

Study of Speciation of Mercury under Simulated SCR NO_x Emission Control Conditions

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ABSTRACT

Selective catalytic reduction (SCR) technology is being increasingly applied for controlling emissions of nitrogen oxides (NO_x) from coal-fired boilers. Some recent field and pilot studies suggest that the operation of SCR could affect the chemical form of mercury species (speciation) in the coal combustion flue gases. The speciation of mercury is an important factor that influences the control and environmental fate of mercury emissions from coal combustion. The vanadium and titanium oxides (V₂O₅, TiO₂), used commonly in the vanadia-titania SCR catalyst for catalytic NO_x reduction, may also promote the formation of oxidized mercury (Hg²⁺).

The work reported in this paper focuses on the impact of SCR on elemental mercury (Hg⁰) oxidation. Bench-scale experiments were conducted to investigate Hg⁰ oxidation in the presence of simulated coal combustion flue gases and under SCR reaction conditions. Flue gas mixtures with different concentrations of hydrogen chloride (HCl) and sulfur dioxide (SO₂) for simulating the combustion of bituminous coals and sub-bituminous coals were tested in these experiments. The effects of HCl and SO₂ in the flue gases on Hg⁰ oxidation under SCR reaction conditions were studied. It was observed that HCl is the most critical flue gas component that causes conversion of Hg⁰ to Hg²⁺ under SCR reaction conditions. The importance of HCl for Hg⁰ oxidation found in the present study provides a strong scientific base for the apparent coal-type dependence observed for Hg⁰ oxidation occurring across the SCR reactors in the field.

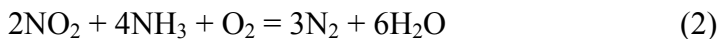
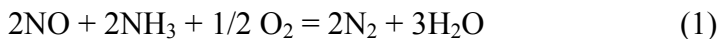
INTRODUCTION

Coal combustion is a major source of mercury emissions that have caused great environmental health concern.¹ The chemical form of mercury species (speciation) is an important factor that influences the control and environmental fate of mercury emissions from coal combustion.² It is known that controlling the emissions of oxidized mercury, the water-soluble mercuric chloride (HgCl₂) in particular, is much easier than controlling the insoluble elemental mercury Hg⁰.³ Emissions of nitrogen oxides (NO_x) are another major environmental concern from coal combustion; the emissions are associated with ground level ozone, acid rain formation, forest damage, degradation of visibility, and formation of fine particles in the atmosphere.⁴ Several

regulatory programs such as the Acid Rain NO_x regulations, the Ozone Transport Commission's NO_x Budget Program, and Ozone Transport rulemakings require significant reductions in NO_x emissions from coal combustion sources. As a result of these more stringent NO_x emission control requirements, selective catalytic reduction (SCR) technology is being increasingly applied for controlling NO_x emissions from coal-fired boilers due to its high NO_x emission control efficiency (>80%). The operation of SCR could affect the speciation of mercury in the coal combustion flue gases. Early field measurements conducted in Europe indicated that SCR reactors installed for NO_x emission control promote the formation of oxidized Hg species.⁵ A recent field study conducted in the United States also found increases in Hg²⁺ species across the SCR reactors.⁶

The effect of SCR operation on mercury speciation appears to be dependent on coal type. Boilers burning bituminous coals, which typically have high chloride (Cl) and sulfur (S) content, have been observed to have significant increases in Hg²⁺ species across their SCR reactors.⁶ However, a subbituminous coal from the Powder River Basin (PRB), which contains significantly lower Cl and S content and higher calcium (Ca) content compared to those of the bituminous coals, showed very little change in mercury speciation across a SCR reactor.⁶

In coal combustion flue gases, NO_x are reduced to nitrogen (N₂) and water (H₂O) in a SCR reactor consisting of metal oxide catalysts such as TiO₂-supported V₂O₅. A reducing agent such as ammonia (NH₃) or urea (CH₄ON₂) is injected into the SCR reactor operating at temperatures of about 350 °C. The NO_x reductions proceed according to the following reactions:



It has been indicated in laboratory-scale experiments that metal oxides, including V₂O₅ and TiO₂, could promote the transformation of Hg⁰ to Hg²⁺ in combustion flue gases.⁷ Recent pilot-scale tests also suggested that SCR has the potential to impact mercury speciation, and the effects appear to be dependent upon coal type.⁸ However, the reactions that are important to transform Hg⁰ into Hg²⁺ in the SCR reactor are not well understood.

The work reported here is bench-scale research that focus on studying Hg⁰ oxidation in the presence of simulated coal combustion flue gases and under SCR NO_x emission control conditions. The effects of HCl and SO₂ in the flue gases on Hg⁰ oxidation were studied. Our previous bench-scale study found that NO_x and HCl promote Hg⁰ oxidation, while SO₂ and H₂O reduce Hg⁰ oxidation in the presence of coal combustion fly ashes.⁹ It was hypothesized that transition metal oxides, such as iron oxide (Fe₂O₃), present in coal fly ash may be instrumental in catalytic oxidation of Hg⁰. In the present study, the effects on Hg⁰ oxidation of the acidic flue gas components and the basic NH₃ gas added for NO_x reduction reactions were evaluated in the presence of a SCR catalyst. The emissions of acidic flue gases generated by the combustion of different types of coals depend upon the coal type and its composition. Emission of acidic flue gases may be reduced significantly in the presence of NH₃, and they may also be catalytically converted into other, more reactive, species under the SCR emission control conditions. Such

changes could, in turn, have a significant effect on Hg^0 oxidation. The objective of the study is to gain a better understanding of the complex, confounding interactions of flue gas components occurring under SCR emission control conditions and resulting in changes in mercury speciation in coal combustion flue gases.

EXPERIMENTAL

SCR Reactor System

A bench-scale reactor system was designed and constructed to simulate the SCR reaction conditions for studying Hg^0 oxidation. A schematic of the reactor system is shown in Figure 1. The system consists of the preheating and premixing sections, the mercury generation unit, the SCR reactor, and the on-line reactor effluent measurement unit. Flue gas components, including carbon dioxide (CO_2), SO_2 , air, and N_2 , were mixed and preheated to 350 °C and then mixed with another preheated stream of nitrogen oxide (NO) and HCl at the main heating section. Water was also pumped into the hot section at a calibrated rate, for simulating the moisture content in flue gas, and mixed with the other flue gas components. Ammonia was preheated and flowed into the static mixing section to get better mixing with the other flue gas components. The gas mixture then flowed into the Pyrex reactor. The alkaline NH_3 reacts with the acid gas components in the gas mixture to form ammonium salts at temperatures lower than the SCR reaction temperatures. Therefore, the simulated flue gas mixture was preheated by the electrical furnaces and maintained at 350 °C by the temperature-controlled electrical heating tapes to prevent such reactions taking place and reducing the amount of NH_3 available for reducing NO_x . The fine-particle ammonium salts formed from the neutralization of NH_3 are sticky and would otherwise tend to deposit on the reactor walls and adsorb mercury species in the flue gas mixture. If not avoided through the use of such precautions, the deposition of ammonium salt particles on the reactor walls would make the measurement of trace mercury species in the reactor very difficult. Proper preheating and mixing of the gas mixture are very important for simulating the SCR reaction conditions and for measuring mercury speciation. All the heating and mixing sections upstream of the Pyrex reactor are made of stainless steel to minimize the corrosive effect of the acidic gases. Gas cylinders of nitrogen-diluted gaseous species were used instead of pure gases for some components (i.e., SO_2/N_2 , NO/N_2 , O_2/N_2 , and NH_3/N_2).

A mercury generation unit consisting of a mercury permeation tube surrounded by a temperature-controlled water bath was used to generate Hg^0 vapor for the oxidation experiments. The Hg^0 vapor was carried by a nitrogen stream and mixed with the flue gas mixture near the top of the reactor (4 cm in inner diameter and 35 cm in length). Details of the generation system are described elsewhere.³ The flue gas mixture containing Hg^0 and NH_3 passed through a honeycomb flow straightener to produce good velocity distribution before passing through the honeycomb SCR catalyst. A thermocouple was positioned near the inlet of the catalyst for controlling and monitoring the SCR catalyst temperature. A sampling tube was also positioned near the inlet of the flow straightener for taking gas samples close to the inlet of the SCR catalyst. The gaseous effluent stream exiting the SCR reactor passed through a drying tube to remove moisture in the flue gas. The dried gas then passed to an online ultraviolet dry SO_2 analyzer (Bovar Engineering, Inc., model 721AT2; accuracy of about $\pm 5\%$) for measuring SO_2 . An online chemiluminescent NO_x analyzer (Advanced Pollution Instrumentation, Inc., model

200AH; accuracy of about $\pm 5\%$) downstream of the SO₂ analyzer was used for measuring NO_x.

Test Procedures

A commercial SCR catalyst with a vanadia/titania formulation and a honeycomb configuration, designated as Catalyst A, was used in the study. A small piece (2.2 cm for both sides and 1.9 cm in length) of the catalyst sample was placed into the catalyst compartment of the SCR reactor. The reactor system was heated overnight at 425 °C under an N₂ atmosphere. This cleanup procedure was employed prior to each test for removing any adsorbed species in the catalyst in order to minimize the residue effect caused by the previous test. The temperature of the reactor was then lowered to 220 °C, and SO₂ and HCl gases were added to the reactor at the same levels for the next day's experiment. The temperature of the reactor was raised to 350 °C and maintained overnight. This catalyst preconditioning procedure was applied prior to each test. The next morning, the remaining components of the simulated flue gas mixture, CO₂, NO, H₂O, and NH₃, plus Hg⁰ were added into the reactor. The flows of the flue gas components were maintained at the levels designed for the test by using the mass flow controllers. The variability of the gas concentrations is about $\pm 2.5\%$. A constant total flow rate of 400 cm³/min [at a standard temperature of 25 °C and pressure of 101.4 kPa (STP)] was used for all tests. The concentrations of SO₂ and NO_x at the outlet of the reactor were monitored continuously for four hours by using the SO₂ and NO_x analyzers, respectively, to ensure that the NO_x reduction reached a steady state. Then the two gas analyzers were disconnected from the outlet of the reactor and the reactor outlet was connected to a sampling train. Sampling for mercury (under positive pressure) was then started with a sampling time for each test of 2 hr. The outlet flow rate was measured by using a Gilibrator electronic flow meter (model 2) every 10 minutes during the sampling.

Mercury Sampling Method

The mercury speciation method¹⁰ developed by Ontario Hydro (OH) was used for measuring Hg⁰ and Hg²⁺ in this study. The method was modified slightly for the bench-scale experiments with small volumetric gas flow, and the simulated flue gases sampled in the present study were free of particulates. No filter designed for capturing particulates, and smaller impingers (25 cm³) instead of the 100 cm³ impingers specified by the OH method, were used for the sampling. Smaller impingers were used for the much smaller gas volume (0.05 m³) sampled in this study than those (1 – 2.5 m³) targeted by the OH method. The smaller impingers were also equipped with screw caps designed for positive pressure sampling. In summary, the sampling train consists of a series of eight impingers immersed in an ice water bath. The first three impingers contain a 1 N potassium chloride (KCl) aqueous solution for capturing Hg²⁺. Hg⁰ is insoluble in the KCl solution and passed through the first three impingers. The next impinger contains a solution of 5 % nitric acid and 10 % hydrogen peroxide (H₂O₂) followed by three impingers containing a solution of 10 % sulfuric acid (H₂SO₄) and 4% potassium permanganate (KMnO₄), for capturing Hg⁰. The last impinger was empty to condense and remove any moisture remaining in the flue gas. The sample fractions captured by the different impingers were prepared and analyzed as specified in the OH method.¹⁰ Each prepared fraction was analyzed for total mercury by cold vapor atomic absorption.

Experimental Approach and Test Conditions

The approach employed in this study was to pass a simulated coal combustion flue gas mixture containing NH_3 and trace Hg^0 through a bench-scale SCR reactor under NO_x reduction conditions and measure mercury speciation at the outlet of the reactor. The HCl and SO_2 levels in the simulated flue gases were varied for simulating the combustion of PRB coals and bituminous coals with different sulfur and chlorine contents. Mercury speciation samplings near the inlet of the SCR reactor with the catalyst removed were conducted at the beginning and in the middle of the test program. It was found that the inlet concentration obtained from the two samplings was 19.3 ppb (± 1 ppb) with very little Hg^{2+} ($\sim 2\%$). The small volumetric flow of the bench-scale reactor makes the simultaneous samplings of inlet and outlet of the SCR reactor impractical. The simulated flue gas mixtures preheated to $350\text{ }^\circ\text{C}$ in order to avoid the formation of ammonium salts also makes the construction a by-pass of the SCR reactor for sampling reactor inlet mercury speciation impractical. Inert, Teflon valves needed for making the by-pass to be operated at such a high temperature are not available.

It has been suggested in the previous field^{5,6} and pilot⁸ studies that coal type seems to be the major factor which determines the impact of SCR on mercury speciation in the coal combustion flue gas. A series of four bench-scale tests were conducted in the present study to simulate the combustion of PRB and bituminous coals with different Cl and S contents. The compositions of the simulated flue gas mixtures used for these tests are shown in Table 1. A base flue gas mixture consists of 350 ppm NO_x , 315 ppm of NH_3 , 15% CO_2 , 3.5% O_2 , 5.3% H_2O , 19 ppb Hg^0 , and balance in N_2 , was used for all the tests. A sub-stoichiometric NH_3/NO_x ratio was used in the all tests in order to simulate the low NH_3 slip commonly practiced in the field. The concentrations of HCl and SO_2 gases added to the base gas mixture were varied for simulating the combustion of different types of coals. Test P1 simulated the combustion of a PRB coal that has low S and Cl contents such that most of the Cl in the coal would be converted into HCl in the combustion flue gas. Test P2 also simulated combustion of a PRB coal, but no HCl was added to the flue gas mixture in order to simulate a PRB coal in which a high calcium (Ca) content would result in all Cl released from the combustion of this coal being reacted with Ca such that no HCl would exist in the flue gas. Test B1 simulated the combustion of a low-sulfur and high-chlorine bituminous coal, and the combustion of a high-sulfur and low-chlorine coal was simulated by the B2 test. All the tests were conducted at $350\text{ }^\circ\text{C}$ and at a constant total flue gas flow rate of $400\text{ cm}^3/\text{min}$ (at STP). The calculated space velocity for the tests was 2609 hr^{-1} , similar to those used in the field. Sampling for mercury speciation using the OH method was started after the simulated flue gas mixture passed through the SCR reactor for four hours and the NO_x concentration measured at the outlet of the reactor by the NO_x analyzer showed that the concentration reached a steady state value.

RESULTS AND DISCUSSION

NO_x Reduction Results

Results of the NO_x reduction for the tests are summarized in Table 2. The NO_x reductions of 85 to 88% were observed for the tests. Figure 2 shows the NO_x concentration profile at the outlet of the SCR reactor for Test B1. Similar NO_x concentration profiles were observed for the other three tests. During the first few minutes after the remaining components of the simulated flue gas mixture (CO_2 , NO , H_2O , and NH_3 , plus Hg^0) were added into the reactor, the NO_x

concentration remained near zero. The pre-conditioning of the SCR catalyst by SO₂ overnight prior to the NO_x reduction experiment started may make the catalyst very reactive for the initial period of reduction of NO_x. Then the outlet NO_x concentration increased quickly to about 120 ppm and reduced slowly to reach a steady state concentration of less than 50 ppm in about 3 hr.

Table 1. Summary of simulated flue gas composition

Test No.	P1	P2	B1	B2
HCl Concentration (ppm)	8	0	204	134
SO ₂ Concentration (ppm)	280	280	934	2891
NO _x Concentration (ppm)	350	350	350	350
NH ₃ Concentration (ppm)	315	315	315	315
CO ₂ Concentration (%)	15	15	15	15
O ₂ Concentration (%)	3.5	3.5	3.5	3.5
H ₂ O Concentration (%)	5.3	5.3	5.3	5.3
Hg ⁰ concentration (ppb)	19	19	19	19

Table 2. Summary of NO_x reduction results

Test No.	P1	P2	B1	B2
Outlet NO _x Concentration (ppm)	44	52	44	43
NO _x Reduction (%)	87	85	87	88

It seems that the fresh SCR catalyst used in the tests exhibited strong NO_x reduction catalytic activity under the widely different SO₂ concentrations (280 –2891 ppm) employed in the tests. The levels of NO_x reduction (85 –87%) obtained from the bench-scale SCR reactor are similar to those observed in the field, suggesting that the reactor system used in the present study simulates closely the SCR NO_x emission control conditions in the field.

Effect of Flue Gas Species on Hg⁰ Oxidation

The mercury speciation results measured in the present study are summarized in Table 3. Two mercury speciation samples were taken at the inlet of the SCR catalyst by using the OH method after Tests P1 and B1. Two OH samples were also taken at the outlet of the mercury generation unit at the beginning of the test program. The total mercury concentrations (19.3±1.0 ppb) with very little Hg²⁺ (0.5 ppb) measured near the inlet of the catalyst are the same as those (19.2 ± 0.1 ppb total Hg with 0.5 ppb Hg²⁺) measured at the outlet of the mercury generation unit. The two inlet OH speciation results obtained in the presence of two different simulated flue gas mixtures (Test P1 and Test B1) showed Hg⁰ as the only mercury species. The consistent inlet results suggest that the presence of HCl and SO₂ at different concentrations in the simulated flue gas mixture have very little effect to change the speciation of mercury in the mixture prior to passing through the SCR catalyst. The results also suggest no gas phase, homogeneous oxidation of Hg⁰ at the inlet.

Table 3. Summary of mercury speciation results

Test No.	Inlet	P1	P2	B1	B2
Hg ⁰ Concentration (ppb)	18.8±1.0	0.7±0.1	12.6±0.4	0.73	3.3
Hg ²⁺ Concentration (ppb)	0.5±0.0	17.8±0	0.46±0.0	16.2	29.4
Total Hg Concentration (ppb)	19.3±1	18.4±0.2	13.1±0.4	16.9	32.7
% Hg ²⁺	2.6	96.7	3.5	95.9	89.9

The speciation results shown in Table 3 suggest that HCl has a significant effect on the conversion of Hg⁰ into Hg²⁺ under SCR NO_x emission control conditions. All tests except P2 showed most of the mercury sampled at the outlet of the SCR catalyst was in the Hg²⁺ form. As shown in Table 1, the gas mixtures used for Test P1 and P2 were identical except that 8 ppm of HCl was added to the simulated flue gas mixture for the P1 Test, and no HCl was added in the P2 Test. Almost all the mercury measured in the P1 Test was Hg²⁺, but very little Hg²⁺ was

measured in the P2 Test. The total mercury concentration (13.1 ppb) at the outlet of the SCR catalyst measured for the P2 Test is about 60% of that (19.3 ppb) measured at the inlet. The results of the two outlet replicate tests are similar to each other, indicating good precision of the tests. One possible explanation for lower mercury being measured at the outlet for the P2 Test is that the adsorption of Hg^0 by the SCR catalyst may take place in the absence of HCl in the flue gas mixture. Evidence of Hg^0 adsorption was observed during the initial scoping tests of the SCR reactor system. When a gas mixture with 3% O_2 , 10% CO_2 , 240 ppm NO, and 30 ppb Hg^0 (balance N_2) was passed through the SCR catalyst at 350 °C, very little Hg^0 was measured at the outlet of catalyst by using an on-line ultraviolet (UV) mercury analyzer. Hg^0 was measured at detectable levels (>2 ppb) at the outlet immediately after NH_3 was added to this gas mixture. The effect of flue gas species on the adsorption of Hg^0 by the SCR catalyst requires further investigation.

Two tests were conducted for simulating the combustion of bituminous coals that have S and Cl contents much different from those of the PRB coals. The B1 test simulated the combustion of a low-sulfur and high-chlorine bituminous coal, and the combustion of a high-sulfur and low-chlorine coal was simulated by the B2 test. Results presented in Table 3 show Hg^{2+} was the predominant mercury species measured at the outlet of the catalyst for both tests. It appears that the relatively high HCl concentrations present in the simulated flue gases used for these two tests provide adequate chlorine for converting Hg^0 to Hg^{2+} . The total Hg concentration (16.9 ppb) measured at the outlet of the catalyst for Test B1 is comparable to that (19.3 ppb) measured at the inlet. However, much higher outlet total Hg concentration (32.7 ppb) was measured for Test B2. The reason for such high total outlet Hg concentration is not known at the present time. A sudden rapid increase in Hg^0 (spike) produced by the Hg^0 generation unit may be the underlying reason. The Hg^0 spike had been observed by using the on-line mercury analyzer during the initial scoping tests. Test B2 is planned to be replicated in the near future.

Importance of Chlorine on Hg^0 Oxidation

It appears that HCl is the most critical flue gas component for converting Hg^0 to Hg^{2+} under SCR emission control conditions. Hg^{2+} was measured as the predominant species at the outlet of the SCR catalyst for the three tests (P1, B1, and B2) that had widely different HCl and SO_2 concentrations. The HCl concentration (8 ppm) used for test P1 was much lower than those used for the other two simulated bituminous coal combustion tests. However, it is still much higher than that (19 ppb) of the Hg^0 in the flue gas and appears to be adequate for converting most of the Hg^0 to Hg^{2+} . Hg^0 was found to be the predominant species for the only test (P2) that had no HCl present in the flue gas. The observation of no Hg^0 oxidation for this test may be due to the lack of a chlorine source in this test. The importance of HCl in the flue gas for oxidation of Hg^0 under SCR emission control conditions observed in the present study seems to be consistent with a recent field study. Significant increases in Hg^{2+} across the SCR reactor were observed for boilers burning bituminous coals with total chloride concentration measured in the flue gases ranging from 108 to 19 ppm.⁶ However, a boiler burning a PRB coal with 1.5 ppm total chloride measured in the flue gas showed very little change in mercury speciation across the SCR reactor.⁶ The PRB coal tested has a Cl content below the detection limit (< 60 $\mu\text{g/g}$), and high Ca content (25%) was found in the fly ash. The lack of HCl available for Hg^0 oxidation for this coal with low Cl content and high Ca content may be the main reason for very little Hg^0 oxidation

observed across the SCR reactor.

The importance of a chlorine source in low temperature, heterogeneous oxidation of Hg^0 catalyzed by metal oxides present in fly ash has been suggested in a recent modeling study.¹¹ The study suggested that Hg^0 oxidation may be promoted by a Deacon-type process for producing Cl_2 from HCl which further reacts with Hg^0 leading to Hg^0 oxidation. The SCR catalyst used in the present study contains V_2O_5 and TiO_2 , which may be reactive for promoting the Deacon-type process. Such oxides have been shown to promote the transformation of Hg^0 to Hg^{2+} in combustion flue gas.⁷ Another recent modeling study suggested that low-temperature oxidation of Hg^0 by a reactive source of chlorine is significant for coals with appreciable chlorine content.¹² The large Cl storage capacity of unburned carbon in coal fly ash is believed to provide a source of reactive Cl for Hg^0 oxidation over a broad temperature range below that for the homogeneous reaction.¹² The SCR catalysts also have significant surface areas as well as active sites, which could retain the reactive Cl generated at the catalyst surfaces and serve as a reactive Cl source for reactions with Hg^0 to oxidize the Hg^0 .

SUMMARY

A bench-scale reactor system has been constructed in the present study for closely simulating the SCR emission control conditions in the field. The system was tested using different gas mixtures simulating the combustion of bituminous and subbituminous coals with different Cl and S contents. NO_x reduction levels (~85%) similar to those observed in field units were achieved by the system under the different simulated coal combustion conditions tested in this study. Almost complete oxidation of Hg^0 to Hg^{2+} was observed in all but one of the four simulated coal combustion tests. In the fourth test, no Hg oxidation was observed when HCl was absent from the simulated flue gas. This result strongly suggests that HCl is important for providing the source of Cl for Hg^0 oxidation under SCR emission control conditions. The importance of HCl for Hg^0 oxidation found in the present study provides a strong scientific base for the apparent coal-type dependence observed for Hg^0 oxidation reported across SCR reactors in prior field studies.

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KEY WORDS

Mercury speciation, Selective catalytic reduction, Coal combustion, Elemental mercury, oxidized mercury, Chlorine content

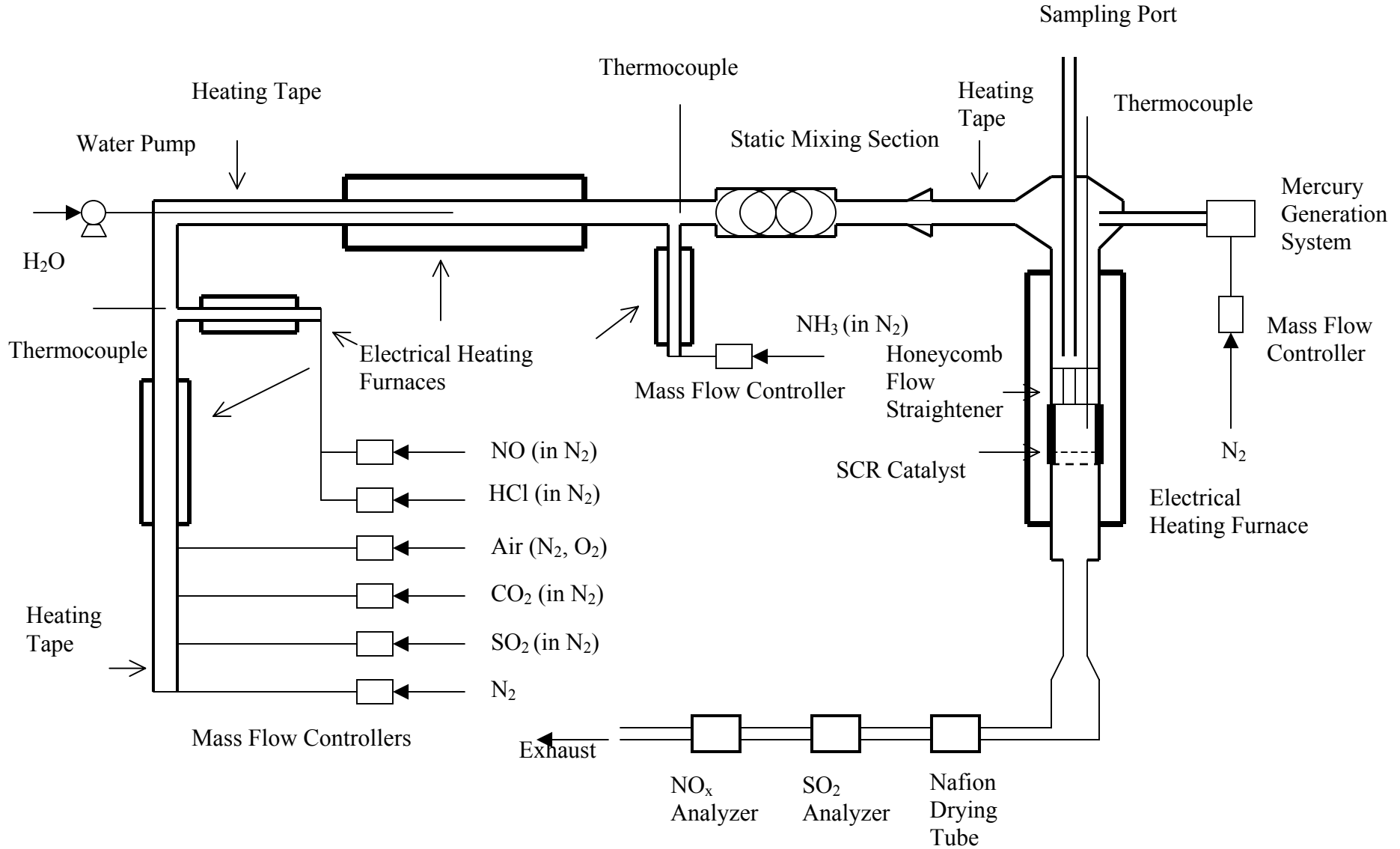


Figure 1. Schematic of SCR reactor system

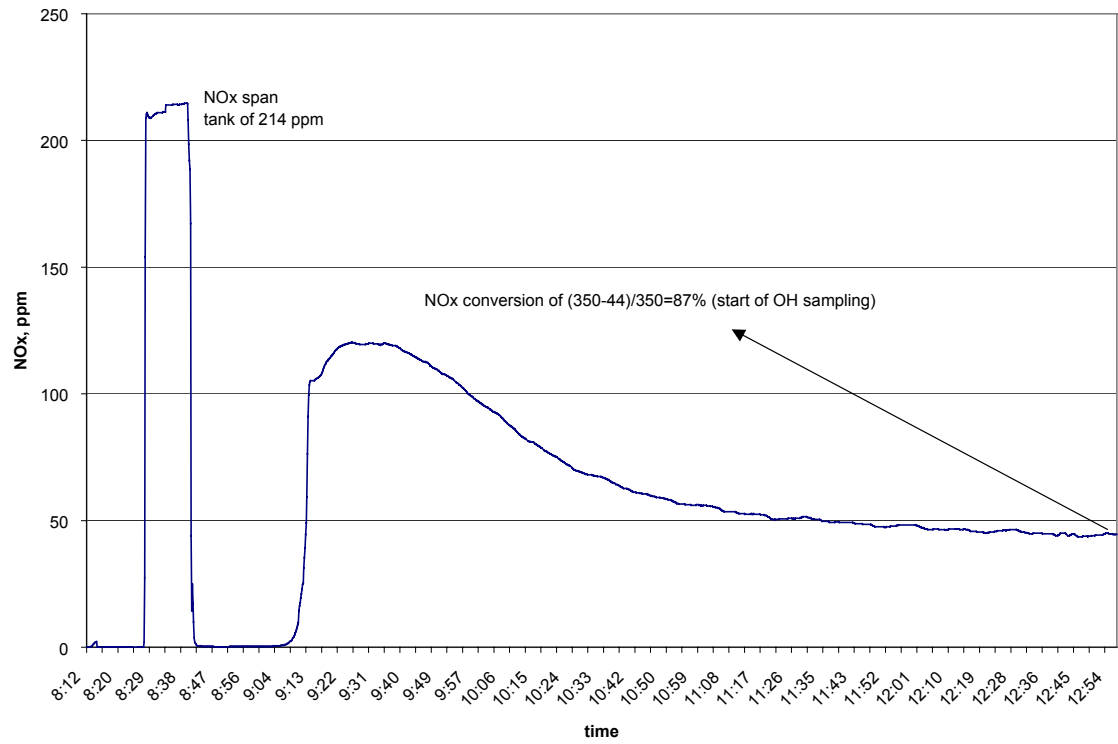


Figure 2. NO_x concentration vs. time (hours:minutes) measured at SCR reactor outlet for Test B1