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Characteristics of commercial selective catalytic reduction catalyst for the oxidation of gaseous elemental mercury with respect to reaction conditions

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Abstract−The performance of V_2O_5/TiO_2-based commercial SCR catalyst for the oxidation of gaseous elemental mercury (Hg^0) with respect to reaction conditions was examined to understand the mechanism of Hg^0 oxidation on SCR catalyst. It was observed that a much larger amount of Hg^0 adsorbed on the catalyst surface under oxidation condition than under SCR condition. The activity of commercial SCR catalyst for Hg^0 oxidation was negligible in the absence of HCl, regardless of reaction conditions. The presence of HCl in the reactant gases greatly increased the activity of SCR catalyst for the oxidation of Hg^0 to oxidized mercury (Hg^{2+}) such as $HgCl_2$ under oxidation condition. However, the effect of HCl on the oxidation of Hg^0 was much less under SCR condition than oxidation condition. The activity for Hg^0 oxidation increased with the decrease of NH_3/NO ratio under SCR condition. This might be attributed to the strong adsorption of NH_3 prohibiting the adsorption of HCl which was vital species promoting the oxidation of Hg^0 on the catalyst surface under SCR condition.

Key words: Mercury Oxidation, Selective Catalytic Reduction, Hydrogen Chloride, Elemental and Oxidized Mercury

INTRODUCTION

Among the various environmental problems, mercury emission from combustion sources is a major concern, since mercury is a toxic and persistent pollutant that accumulates in the food chain and has neurological health effect [1,2]. Atmospheric mercury is a global problem with many natural and anthropogenic emission sources. Coal-fired power plants have been known to be the major anthropogenic source of mercury emissions [3,4]. The U.S. EPA recently promulgated the Clean Air Mercury Rule (CAMR) to permanently cap and reduce emissions of mercury from coal-fired power plants [5]. In Korea, more strict regulation of mercury emission is also notified to apply to major emission sources such as power plants, sintering plants and cement manufacturing plants from 2010.

A variety of technologies have been studied to control the mercury emission from coal-fired power plants, such as conventional air pollution control devices [6,7], sorbent injection [8-10], electrocatalytic oxidation [11], photochemical oxidation [12], oxidizing agents injection and catalytic oxidation [13,14]. Mercury exists in three forms in coal-fired flue gas: elemental (Hg^0), oxidized (Hg^{2+}), and particle-bound (Hg(p)). Hg^{2+} and Hg(p) are relatively easy to remove from flue gas by using typical air pollution control devices (APCDs) such as ESPs (electrostatic precipitators) and wet-FGD (flue gas desulfurization). Elemental mercury (Hg^0), however, is difficult to capture, since it is insoluble in water. Among the technologies being considered for mercury reduction in coal-fired power plants is thus the combination of a catalyst and a wet scrubber; the catalyst oxidizes Hg^0 to Hg^{2+} and the oxidized mercury is subsequently absorbed by the scrubber system [15]. Catalysts capable of significant conversion of Hg^0 to Hg^{2+} would have tremendous value because the oxidized mercury can be removed concurrently with acid gases during the FGD process. Oxidation catalysts studied to date fall into one of three groups: V_2O_5/TiO_2-based SCR catalysts [16-18], carbon-based catalysts [19-21] and metals and metal oxides [22-25]. Selective catalytic reduction (SCR) has been a well-developed and commercialized technology for controlling emissions of NOx from power plants [26-28]. In addition to NOx control SCR catalyst has been found to affect the mercury speciation by altering Hg^0 to Hg^{2+}. The efficacy of SCR has been tested at the laboratory, pilot and full scale operation for a variety of conditions of HCl and NO concentrations, NH_3/NO ratios, temperatures and coal types [16-18]. Increasing the emissions of Hg^{2+} across SCR catalyst allows for high reduction of mercury emission because highly water soluble Hg^{2+} or Hg^{2+}-derived species such as $HgCl_2$ can be removed in downstream equipments such as ESPs and wet-FGD systems. Therefore, the co-benefit of increased Hg^{2+} through the SCR catalyst is very important to the overall control of mercury emissions from coal-fired power plants.

In the present study, the performance of a V_2O_5/TiO_2-based commercial SCR catalyst for the oxidation of Hg^0 was studied to understand the mechanism of mercury oxidation on SCR catalyst. For the better understanding of mercury oxidation on SCR catalyst, the adsorption behavior of Hg^0 on the catalyst surface and the activities of mercury oxidation were compared with respect to reaction conditions such as oxidation and SCR conditions. The effect of HCl present in the reactant gas stream on the mercury oxidation was also examined for both reaction conditions.

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Table 1. Chemical compositions of commercial SCR catalyst employed in this study

Content (wt%)		
V_2O_5	WO_3	S
1.68	7.60	0.29

Table 2. Physical properties of commercial SCR catalyst employed in this study

BET surface area (m^2/g)	Pore volume (m^3/g)	Average pore diameter (Å)
79.4	0.278	132.7

EXPERIMENTAL

1. Catalyst Properties

The catalyst employed in this study was a V_2O_5/TiO_2-based commercial SCR catalyst for coal-fired power plants. The physicochemical properties of the commercial SCR catalyst employed in this study are summarized in Table 1. The catalyst composition was determined by inductively coupled plasma (ICP) analyzer. The catalyst contains 1.68 and 7.60 wt% of V_2O_5 and WO_3, respectively, which are typical compositions of SCR catalyst for commercial application to coal-fired power plant. The catalyst has a small amount of sulfur originating from the TiO_2, since the catalyst support, TiO_2, is generally manufactured by sulfuric process. The catalyst surface area measured by BET method with Micromeritics ASAP 2010 apparatus using liquid N_2 at 77 K was 79.4 m^2/g. The average pore diameter and pore volume obtained from N_2 desorption isotherm were 132.7 Å and 0.278 m^3/g, respectively.

2. Catalytic Reactor System

The catalytic activity for mercury oxidation was examined in a fixed-bed flow reactor typically containing 1 g of 20/30 mesh size pellet obtained by crushing honeycomb-type SCR catalyst. A gas mixture containing 3% oxygen in N_2 balance was fed to the reactor system through mass flow controller (Brooks Model 5850E) under oxidation condition experiment. Under SCR condition experiment, NO and NH_3 of 500 ppm, respectively, were additionally fed to the compositions of oxidation condition. To examine the effect of HCl on the oxidation of mercury, 10-50 ppm of HCl was mixed with reactant gas stream, when added. HCl was injected just above the catalyst bed to avoid the possibility of NH_4Cl formation under SCR condition. The total reactant gas flow rate was maintained as 2 L/min. A quartz tube of inner diameter 3/8" was used as the reactor to avoid the adsorption and reaction of mercury species on the reactor surface. For the mercury oxidation experiment, the gaseous Hg^0 was generated by flowing N_2 carrier gas to a temperature-controlled impinger containing liquid mercury. The concentration of mercury could be controlled by adjusting the impinger temperature and carrier gas flow rate. The mercury vapor in N_2 carrier gas was mixed with simulated reactant gas mixture just before the catalyst bed. The Hg^0 concentration in the reactant gases was maintained as about 50 μg/m^3 for all experiments. Thermocouples were positioned near the inlet and outlet of the catalyst bed for controlling and monitoring the reactor temperature.

Mercury concentration was continuously measured by cold-vapor atomic absorption spectrometer (VM-3000, Mercury Instruments Analytical Technologies) employing the resonance absorption of the mercury atoms at a wavelength of 253.7 nm. The spectrometer can measure only the concentration of Hg^0. Therefore, to measure the concentration of total mercury including elemental and oxidized mercury in reactant gases, an impinger containing 10 wt% stannous chloride ($SnCl_2$) solution was located at just after the reactor. The $SnCl_2$ solution reduces all oxidized mercury species to elemental mercury. The concentration of oxidized mercury can be measured by the difference of mercury concentration after and before passing the $SnCl_2$ solution. The mercury oxidation efficiency can be calculated by the following formula:

$$\text{Hg oxidation efficiency } (\%) = \frac{[\text{Hg}^{2+}]_{out}}{[\text{Hg}^0]_{out} + [\text{Hg}^{2+}]_{out}} \qquad (1)$$

where, $[Hg^0]_{out}$ and $[Hg^{2+}]_{out}$ are the concentrations of elemental and oxidized mercury at the reactor outlet, respectively.

The concentration of NO was analyzed by on-line chemiluminescence NO-NO_2 analyzer (Thermo Electron Co., Model 42C).

RESULTS AND DISCUSSION

1. Adsorption Behavior of Elemental Mercury (Hg^0) with Respect to Reaction Conditions

To investigate the adsorption behavior of Hg^0 on the catalyst surface, the Hg^0 concentration at the reactor outlet was continuously monitored with time on-stream under SCR condition, as shown in Fig. 1. The Hg^0 concentration at the reactor outlet showed a transient behavior with time on-stream. The time required to reach the steady concentration of Hg^0 was significantly dependent on the reaction temperature under SCR condition. It took several hours to reach steady concentration of Hg^0 at 250 °C, whereas less than an hour at higher temperature of 300 °C. The steady concentration of Hg^0 was about 45 μg/m^3, indicating that the removal efficiency of Hg^0 was less than 10% even at 250 °C. Since the time required to reach steady concentration was much longer at lower temperature, the transient

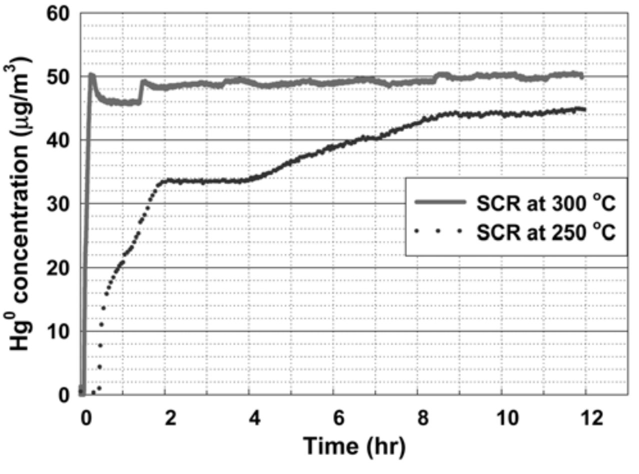

Fig. 1. Change of the concentration of elemental mercury at the reactor outlet under SCR condition; Gas flow rate=2 L/min, [NO]=[NH_3]=500 ppm, [O_2]=3%, [Hg]=50 μg/m^3, [N_2] balance.

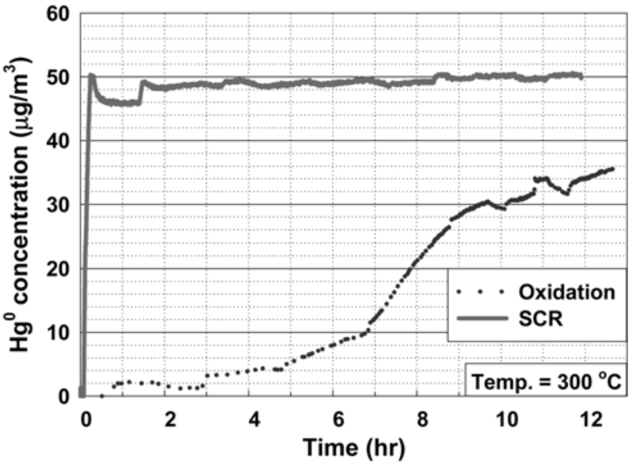

Fig. 2. Comparison of the concentration of elemental mercury at the reactor outlet under SCR and oxidation conditions; Gas flow rate=2 L/min, $[O_2]$=3%, [Hg]=50 μg/m³, $[N_2]$ balance, [NO]=$[NH_3]$=500 ppm (under SCR condition).

behavior of Hg^0 concentration at the reactor outlet might be due to the adsorption of Hg^0 on the catalyst surface. The strong dependence of Hg^0 adsorption on the temperature might suggest the physical adsorption rather than chemical adsorption of Hg^0 on the SCR catalyst.

The concentration of Hg^0 at the reactor outlet was compared for both oxidation and SCR conditions at 300 °C, as shown in Fig. 2. The transient behavior of Hg^0 concentration at the reactor outlet was much more apparent under oxidation condition than under SCR condition. The catalyst surface was saturated with Hg^0 in an hour under SCR condition, whereas much longer time over 12 hrs was needed to saturate the catalyst surface with Hg^0 under oxidation condition. The breakthrough curve for Hg^0 adsorption showed that much larger amount of Hg^0 could be adsorbed on the catalyst surface under oxidation condition than under SCR condition.

The Hg^0 concentration at the reactor outlet was continuously monitored under the alternate operation of oxidation and SCR conditions as shown in Fig. 3. The transient behavior of Hg^0 concentration at the reactor outlet was fully reversible with respect to reaction conditions. When the flow of NH_3 and NO was shut off (oxidation condition), the concentration of Hg^0 rapidly decreased and maintained nearly zero concentration. This result reveals the high capacity of SCR catalyst for Hg^0 adsorption under oxidation condition. However, the Hg^0 concentration abruptly increased and reached the original concentration of Hg^0 by the injection of NH_3 and NO again (SCR condition). The NH_3 injected seems to cause mercury to desorb from the catalyst surface by changing the oxidation condition to SCR condition. This means that NH_3 and Hg^0 compete for the same sites on the catalyst surface. However, NH_3 preferentially adsorbs and inhibits the adsorption of Hg^0 on the catalyst surface under SCR condition. Therefore, NH_3 dominantly adsorbs on the SCR catalyst surface, when both components of NH_3 and Hg^0 are present in the reactant gases. Note that NH_3 present in the reactant gas stream under SCR condition has been well known to easily and strongly adsorb on the V_2O_5/TiO_2-based SCR catalyst surface [29,30]. This may be the reason for the much less adsorption of Hg^0 under SCR condition than oxidation condition as identified by the longer transition time to reach steady concentration in Fig. 2.

2. Effect of HCl on the Oxidation Efficiency of Hg^0 with Respect to Reaction Conditions

In the coal-derived flue gases, chlorine is believed to be mainly present in the form of HCl. HCl is a most important species affecting mercury oxidation, since the major oxidized mercury species in coal-derived flue gas is $HgCl_2$ [31]. For the removal of Hg^0 by conventional APCDs such as wet-FGD and ESPs, Hg^0 has to be converted into oxidized Hg such as $HgCl_2$ which is much more soluble than Hg^0. Therefore, the effect of HCl on the oxidation of Hg^0 to oxidized Hg such as $HgCl_2$ has been most widely studied in the area of mercury removal technologies [21-23]. In this study, we already observed that the SCR catalyst had negligible activity for Hg^0 oxidation in the absence of HCl, regardless of reaction conditions of oxidation and SCR as shown in Figs. 1 and 2. The effect of HCl on the oxidation of Hg^0 was examined with respect to reaction conditions.

Fig. 4 shows a typical concentration profile of Hg^0 measured by

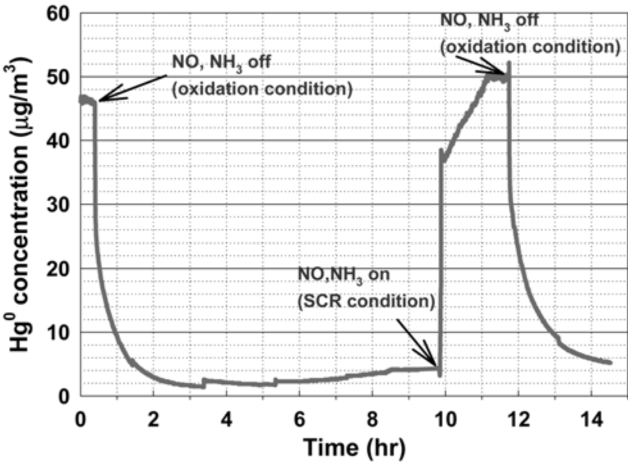

Fig. 3. Concentration of elemental mercury at the reactor outlet under the cyclic operation of oxidation and SCR conditions; Gas flow rate=2 L/min, $[O_2]$=3%, [Hg]=50 μg/m³, $[N_2]$ balance, [NO]=$[NH_3]$=500 ppm (under SCR condition).

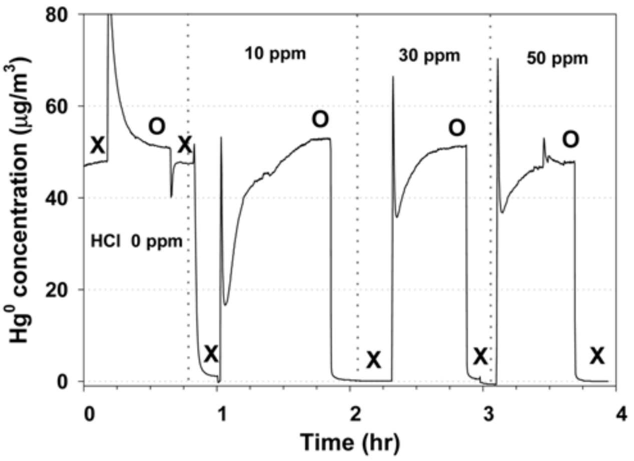

Fig. 4. Typical concentration profile of elemental mercury before (symbol X) and after (symbol O) $SnCl_2$ solution at the reactor outlet under oxidation condition; Gas flow rate=2 L/min, $[O_2]$=3%, [Hg]=50 μg/m³, [HCl]=0-50 ppm, $[N_2]$ balance.

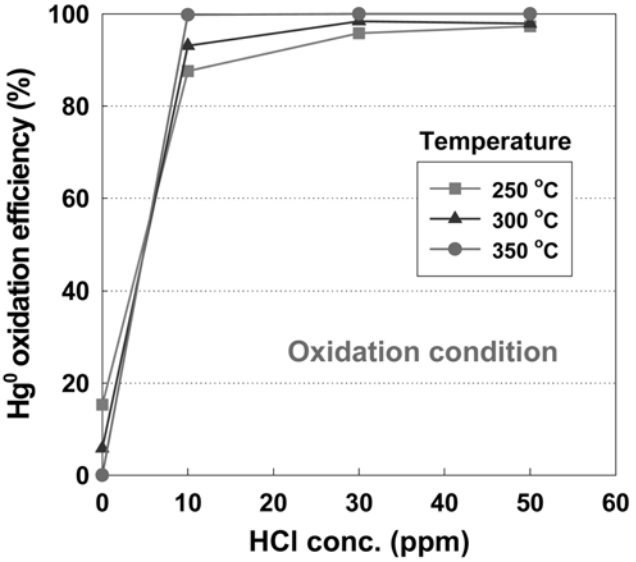

Fig. 5. Effect of HCl concentration on the oxidation of elemental mercury to oxidized mercury under oxidation condition; Gas flow rate=2 L/min, [O_2]=3%, [Hg]=50 μg/m^3, [HCl]=0-50 ppm, [N_2] balance.

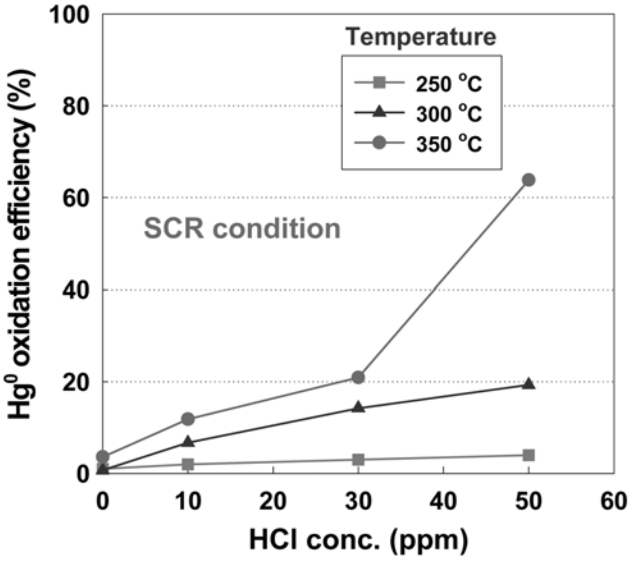

Fig. 6. Effect of HCl concentration on the oxidation of elemental mercury to oxidized mercury under SCR condition; Gas flow rate=2 L/min, [NO]=[NH_3]=500 ppm, [O_2]=3%, [Hg]=50 μg/m^3, [HCl]=0-50 ppm, [N_2] balance.

continuous Hg^0 monitoring system in the presence of HCl of 0-50 ppm under oxidation condition. When the reactant gases were passing through the $SnCl_2$ solution, oxidized mercury could be reduced to Hg^0. Therefore, mercury concentration measured after passing the $SnCl_2$ solution is total mercury including elemental and oxidized mercury, while the mercury concentration before passing the $SnCl_2$ solution is only the Hg^0 in the reactant gases. In the absence of HCl, only less than 10% of Hg^0 was converted to oxidized mercury. On the other hand, almost all of Hg^0 was converted to oxidized mercury in the presence of HCl of 10-50 ppm as shown in Fig. 4. The gas phase HCl was identified as a crucial species for the oxidation of Hg^0.

Based on the concentration profile obtained for each reaction condition, the effect of HCl on the activity of Hg^0 oxidation to oxidized mercury was examined at a reaction temperature of 250-350 °C under oxidation condition as shown in Fig. 5. In the absence of HCl, the activity of commercial SCR catalyst for mercury oxidation was very low less than 20% at all reaction temperatures examined in this study. However, the activity greatly increased by the addition of HCl to the reactant gas stream. More than 90% conversion for mercury oxidation was observed at all reaction temperatures over 250 °C. The oxidation of Hg^0 by HCl has been known to take place by the following reaction [31].

$$Hg^0 + 2HCl + \frac{1}{2}O_2 \leftrightarrow HgCl_2 + H_2O \tag{2}$$

This result indicates that the presence HCl is of importance to the oxidation of Hg^0 to oxidized mercury.

The activity of SCR catalyst for Hg^0 oxidation under SCR condition was examined and shown in Fig. 6. The activity of SCR catalyst for Hg^0 oxidation was also negligible in the absence of HCl. The activity increased with the increase of HCl concentrations and reaction temperatures. However, the effect of HCl on the oxidation of Hg^0 to oxidized mercury was very low under SCR condition compared to that under oxidation condition. At lower temperature of 250 °C, the activity for Hg^0 oxidation was negligible even in the presence of HCl up to 50 ppm. Although the activity for Hg^0 oxidation increased with the increase of HCl concentration, the conversion was still very low less than 20% even in the presence of HCl 50 ppm at 300 °C. The maximum oxidation efficiency obtained under SCR condition was about 60% at the reaction temperature of 350 °C and HCl concentration of 50 ppm in our experimental conditions. The large difference in the oxidation efficiency between oxidation and SCR condition seems to be closely related to the difference in gas compositions. Note that NH_3 and NO, which were not present under oxidation condition, were fed with the reactant gas under SCR condition. HCl should adsorb on the catalyst surface to oxidize Hg^0 to oxidized mercury. However, NH_3 might prohibit the adsorption of HCl under SCR condition, since NH_3 is the dominant species adsorbed on V_2O_5/TiO_2-based SCR catalyst during the course of SCR reaction. The NO reduction by NH_3 on V_2O_5/TiO_2-based SCR catalyst has been well known to proceed via Eley-Rideal mechanism between NH_3 adsorbed on the catalyst surface and gas phase or weakly-bound NO [32,33]. This seems to the main reason for low activity for Hg^0 oxidation under SCR condition than oxidation condition.

To identify the effect of NH_3 on the oxidation of Hg^0, the oxidation efficiency was examined with the change of NH_3/NO ratio as shown in Fig. 7. The Hg^0 oxidation efficiency significantly increased with the decrease of NH_3/NO ratio and was nearly 100% at NH_3/NO ratio less than 0.5. As already mentioned, NH_3 would preferentially adsorb on the catalyst surface when NH_3 and HCl were present simultaneously in the reactant gases. However, the surface coverage of NH_3 on the catalyst surface would decrease with the decrease of NH_3/NO ratio by the SCR reaction occurring simultaneously with Hg^0 oxidation, since NH_3 could be consumed by SCR reaction pro-

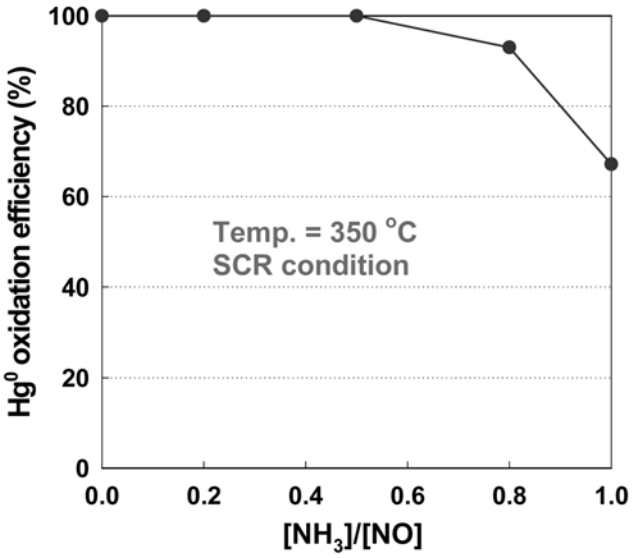

Fig. 7. Oxidation efficiency of elemental mercury to oxidized mercury with respect to NH_3/NO ratio under SCR condition at 350 °C; Gas flow rate=2 L/min, [NO]=500 ppm, $[NH_3]$= 0-500 ppm, $[O_2]$=3%, [Hg]=50 μg/m^3, [HCl]=50 ppm, $[N_2]$ balance.

ceeding at equimolar ratio between NH_3 and NO. Therefore, there would be more available sites for the adsorption of HCl promoting the Hg^0 oxidation with the decrease of NH_3/NO ratio even under SCR condition. This result might give concrete evidence of the role of NH_3 reducing the activity for Hg^0 oxidation under SCR condition than under oxidation condition at the same reaction temperature and HCl concentration as observed in Figs. 5 and 6. This result indicates that HCl competes with NH_3 for the same sites on the surface of SCR catalyst. It also means the adsorption of HCl on the catalyst surface is of importance for the oxidation of Hg^0 to oxidized mercury.

Based on the effect of NH_3/NO ratio and reaction conditions on Hg^0 oxidation, it can be proposed that Hg^0 oxidation occurs via an Eley-Rideal mechanism by which adsorbed HCl reacts with gas phase or weakly bound Hg^0 under SCR condition. This mechanism explains well the experimental results on the adsorption behavior and oxidation of Hg^0 with respect to reaction conditions of oxidation and SCR obtained in this study.

CONCLUSIONS

The adsorption behavior of Hg^0 on the V_2O_5/TiO_2-based commercial SCR catalyst significantly depended on the reaction conditions. A much larger amount of Hg^0 adsorbed on the catalyst surface under oxidation condition than under SCR condition. This might be mainly due to the strong adsorption of NH_3 inhibiting the adsorption of Hg^0 on the catalyst surface. The activity of commercial SCR catalyst for mercury oxidation was negligible in the absence of HCl, regardless of reaction conditions of oxidation and SCR. The presence of HCl in the reactant gases greatly increased the activity of SCR catalyst for the oxidation of Hg^0 to oxidized mercury under oxidation condition. However, the effect of HCl on the oxidation of Hg^0 was much less under SCR condition than oxidation condition. NH_3 is also believed to prohibit the adsorption of HCl promoting the oxidation of Hg^0 to oxidized Hg on the catalyst surface under SCR condition. The inhibition of Hg^0 oxidation by NH_3 was also confirmed by the observation of the increase of Hg^0 oxidation with the decrease of NH_3/NO ratio under SCR condition. Based on the experimental results observed in this study, it can be proposed that Hg^0 oxidation occurs via an Eley-Rideal mechanism by which adsorbed HCl reacts with gas phase or weakly bound Hg^0 under SCR condition.

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