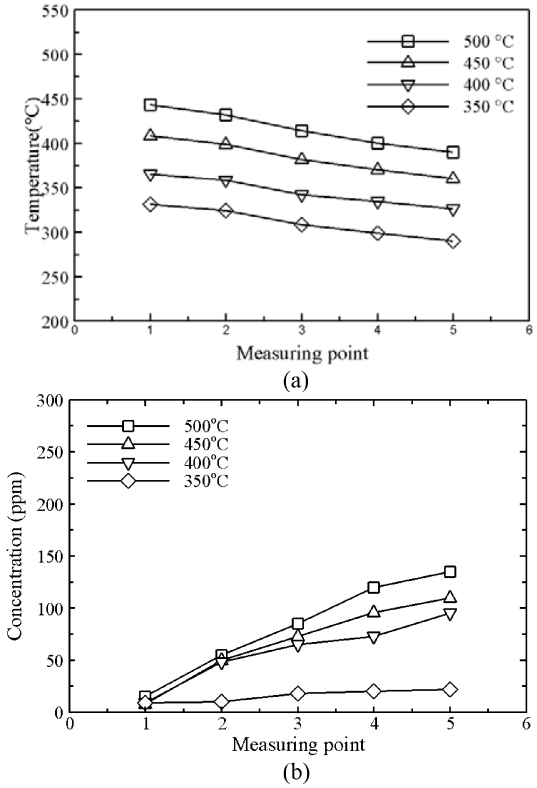

Figure 5 Gas temperature (a) and NH_3 concentration (b) distributions along the pipe with respect to inlet gas temperature at a fixed inlet velocity of 10 m/s without mixing chamber

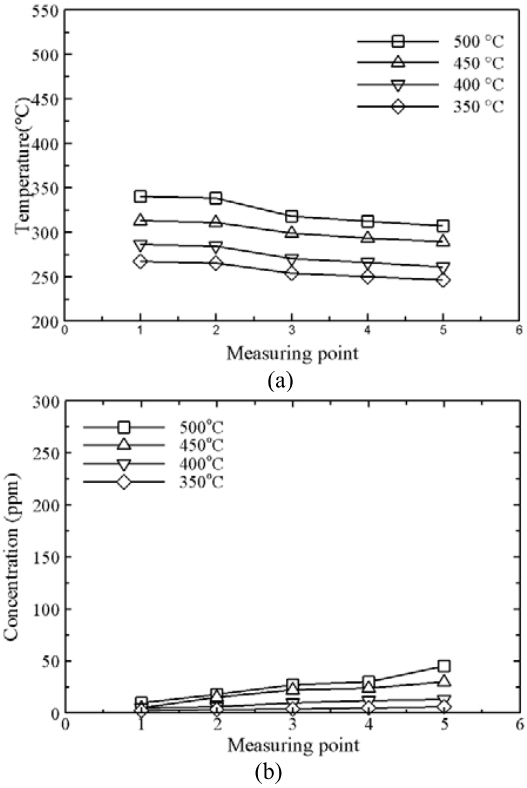

Figure 6 Gas temperature (a) and NH_3 concentration (b) distributions along the pipe with respect to inlet gas temperature at a fixed inlet velocity of 12 m/s without mixing chamber

and it decreases with increasing inlet temperature. However, at the inlet temperatures below 350℃ the NH_3 concentration produced through urea thermolysis is very small due to the deactivated thermal-decomposition of urea. Because the characteristics of gas temperature and NH_3 concentration with 4 m/s are similar to the case with 6 m/s, the plots are omitted here.

As shown in Figure 4 for the inlet velocity of 8 m/s, the maximum NH_3 concentration is about 150 ppm at the fifth measuring point with the inlet temperature of 500℃. The maximum increasing rate of the NH_3 concentration along the distance is about 59 ppm/m between the first and the second measuring points at the inlet temperature of 500℃, and the increasing rate decreases upon increasing length. As inlet temperature increases from 350℃ to 400℃, the increasing rate of the NH_3 concentration is the highest as about 1.06 ppm/℃ at the fifth measuring point, and it decreases with an increase in the inlet temperature.

Figure 5 denotes the characteristics of urea thermal-decomposition along increasing inlet temperature and distance at the inlet velocity of 10 m/s. The maximum NH_3 concentration is about 135 ppm at the fifth measuring point with the inlet temperature of 500℃. The maximum increasing rate of the NH_3 concentration along the distance is the highest as about 40 ppm/m between the first and the second measuring points at the inlet temperature of 500℃. With increasing the inlet temperature from 350℃ to 400℃, the highest increasing rate of the NH_3 concentration is about 1.46 ppm/℃ at the fifth measuring point.

As depicted in Figure 6 with the inlet velocity of 12 m/s, the maximum NH_3 concentration is about 45 ppm at the fifth measuring point for the inlet temperature of 500℃, and it is significantly lower, about 90 ppm, than that of the case with 10 m/s. At between the first and the second measuring points with the inlet temperature of 500℃, the increasing rate of the NH_3 concentration along the distance is the highest, and its value is about 15 ppm/m. And the maximum increasing rate of the NH_3 concentration along increasing inlet temperature is about 0.34 ppm/℃. When the inlet velocity is 12 m/s, regardless of the inlet temperature and measuring points, the NH_3 concentration is very small because of shortened residence time of UWS in exhaust pipe.

As can be seen from experimental results shown above, the NH_3 concentration at the first measuring point is very low, regardless of inlet temperature and inlet velocity. This is due to an inactivated atomization of urea-water solution, which is caused by the very short residence time of UWS in the exhaust gas. Also, as inlet velocity increases from 4 m/s to 12 m/s, the average NH_3 concentration over all measuring points decreases

because the penetration length of urea spray increases and thus it needs more times to achieve fine atomization of UWS. [7]

EFFECT OF INLET VELOCITY AND TEMPERATURE ON THE UREA THERMOLYSIS WITH A-TYPE MIXING CHAMBER.

In order to increase the residence time of UWS in exhaust pipe and the mixing of urea with the exhaust gas, the A-type mixing chamber shown in Figure 2 is implemented. Under the operating conditions of inlet velocity (6, 8 m/s) and temperature (400, 500℃), the NH_3 concentration produced from urea thermal-decomposition is measured along the axial distance, except for the first measuring point because of the set-up of mixing chamber at 1.3 m away from the injection nozzle.

Figure 7 presents the NH_3 concentrations along the axial distance as a function of inlet gas temperature with and without an implementation of the A-type mixing chamber at the inlet velocity of 6 m/s. When the inlet temperatures are 400℃ and 500℃, the average NH_3 concentrations over all measuring points with the mixing chamber increase by about 170% and about 157%, respectively, compared to those without the mixing chamber. With the inlet temperatures of 400℃ and 500℃, at the fifth measuring point the NH_3 concentrations are 160 ppm and 228 ppm, respectively, and are about 193% and about 157% higher than those without the mixing chamber.

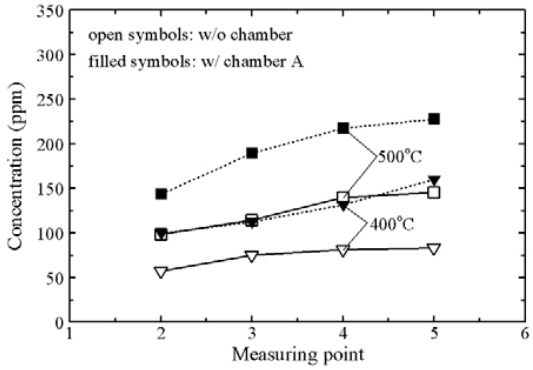

Figure 7 NH_3 concentration along the pipe with respect to inlet gas temperature at a fixed inlet velocity of 6 m/s with A-type mixing chamber: open and filled symbols denote NH_3 concentration with and without A-type mixing chamber, respectively

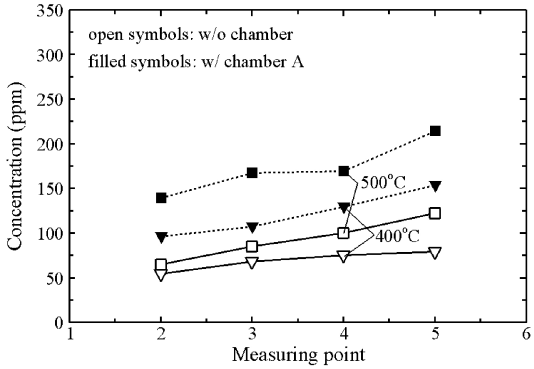

Figure 8 NH_3 concentration along the pipe with respect to inlet gas temperature at a fixed inlet velocity of 8 m/s with A-type mixing chamber: open and filled symbols denote NH_3 concentration with and without A-type mixing chamber, respectively

When the inlet velocity is 8 m/s, the NH_3 concentrations along the length are shown in Figure 8. For the inlet temperatures of 400℃ and 500℃, the average NH_3 concentrations for all measuring points with the mixing chamber are about 177% and about 164% greater, respectively, than those without the mixing chamber. With an implementation of the mixing chamber, the NH_3 concentrations for the inlet temperatures of 400℃ and 500℃ are 154 ppm and 215 ppm, respectively, at the fifth measuring point, and the NH_3 concentrations are about 195% and 143% higher, respectively, compared to those without the mixing chamber.

EFFECT OF INLET VELOCITY AND TEMPERATURE ON THE UREA THERMOLYSIS WITH B-TYPE MIXING CHAMBER.

The characteristics of urea thermal-decomposition are examined with the B-type mixing chamber under the conditions

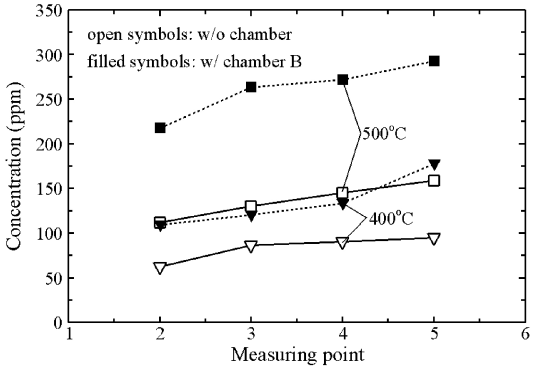

Figure 9 NH_3 concentration along the pipe with respect to inlet gas temperature at a fixed inlet velocity of 4 m/s with B-type mixing chamber: open and filled symbols denote NH_3 concentration with and without B-type mixing chamber, respectively

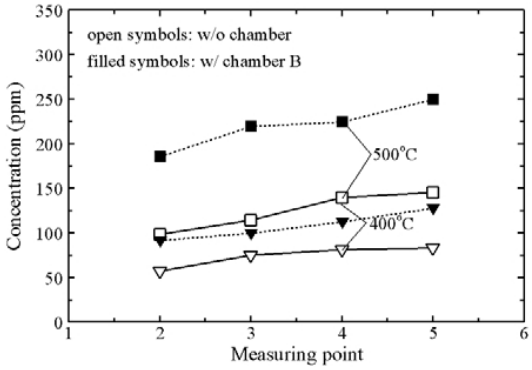

Figure 10 NH_3 concentration along the pipe with respect to inlet gas temperature at a fixed inlet velocity of 6 m/s with B-type mixing chamber: open and filled symbols denote NH_3 concentration with and without B-type mixing chamber, respectively

of inlet velocity (4, 6 m/s) and temperature (400, 500℃).

Figure 9 represents the NH_3 concentration profiles along the axial distance with and without the mixing chamber for varying inlet temperature at the inlet velocity of 4 m/s. At the inlet gas temperatures of 400℃ and 500℃, the average NH_3 concentrations over all measuring points with the mixing chamber are about 164% and about 191% larger, respectively, compared to those without the mixing chamber. The NH_3 concentrations at the fifth measuring point are 178 ppm and 293 ppm for respective inlet gas temperatures, and the concentrations are about 187% and about 184% higher, respectively, than those without the mixing chamber.

The NH_3 concentration distributions along the distance corresponding to the inlet velocity of 6 m/s are depicted in Figure 10. For the inlet gas temperatures of 400℃ and 500℃, the average NH_3 concentrations with the mixing chamber are about 147% and about 179% greater, respectively, than those without the mixing chamber. For respective inlet temperatures, the NH_3 concentrations at the fifth measuring point are 128 ppm and 250 ppm, respectively, and the NH_3 concentrations are about 154% and about 172% higher, respectively, than those without the mixing chamber.

CONCLUSIONS

In the present work, experiments are carried out by varying inlet velocities (4~12 m/s) and temperatures (350~500℃) to analyze the characteristics of urea thermal-decomposition. Two types of the mixing chambers are designed and fabricated to maximize the thermal-decomposition of urea. The thermal-decomposition characteristics of urea to isocyanic acid (HNCO) and ammonia (NH_3) are experimentally examined without and with mixing chamber.

NH_3 concentration produced from urea thermal-decomposition increases with increasing inlet gas temperature and it is caused by activation of urea thermolysis. And the concentration increases along the axial distance from injection nozzle due to an increase residence time in exhaust pipe. And NH_3 concentration decrease with increasing inlet velocity because of the shorten residence time in exhaust pipe.

At the inlet temperatures below 350℃ the NH_3 concentration produced through urea thermolysis is very small due to the deactivated thermal-decomposition of urea. And when the inlet velocity is 12 m/s, regardless of the inlet temperature and measuring points, the NH_3 concentration is very small because of shortened residence time of UWS in exhaust pipe.

At the inlet velocities are 6 m/s, 8 m/s, the average NH_3 concentrations with A-type mixing chamber over all measuring points are about 170%, about 177% larger, respectively, compared to those without the mixing chamber at the inlet gas temperature of 400℃, and about 157%, about 164% higher, respectively, than those without the mixing chamber at the inlet gas temperature of 500℃.

The average NH_3 concentrations with an implementation of B-type mixing chamber over all measuring points at the inlet velocities of 4 m/s and 6 m/s increase by about 164% and 147%, respectively, compared to those without the mixing chamber at the inlet gas temperature of 400℃, and increase by about 192% and 179%, respectively, compared to those without the mixing chamber at the inlet gas temperature of 500℃

ACKNOWLEDGMENTS

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REFERENCES

[1]Shimizu K., and Satsuma A., Hydrogen assisted urea-SCR and NH_3-SCR with silver-alumina as highly active and SO_2-tolerant de-NO_X catalysis, *Applied Catalysis B: Environmental*, Vol.77, 2007, pp. 202-205

[2]Koebel M., Elsener M., Kleemann M.,Urea SCR: A Promising Technique to Reduce NO_X Emissions from Automotive Diesel Engines, *Catalysis Today*, Vol. 59, 2000, pp. 335-345

[3]Kleemann M., Elsener M., Koebel M., Wokaun A., Hydrolysis of Isocyanic Acid on SCR Catalysts, *Industrial & engineering chemistry research*, Vol. 39, 2000, pp. 4120-4126

[4]Nishioka A., Amou K., Yokota H., and Murakami T., A Urea-Dosing Device for Enhancing Low-Temperature Performance by Active-Ammonia Production in an SCR System, *SAE International*, 2008-01-1026

[5]Sullivan J.A., and Doherty J.A., NH_3 and urea in the selective catalytic reduction of NO_X over oxide-supported copper catalysts, *Applied Catalysis B: Environmental*, Vol.55, 2005, pp. 185-194

[6]Koebel M., and Strutz E.O., Thermal and Hydrolytic Decomposition of Urea for Automotive Selective Catalytic Reduction Systems: Thermochemical and Practical Aspects, *Industrial & engineering chemistry research*, 2003, Vol. 42, pp. 2093-2100

[7] Karin Ax., Hermann F., Robert S., Michael H., and Agba S., Influ-ence of liquid binder dispersion on agglome-ration in an intensive mixer, *Powder Technology*, 2008, Vol.179 pp.190–194