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**Project Title: EVALUATION OF SCR CATALYSTS FOR COMBINED
CONTROL OF NO_x AND MERCURY**

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ABSTRACT

Selective catalytic reduction (SCR) catalysts, in addition to reduction of nitrogen oxides (NO_x), can oxidize elemental mercury (Hg⁰) in flue gases of coal-fired boilers. Therefore, Hg⁰ oxidation by SCR catalysts upstream of flue gas desulfurization (FGD) systems should create an opportunity for simultaneous and cost-effective control of mercury, NO_x, and sulfur dioxide (SO₂) emissions at power plants. This is because, unlike Hg⁰, oxidized mercury (Hg²⁺) is highly soluble in aqueous solutions and, therefore, can be easily removed in FGD systems.

A two-task, bench- and pilot-scale research study was conducted to investigate the effect of SCR catalysts on mercury speciation in Illinois and Powder River Basin (PRB) coal combustion flue gases. Task I studied Hg⁰ oxidation by a SCR catalyst (provided by Cormetech, Inc.) using a bench-scale reactor. Illinois and PRB coal flue gases were simulated in the bench-scale system. Very good oxidation of Hg⁰ was observed in all the coal flue gases (~ 90%). It was shown that hydrogen chloride (HCl) may be the critical flue gas component that causes the conversion of Hg⁰ to Hg²⁺ under SCR reaction conditions. Since high HCl concentrations are expected during combustion of all Illinois coals, firing of these coals should result in significant Hg⁰ oxidation occurring across the SCR reactors in the field.

Based on bench-scale results, an appropriate SCR catalyst was produced in a pilot-scale size (by Cormetech, Inc.) and installed in a pilot-scale combustor for Task II studies. Three different Illinois coals (from high to low sulfur and chlorine) and one PRB coal were combusted in the pilot-scale unit. For the high sulfur/low chlorine Illinois coals, about 84–68 % Hg⁰, and 15–9% Hg²⁺ were measured at the inlet of the pilot-scale SCR reactor. The SCR catalyst induced high oxidation of Hg⁰, decreasing the percentage of Hg⁰ at the outlet of the SCR to values below 16%. The low sulfur/high chlorine Illinois coal had a relative high amount (21%) of Hg²⁺ at the inlet of SCR. The Hg⁰ content of this coal flue gas was decreased from about 73% at the inlet to about 4% at the outlet of the SCR reactor. The PRB coal tests indicated a low oxidation of Hg⁰ by the SCR catalyst. The percentage of Hg⁰ decreased from about 96% at the inlet of the reactor to about 80% at the outlet.

EXECUTIVE SUMMARY

The Air Pollution Prevention and Control Division (APPCD) of the United States Environmental Protection Agency (EPA), ARCADIS, Inc., and Cormetech, Inc. conducted an Illinois Clean Coal Institute (ICCI) co-funded research to evaluate the viability of a mercury control technology utilizing oxidation of elemental mercury (Hg^0) to ionic mercury (Hg^{2+}) for subsequent removal by conventional flue gas desulfurization (FGD) systems. Mercury speciation modification was implemented via selective catalytic reduction (SCR) catalysts, currently used for nitrogen oxides (NO_x) control. In December of 2001, the EPA determined that control of mercury emissions from coal-fired power plants is appropriate and necessary. For Illinois coal-fired utilities, control of mercury emissions may be needed to comply with future regulations. Oxidation of Hg^0 to Hg^{2+} prior to an FGD system is a viable option for controlling mercury emissions, as FGD systems are known to remove Hg^{2+} at high efficiencies. This research generated valuable data, which may be used by utilities using Illinois coals to cost-effectively control mercury, NO_x , and sulfur dioxide (SO_2) emissions using SCR/FGD systems.

Currently there are no commercial technologies for modifying mercury speciation prior to an FGD unit. This study evaluated the viability of SCR technology for mercury speciation modification in Illinois and also Powder River Basin (PRB) coal combustion processes. Since SCR and FGD are increasingly being used at power plants to control NO_x and SO_2 emissions, respectively, their combination has the potential to provide effective mercury control.

The objectives of the research were to conduct bench- and pilot-scale studies in order to demonstrate Illinois and PRB coal combustion conditions that favor conversion of Hg^0 to Hg^{2+} by a Cormetech, Inc. formulated SCR catalyst. These objectives were accomplished, initially, through a systematic bench-scale study of the effect of the simulated flue gas compositions (three different Illinois and one PRB coals) on the oxidation of Hg^0 to Hg^{2+} in the presence of the SCR catalyst. Hg^0 oxidation activity of the SCR catalyst was established during the bench-scale testing phase of this study. A pilot-scale version of the SCR catalyst, designed and manufactured by Cormetech, Inc., was installed in a pilot-scale combustor to evaluate and establish Hg^0 oxidation capabilities of this catalyst during combustion of three different Illinois coals (from high to low sulfur/chlorine) and one PRB coal (very low sulfur and chlorine).

The research was performed over a 12-month period (from September 2002 to August 2003) and consisted of two main tasks. Task I involved bench-scale testing of a SCR catalyst formulation (designed by Cormetech, Inc.) to characterize oxidation of Hg^0 to Hg^{2+} in the presence of coal combustion/SCR flue gas species such as NO_x , ammonia (NH_3), SO_2 , and hydrogen chloride (HCl). The ranges of NO_x , NH_3 , HCl , SO_2 , and water (H_2O) concentration employed during Task I were those expected to be found in three representative Illinois coals and one PRB coal combustion flue gases. The Illinois coals tested include Galatia [low sulfur (1.13 wt%) and high chlorine (0.29 wt%)], Turriss [medium to high sulfur (3.11 wt%) and low chlorine (0.17 wt%)], and Crown II [high sulfur (3.48 wt%) and low chlorine (0.13 wt%)]. The PRB coal tested (from Black

Thunder) is a Southern PRB coal with very low sulfur (0.29 wt%) and chlorine content (about 0.01 wt%). Task I results showed that HCl is the 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 in the four simulated coal tests (PRB, Galatia, Turris, and Crown II). The HCl concentration (8 ppm) used for PRB test was much lower than those used for the Illinois coal combustion tests (100-200 ppm). However, it was still much higher than that of the doped Hg^0 (19 ppb) in the simulated flue gas, and may be adequate for converting most of the Hg^0 to Hg^{2+} . Hg^0 was found to be the predominant species at the exit of the SCR catalyst for only one test, PRB coal that had no HCl present in the flue gas. This is believed to be due to the lack of a chlorine source in this test. The final result of Task I was the confirmation of the SCR catalyst formulation to be used in Task II.

Task II was performed in two subtasks. In Subtask I, a SCR catalyst was installed in a vertical downward flow configuration in the post-combustion region of a pilot-scale 34.9 kW (150,000 Btu/h), refractory lined, down-fired cylindrical furnace. This furnace, termed the “innovative furnace reactor” (IFR) has the capability of firing natural gas or pulverized coal. In Subtask I, the natural gas combustion flue gas was doped with appropriate amounts of Hg^0 , HCl, NO_x , NH_3 , and SO_2 . It was determined that the catalyst contribution to the oxidation of Hg^0 was very low (only about 8% increase). It was hypothesized that the absence of particulates (fly ash) might have a bearing on Hg^0 oxidation reactions. Indeed, the coal combustion tests performed later indicated a very good Hg^0 oxidation by the SCR catalyst. The natural gas combustion test indicated that the IFR could be operated consistently and most of the injected mercury could be recovered at the outlet of the SCR catalyst. Furthermore, it established reliable operation of the pilot-scale combustion/SCR system.

In Subtask II of Task II, three different Illinois coals were combusted in the IFR facility. The objective of Subtask II was to characterize Hg^0 oxidation for a spectrum of Illinois coals and to establish any additional increases of Hg^{2+} resulting from the SCR catalyst used in Task I and Subtask I of Task II. Since combustion of PRB coals will result in the production of Hg^0 -dominated flue gases, the effect of the pilot-scale SCR catalyst on oxidation of Hg^0 in a typical PRB coal combustion flue gas was also studied. Mercury speciation of the flue gas (Hg^0 , Hg^{2+} , and Hg associated with particulate matter, Hg_p) across the SCR catalyst was measured using the Ontario Hydro (OH) method. Attempts were made to measure gas-phase Hg (Hg^0 and Hg^{2+}) by using a specially designed dual isokinetic/Inertial Gas Sampling (IGS) probe. The Hg speciation samplings were performed at the inlet of the baghouse where high particulate (fly ash) loading on the filter of the isokinetic OH sampling train may change the speciation of mercury. Deposition of white powder was observed occurring on the walls of the first impinger connected to the IGS probe. Comparison of the data measured by using the OH method and those using the IGS probe showed the total Hg in solid and gaseous phases measured by the OH method was significantly higher than the gas-phase Hg. The white ammonium salt aerosols were formed near the inlet of the first impinger connected to the IGS probe as the ammonia-containing flue gas cooled down. The aerosols may retain gas-phase Hg and change in Hg speciation may result. Further work is needed for the development of

the IGS probe for sampling ammonia-containing flue gas in SCR systems. NO_x conversions by the Cormetech, Inc.-formulated SCR catalyst for all these coals were around 90%.

It was found that Hg⁰ was the predominant species in the flue gases measured at the inlet of SCR catalyst for the three Illinois coals tested. The combustion of the low sulfur/high chlorine Galatia coal produced a relative high amount (21%) of Hg²⁺ at the inlet of SCR. The Hg⁰ content of this coal flue gas was decreased from about 73% at the inlet to about 4% at the outlet of SCR reactor. Turris and Crown II coal combustion tests exhibited similar behavior with regard to mercury speciation and oxidation. These two coals are very similar in their properties. During the combustion of these coals about 84 – 68 % Hg⁰, and 15 - 9% Hg²⁺ were measured at the inlet of the pilot-scale SCR reactor. The SCR catalyst induced high oxidation of Hg⁰, dropping the percentage of Hg⁰ at the outlet of the SCR to values below 16%. These results suggest that the use of SCR systems in Illinois coal-fired boilers that are equipped with FGD systems may result in about 85 to greater than 90% control of mercury.

The PRB coal combustion tests showed that almost all the Hg measured at the inlet was Hg⁰. The test results also indicated a very low oxidation of Hg⁰ by the SCR catalyst. Based on the measurements conducted in this study, it appears that SCR applications on PRB coal-fired boilers may not result in significant increase in Hg²⁺ content of flue gas. Therefore, such boilers equipped with wet FGD systems may not achieve increased mercury removal resulting from SCR applications.

OBJECTIVES

The objective of this research program was to investigate elemental mercury (Hg^0) oxidation across selective catalytic reduction (SCR) systems in Illinois coal combustion processes. Additional tests were also performed to characterize SCR catalyst activity in a Powder River basin (PRB) coal combustion environment. The specific goals of the proposed research were to identify the effect of SCR catalysts, flue gas compositions, and combustion conditions (time-temperature history and concentrations of gas-phase species) on the oxidation of Hg^0 upstream of the flue gas desulfurization (FGD) units. This work was synergized with similar collaborative bench-scale research being conducted by U.S.EPA and Cormetech, Inc.

This research was performed over a 12-month period and consisted of two tasks. Task I involved bench-scale testing of a promising SCR catalyst formulation to characterize oxidation of Hg^0 to Hg^{2+} in the presence of coal combustion flue gas species such as nitrogen oxides (NO_x), ammonia (NH_3), sulfur dioxide (SO_2), and hydrogen chloride (HCl). Previous EPA investigations with synthetic and actual fly ashes have indicated that certain transition metals are very active in catalyzing conversion of Hg^0 to Hg^{2+} in the presence of HCl and NO_x . SCR catalysts contain vanadium (a transition metal) active sites. In Task I, the role of this transition metal on Hg^0 oxidation was identified. The NO_x , NH_3 , HCl, SO_2 , and H_2O concentration ranges employed during Task I were those expected to be found in three representative Illinois coals (low to high sulfur and chlorine) and one PRB coal combustion flue gases. The final result of Task I was the confirmation of the SCR catalyst formulation to be used in Task II.

Task II was performed in two subtasks. In Subtask I, a pilot-scale configuration of the active SCR catalyst was installed in a vertical downward flow configuration in a pilot-scale 34.9 kW (150,000 Btu/h), refractory lined, down-fired cylindrical furnace. This furnace, termed the IFR, has the capability of firing natural gas or pulverized coal. In Subtask I, natural gas combustion flue gas was doped with appropriate amounts of Hg^0 , HCl, NO_x , NH_3 , and SO_2 . Mercury speciation of the flue gas (Hg^0 and HgCl_2 content) across the SCR catalyst was measured using the Ontario Hydro (OH) method. Once the reliable operation of the pilot-scale combustion system was established, in Subtask II of Task II three different Illinois coals were combusted. The objective of Subtask II was to characterize Hg^0 oxidation for a spectrum of Illinois coals and to establish any additional increases of Hg^{2+} using the SCR catalyst characterized in Task I and Subtask I of Task II. The effect of the active pilot-scale SCR catalyst on oxidation of Hg^0 in a typical PRB coal combustion flue gas was also studied.

INTRODUCTION AND BACKGROUND

Metallic air toxics, such as mercury, are found in coal combustion flue gases in elemental or various oxidized forms and exist in the solid, aerosol, or vapor state. These compounds originate from the raw coal and are typically enriched in the fine particles (Markowski and Filby, 1985). Mercury speciation may be potentially modified by SCR

catalysts currently used for NO_x control. In December of 2001, the EPA determined that control of mercury emissions from coal-fired power plants is appropriate and necessary. For Illinois-coal-fired utility boilers, control of mercury emissions may be needed to comply with future regulations. Oxidation of Hg⁰ to Hg²⁺ prior to an FGD system is a viable option for controlling mercury emissions, as FGD systems easily remove Hg²⁺. Research data is needed to assess the contribution of SCR catalysts to the oxidation of Hg⁰ prior to the FGD systems. Preliminary results in the EPA laboratories (Ghorishi and Gullett, 1998) have indicated the effectiveness of alkaline sorbents (dry FGD units) in removing Hg²⁺ (but not Hg⁰) from flue gases. Previous investigations have also indicated that many flue gas and fly ash parameters determine the speciation of mercury (Ghorishi, 1998; Ghorishi et al., 1999).

The distribution of various forms of mercury in flue gases is a function of system-specific properties. For example, various concentrations of chlorine or sulfur have been shown to significantly affect the expected equilibrium product distribution (Linak and Wendt, 1993). Ghorishi et al. (1999) have shown that, in combustion flue gas with the presence of active simulated or actual fly ashes, Hg⁰ is readily oxidized by HCl and NO_x at temperatures typical of air pollution control systems (150-250° C). The presence of these constituents in the flue gas can therefore shift the ratio of Hg²⁺/Hg⁰ to a higher value and result in better removal of mercury in FGD systems. However, quantitative information on speciation of mercury in the SCR systems in the presence of coal combustion flue gases at typical SCR operating temperatures (340-370° C) is lacking at this time.

The control of mercury emissions has been found to be strongly dependent on the form of mercury; these results were observed at the Electric Power Research Institute's High Sulfur Test Center's (HSTC) 4 MW_e pilot-scale wet FGD system and at municipal waste combustor (MWC) tests (Volland, 1991). The presence of HCl and NO_x in the combustion flue gases shifts the mercury speciation toward its oxidized form (Hall et al., 1991, Ghorishi, 1998). The type of coal has also been found to affect different levels of mercury capture (Felsvang et al., 1992; 1993). The chlorine present in coal, which is capable of forming the less volatile and more water-soluble Hg²⁺, is thought to be an important factor with regard to the level of mercury capture in coal combustion (Felsvang et al., 1993; DeVito et al., 1993). Therefore, an important scientific issue that is being investigated is the speciation of mercury in coal combustion flue gases. Knowledge on the transformation of Hg⁰ to HgCl₂ is absolutely crucial in this regard. A detailed review on the state of knowledge on mercury speciation in coal-fired processes has been performed by Senior et al. (1997). Their review and another investigation (Krishnan et al. 1994) concluded that the assumption of gas-phase equilibrium for mercury species in coal-fired flue gases is not valid and major reaction pathways for mercury oxidation in coal combustion flue gas need to be investigated. These investigations should include heterogeneous reactions on active surfaces as well as gas-phase reactions (Senior et al. 1997).

A preliminary investigation was performed at EPA laboratories (Ghorishi, 1998) to provide some insights into these questions by studying mercury speciation in simulated coal combustion flue gases. Gas-phase studies indicated that the in-flight, post-

combustion oxidation of Hg^0 in the presence of HCl is very slow and proceeds at measurable rates only at high temperatures ($>700^\circ\text{C}$) and high HCl concentrations (200 ppm). The presence of SO_2 and H_2O in the simulated flue gas significantly inhibited the gas-phase oxidation of Hg^0 in the presence of HCl. These results indicate that conversion of Hg^0 to Hg^{2+} needs to be mediated by the presence of an active surface such as fly ash (or SCR catalyst, as in this study). The effects of coal fly ash components and compositions were investigated using a fixed-bed of model (simulated) fly ashes. The primary focus was on evaluating the catalytic Hg^0 oxidation activity of major mineral constituents of coal fly ashes: alumina (Al_2O_3), silica (SiO_2), iron (III) oxide (Fe_2O_3), copper (II) oxide (CuO), and calcium oxide (CaO). Copper and iron oxides exhibited significant catalytic activity toward a surface-mediated oxidation of Hg^0 .

SCR has been used extensively to control NO_x emissions from hundreds of utility and industrial boilers in Japan, Europe, and the United States. A large number of SCR systems are under construction or planned to be installed in the United States. A recent study (Cichanowicz and Muzio, 2001) estimated that, by the year 2004, in excess of 100 GW of coal-fired capacity in the United States will be equipped with SCR to meet the requirements of Title I of the clean Air Act (CAA). The study describes how today's U.S. applications of SCR have evolved from two prior generations of design in response to changing application conditions. The first commercial SCR application in Japan occurred in the late 1970s, and a subsequent "wave" of installations took place in Europe (predominantly Germany) in the mid-1980s. As a result of this evolution, present-day SCR catalysts feature thinner walls, improved mass transfer and activity, and better poison resistance than earlier generations of SCR catalysts. Limited field data obtained from European coal-fired boilers equipped with SCR systems suggest that SCR catalysts promote the formation of Hg^{2+} (Gutberlet et al., 2000). Results of recent pilot-scale screening tests on the impact of SCR on mercury speciation also indicate that SCR systems could promote the formation of ionic Hg (EPRI report, 2000). The screening test results reflected that the impact of SCR on mercury speciation appears to be highly coal type dependant. For the coals (three bituminous coals and one subbituminous PRB coal) tested, only the high-sulfur bituminous coal showed significant increase in oxidized mercury in the outlet of the SCR. The low-sulfur (also low chloride) PRB coal showed very little impact of SCR on mercury speciation. The other two bituminous coals' results were between the two extreme cases. Results of the recent field tests conducted in the United States showed there was an increase in Hg oxidation across the SCR catalysts for plants firing eastern bituminous coals with sulfur contents ranging from 1.1 to 4.2% (Laudal et al., 2003).

The increase in oxidation of Hg^0 to ionic Hg across the SCR reactor could result from the changes in flue gas chemistry caused by the SCR catalysts. The SCR catalysts including oxides of vanadium, titanium, and tungsten, have the potential to catalyze the formation of SO_3 (from SO_2) and Cl_2 (from HCl) in coal combustion flue gases. Both SO_3 (Galbreath and Zygarlicke, 2000) and Cl_2 (Ghorishi et al., 1999) have been suggested to react with Hg^0 to cause its oxidation. However, not much is known on the effect of the flue gas species, chemical reactions, and reaction conditions on Hg^0 oxidation across SCR reactors.

EXPERIMENTAL PROCEDURES

Task I studies were performed in a bench-scale SCR/Hg⁰ oxidation reactor system. A schematic of the reactor system is shown in Figure 1. The system consists of preheating and premixing sections, a mercury generation unit, a SCR reactor, and an on-line reactor effluent measurement unit. Flue gas components including carbon dioxide (CO₂), SO₂, air, and nitrogen (N₂) were mixed and preheated to 350 °C and then mixed with another preheated stream of NO_x and HCl at the main heating section. Water, for simulating the moisture content in the flue gas, was also pumped into this section at a calibrated rate and mixed with the other flue gas components. Ammonia was preheated and added into a section equipped with static mixing hardware. The gas mixture flowed into a Pyrex reactor. The alkaline NH₃ reacts with the acid gas components in the gas mixture to form ammonium salts at temperatures lower than the SCR reaction temperatures, so the simulated flue gas mixture was preheated by electrical furnaces and maintained at 350° C by temperature-controlled electrical heating tapes. A mercury generation unit consisting of a mercury permeation tube surrounded by a temperature-controlled water bath was used to generate Hg⁰ vapor for the oxidation experiments. The Hg⁰ vapor was carried by an N₂ stream and mixed with flue gas mixture near the top of the reactor (4 cm in inner diameter and 35 cm in length). The flue gas mixture containing Hg⁰ and NH₃ passed through a honeycomb flow straightener to produce a uniform laminar flow before passing through the honeycomb SCR catalyst. The effluents exiting the SCR reactor passed through a drying tube for removing moisture in the flue gas. The dried gas then passed to an online ultraviolet SO₂ analyzer (Bovar Engineering, Inc., model 721AT2; accuracy of about ±5%) for measuring SO₂. An online chemiluminescent NO_x analyzer (Advanced Pollution Instrumentation, Inc., model 200AH; accuracy of about ±5%) downstream of the SO₂ analyzer was used for measuring NO_x.

A SCR catalyst with a vanadia/titania formulation and a honeycomb configuration, designated as Catalyst A, was used in Task I. A small piece (2.2 cm for both sides and 1.9 cm in length; calculated space velocity of 2609 hr⁻¹) of the catalyst sample was placed into the catalyst compartment of the SCR reactor. The flows of the flue gas components were maintained at the desired levels by using mass flow controllers. The variability of the gas concentrations is about ±2.5%. A constant total flow rate of 400 cm³/min (at a standard temperature of 25 °C and pressure of 101.4 kPa, standard temperature and pressure [STP]) was used for all tests. The concentrations of SO₂ and NO_x at the outlet of the reactor were monitored continuously for 4 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 it was connected to a mercury sampling train. The mercury speciation measurement method known as the OH method was used for measuring Hg⁰ and Hg²⁺ in this study.

In Task II of this study, tests were performed in a pilot-scale 34.9 kW (150,000 Btu/h), refractory lined, down-fired cylindrical furnace fired with coal or natural gas (Figure 2). The furnace, termed the IFR, has an inner diameter of 15.2 cm and overall length of about 4 m. The IFR is used to simulate and generate a coal combustion environment and quench rate conditions similar to those upstream of FGD units in coal-fired utility boilers.

View and injection/probe points traverse the length of the furnace for testing flexibility. The IFR system was retrofitted with two vertical sections, an upward and a downward flow. The objective was to install the two-stage, pilot-scale SCR reactor, based on the bench-scale results, in the downward section. Figure 2 illustrates the dimensions of this two-stage SCR system. A sampling port is installed between the two stages. The first stage of the SCR catalyst is equipped with a flow straightener, a soot blower, and a honeycomb catalyst. The second stage has a soot blower and a honeycomb catalyst. The two honeycomb catalysts are identical and have a pitch of 8.2 mm, wall thickness of 0.83 mm, and length of 1250 mm; they consist of 8x8 cells; a total of 64 cells. The calculated space velocity of the two stages when the IGS probe is operational was 2943 hr^{-1} . This space velocity is comparable to that used in bench-scale tests (2609 hr^{-1}) and is also comparable to those SCR space velocities observed in the field. Sampling ports along the horizontal ducts at the inlet and the outlet of the vertical SCR reactor allow gas and particle monitoring. The IFR is equipped with a continuous emission monitoring system for O_2 , CO_2 , CO , NO_x , and SO_2 . The IFR is also equipped with 12 thermocouples in order to accurately determine the time-temperature history of the combustor.

Isokinetic OH trains were used for making mercury speciation measurements at the inlet and the outlet of the SCR reactor. Two inertial gas sampling (IGS) probes (Mott Co.) were also used in conjunction with the isokinetic OH trains at the inlet and the outlet of the SCR reactor for Hg measurements. The IGS probe has the capability of separating gas from particulate matter without the need for a filter, thus, minimizing the potential Hg speciation biases created by the filter of the isokinetic OH train. The IGS probes were equipped with venturi flow meters for accurate axial flow measurement. The focus of the isokinetic trains for mercury sampling was only on total mercury data (Hg_T), since these samplings were performed at the inlet of the baghouse with significant particulate loading in the flue gas. The IGS probe data are only for the gas-phase elemental (Hg^0) and ionic mercury (Hg^{2+}). The difference between Hg_T results obtained by the isokinetic OH train and the gas-phase Hg^0 and Hg^{2+} obtained by the IGS probe is the amount of Hg associated with particulate matter (Hg_p). The IFR is also equipped with a baghouse and a scrubber to treat the flue gas before releasing it into the atmosphere.

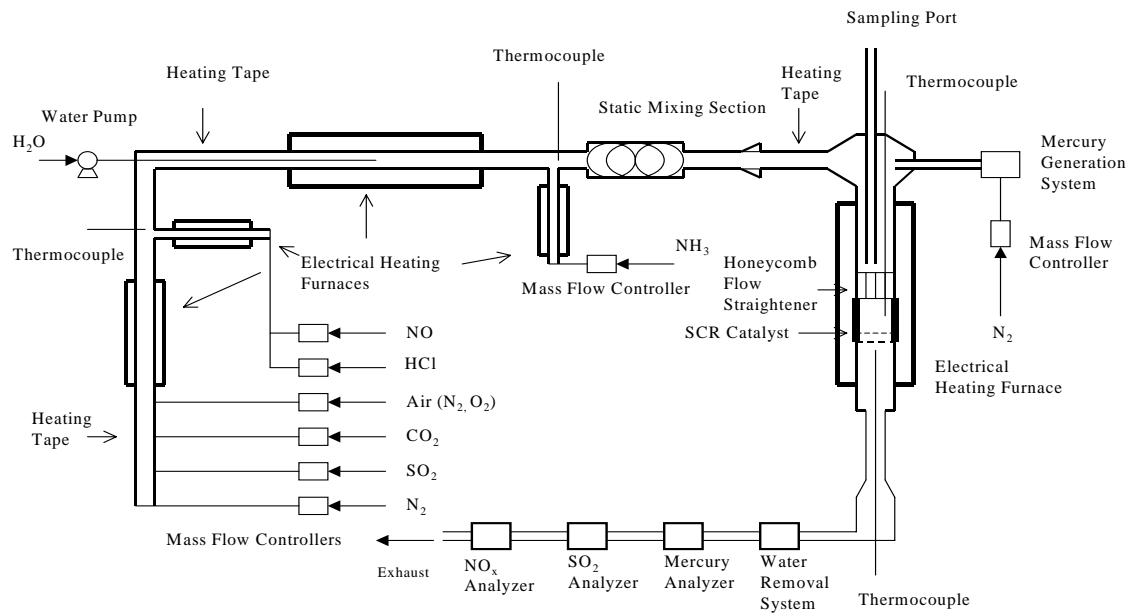


Figure 1: Schematic of bench-scale SCR/Hg⁰ oxidation reactor system

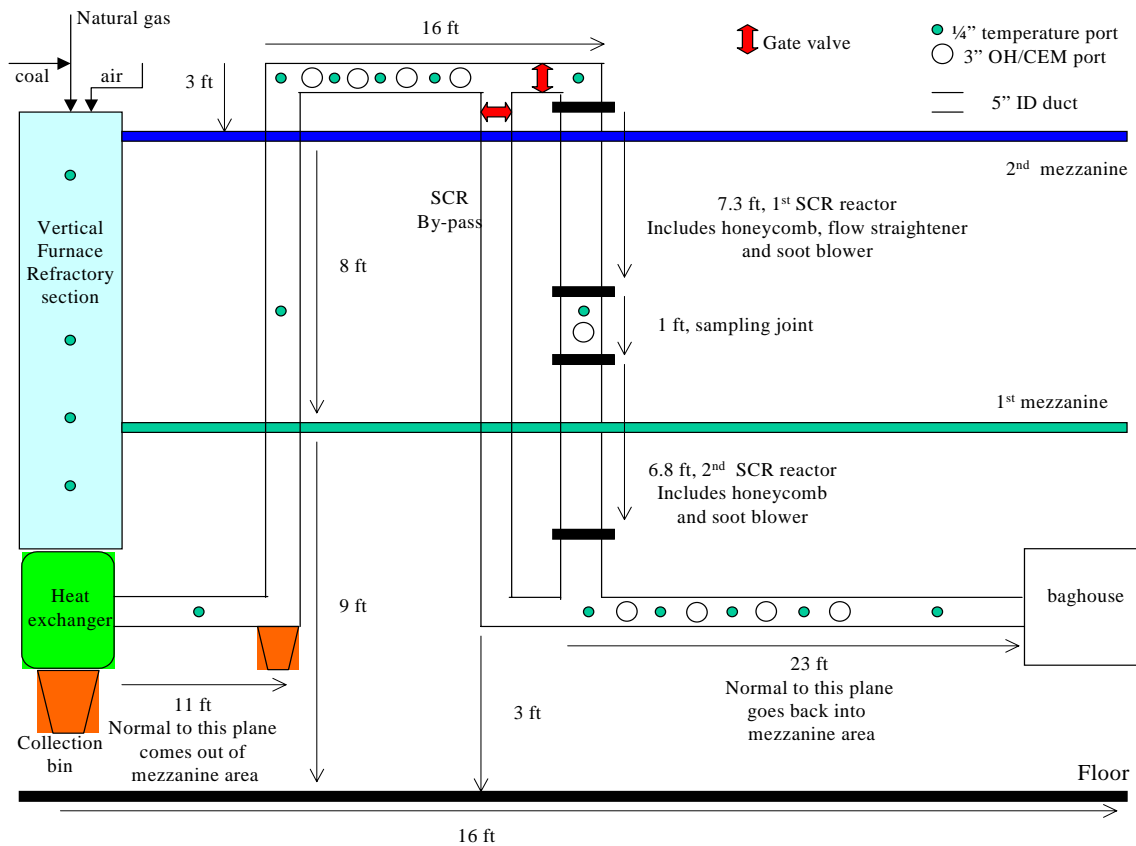


Figure 2: Schematic of the pilot-scale combustor used to evaluate the SCR catalyst

RESULTS AND DISCUSSION

Task I: Bench-Scale Evaluation of SCR Catalyst for Hg⁰ Oxidation in Simulated Illinois and PRB Coal Combustion Flue Gases

Bench-scale testing of the SCR catalyst was performed in the simulated combustion flue gases of three different Illinois coals and one PRB coal. These coals were also tested in the pilot-scale combustor during Task II studies. Table 1 summarizes the characteristic of the obtained Illinois coals (Turriss, Crown II, and Galatia) and the PRB coal (Black Thunder). Turriss coal sample contains medium amounts of sulfur and chlorine. Crown II is a high sulfur/low chlorine coal sample. Galatia contains high amounts of chlorine and low amounts of sulfur.

Table 1: Proximate and ultimate analysis of three Illinois Basin coals and a PRB coal

| Content | PRB Black Thunder | Turriss (Medium S/Cl) | Crown II (High S/low Cl) | Galatia (Low S/high Cl) |
|-------------|----------------------|--------------------------|-----------------------------|----------------------------|
| % Moisture | 14.00 | 16.99 | 16.07 | 11.33 |
| % Ash | 5.92 | 9.26 | 7.34 | 6.29 |
| % Volatile | 37.33 | 33.89 | 37.05 | 34.16 |
| % Fixed C | 42.76 | 39.85 | 39.55 | 48.22 |
| HV (Btu/lb) | 9903 | 10531 | 10877 | 12179 |
| %C | 59.71 | 59.00 | 60.48 | 68.31 |
| %H | 3.83 | 4.32 | 4.70 | 4.50 |
| %N | 0.82 | 1.19 | 1.07 | 1.50 |
| %S | 0.29 | 3.11 | 3.48 | 1.13 |
| %O | 15.44 | 5.96 | 5.73 | 6.94 |
| %Cl | NA | 0.17 | 0.13 | 0.29 |
| Hg, ppmw | NA | 0.07 | 0.07 | 0.09 |

NA: Not Available

The ultimate analysis results of these coals were used to determine the composition of the simulated flue gas in each test case shown in Table 2. The base flue gas mixture consisted of 350 ppm NO_x, 315 ppm of NH₃, 15% CO₂, 3.5% of O₂, 5.3% of H₂O, 19 ppb of Hg⁰, and balance in N₂. The nominal sub-stoichiometric NH₃/NO_x ratio of 315/350 was used in the all tests in order to achieve a nominal 90% NO_x reduction. The PRB/no HCl test represents a situation where the Cl released from the combustion of PRB coal reacts with Ca resulting in no HCl in the flue gas. This simulation was designed to determine the effect of HCl on the oxidation of Hg⁰.

Table 2: Summary of simulated flue gas compositions for Task I studies

| Constituent Concentration | PRB | PRB/no HCl | Galatia | Turris | Crown II |
|---------------------------|-----|------------|---------|--------|----------|
| HCl (ppm) | 8 | 0 | 204 | 134 | 98 |
| SO ₂ (ppm) | 280 | 280 | 934 | 2891 | 3116 |
| NO _x (ppm) | 350 | 350 | 350 | 350 | 350 |
| NH ₃ (ppm) | 315 | 315 | 315 | 315 | 315 |
| CO ₂ (%) | 15 | 15 | 15 | 15 | 15 |
| O ₂ (%) | 3.5 | 3.5 | 3.5 | 3.5 | 3.5 |
| H ₂ O (%) | 5.3 | 5.3 | 5.3 | 5.3 | 5.3 |
| Hg ⁰ (ppb) | 19 | 19 | 19 | 19 | 19 |

Results of the NO_x reduction for these tests are summarized in Table 3. NO_x reductions of 85% to 88% were observed for all the tests. These levels of NO_x reduction are similar to those achieved in the field, suggesting that the reactor system used in the present study closely simulated the SCR NO_x emission control conditions in the field.

Table 3: Summary of NO_x reduction results for Task I studies

| NO _x Results | PRB | PRB/no HCl | Galatia | Turris | Crown II |
|--|-----|------------|---------|--------|----------|
| Outlet NO _x Concentration (ppm) | 44 | 52 | 44 | 43 | 41 |
| NO _x Reduction (%) | 87 | 85 | 87 | 88 | 88 |

The mercury speciation results are summarized in Table 4 (\pm indicates duplicate test). Two mercury speciation samples were taken at the inlet of the SCR catalyst by using the OH method after the PRB and Galatia tests. 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 very close to those (19.2 ± 0.1 ppb Hg_T 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 (PRB and Galatia tests) showed Hg⁰ was the only mercury species. The consistent inlet results suggest that the presences of HCl and SO₂ with different concentrations in the simulated flue gas mixtures do not cause gas-phase oxidation of Hg⁰ at 350° C prior to the SCR catalyst.

The speciation results shown in Table 4 suggest that HCl has a significant effect on conversion of Hg⁰ to Hg²⁺ under SCR/NO_x emission control conditions. All tests except

PRB/no HCl showed mostly Hg^{2+} at the outlet of the SCR catalyst. As shown in Table 2, the gas mixtures used for PRB and PRB/no HCl tests were identical except that 8 ppm of HCl was added to the simulated flue gas mixture for the PRB test. Almost all the mercury measured in the PRB test was Hg^{2+} , but very little Hg^{2+} was measured in the PRB/no HCl test. The total mercury concentration (13.1 ppb) at the outlet of the SCR catalyst measured for the PRB/no HCl 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. One possible explanation for the discrepancy in inlet and outlet results is that adsorption of Hg^0 by the SCR catalyst might have taken place in the absence of HCl in the flue gas mixture during the PRB/no HCl test. Evidence of Hg^0 adsorption was observed during the start-up of the SCR reactor system. When a gas mixture with 3% O_2 , 10% CO_2 , 240 ppm of NO , and 30 ppb of 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 ultraviolet (UV) mercury analyzer. However, this adsorption of Hg^0 by the SCR catalyst requires further investigation.

Table 4 also shows that Hg^{2+} was the predominant mercury species measured at the outlet of the catalyst for all the Illinois coal simulation tests. It appears that the relatively high HCl concentrations present in these simulated flue gases provide adequate chlorine for conversion of Hg^0 to Hg^{2+} . The total Hg concentrations (Hg_T) measured at the outlet of the catalyst for the Galatia and Crown II tests (16.9 and 20.0±0.3 ppb, respectively) are comparable to that measured at the inlet (19.3 ppb). However, much higher outlet total Hg concentration (32.7 ppb) was measured for the Turriss Test. It is possible that, during this test, a spike in the generation of Hg^0 might have occurred. However, this shows that the SCR catalyst was active enough to oxidize the relatively high spike of Hg^0 .

Table 4: Summary of mercury speciation results (Task I)
(results are in ppb; 1 ppb is 8.3 $\mu\text{g}/\text{dscm}$)

| Hg Specie Concentration | Inlet | PRB | PRB/no HCl | Galatia | Turriss | Crown II |
|-------------------------|----------|----------|------------|---------|---------|----------|
| Hg^0 | 18.8±1.0 | 0.7±0.1 | 12.6±0.4 | 0.73 | 3.3 | 2.0±0.1 |
| Hg^{2+} | 0.5±0.0 | 17.8±0 | 0.46±0.0 | 16.2 | 29.4 | 17.9±0.5 |
| Hg_T | 19.3±1 | 18.4±0.2 | 13.1±0.4 | 16.9 | 32.7 | 20.0±0.3 |
| % Hg^{2+} | 2.6% | 96.7% | 3.5% | 95.9% | 89.9% | 89.5% |

It appears that HCl is the 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 four simulated coal tests even though these coals had widely different HCl and SO_2 concentrations. The HCl concentration (8 ppm) used for the PRB test was much lower than those used for the Illinois 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+} in the SCR catalyst. Hg^0 was found to be the predominant species for the only test that had no HCl present in the flue gas. No Hg^0 oxidation observed for this test may be due to the lack of a chlorine source.

Task II: Pilot-scale Evaluation of SCR Catalyst for Hg⁰ Oxidation in the Combustion of Natural Gas, Illinois, and PRB Coal Combustion Flue Gases

Task II, Subtask I: SCR Catalyst Evaluation in Natural Gas Combustion

Natural gas tests were used as shakedown tests. They were also performed to determine the temperature profiles in the IFR and assess the feasibility of operating the pilot-scale SCR reactor at temperatures around 350° C. Another objective of Task II, Subtask I was to assess the change in Hg speciation across the SCR in a particulate-free environment. Natural gas tests were also used to determine whether mercury speciation in the IFR duct could be biased by use of the IGS probes.

Initial temperature profile measurements across the SCR catalyst indicated that, at the nominal IFR firing rate of 120,000 Btu/hr, a temperature of 350° C could be easily achieved in the first section of the SCR reactor. The axial temperature gradient along the SCR reactor was about 80° C and it was determined that the axial temperature gradient needed to be reduced. Accordingly, an additional layer of insulation was added to the SCR reactor wall, and all the flanges and exposed surfaces were insulated. The new axial temperature gradient was reduced to 50° C. This temperature gradient was deemed to be appropriate for the operation of the SCR reactor.

Ontario Hydro sampling for the natural gas tests was performed on 6/5/03. The new insulation was not in place during this test, and the axial temperature profile along the SCR reactor was 80° C. Natural gas was combusted at a firing rate of 118,674 Btu/hr. The excess air was calculated to be 20%. The O₂ concentration measured at the outlet of the SCR was 4.8% (predicted to be 3.8%), and the measured CO₂ concentration was 8.9% (predicted to be 9.6%). Comparison between the measured and the calculated values indicated a 7% in-leakage based on CO₂ and a 6% in-leakage based on O₂. The flue gas compositions measured at the inlet and outlet of the SCR reactor were identical, indicating no in-leakage between the inlet and the outlet of the SCR system. The total flow (including the in-leakage) was calculated to be 29.3 scfm (0.829 scm/min). NO_x and SO₂ were doped into the flue gas in the vertical section, near the 2nd temperature location, at 600-ppm levels. NH₃ and HCl were doped at 500- and 50-ppm levels (calculated), respectively. HCl was injected with SO₂ and NO_x. NH₃ was injected at the entrance of the top horizontal duct before the SCR system. The injected NH₃ level corresponded to an NH₃/NO_x ratio of 0.83. The SCR catalyst was conditioned with SO₂ and HCl for 4 hours prior to the test. A 90% NO_x removal across the SCR catalyst was observed. Hg⁰ was introduced in the vertical furnace section of the IFR via a separate injection nozzle.

Simultaneous isokinetic OH sampling was performed at the inlet and the outlet of the SCR catalyst. IGS probes were not used since there were no particulates during the natural gas combustion tests. Mercury speciation results are shown in Table 5.

Table 5: Hg speciation across the SCR catalyst for the natural gas test day 6/5/03
(results are in $\mu\text{g}/\text{dscm}$)

| Location | Hg^0 | Hg^{2+} | Hg_T | % Hg^{2+} |
|------------|---------------|------------------|---------------|--------------------|
| SCR inlet | 7.22 | 1.86 | 9.08 | 20% |
| SCR outlet | 6.35 | 2.48 | 8.83 | 28% |

Very good total Hg recovery was obtained across the SCR catalyst. The catalyst showed very little contribution to the oxidation of Hg^0 (only an 8% increase). It was hypothesized that the newly installed catalyst may need more aging in order to achieve a higher Hg^0 oxidation rate. The absence of particulates (fly ash) might also have a bearing on the catalytic Hg^0 oxidation reactions. A decision was made to continue with natural gas and Illinois coal combustion tests to induce aging in this catalyst. The natural gas combustion tests indicated that the IFR can be operated consistently and most of the injected mercury can be recovered at the outlet of the SCR catalyst.

The doped natural gas combustion test was repeated on 6/17/03 after the additional SCR reactor insulation was installed. A mercury continuous emission monitor (CEM) known as PS analytical (PSA) was used to measure Hg speciation on this test day. Natural gas was combusted at a firing rate of 127,630 Btu/hr. The excess air was calculated to be 16%. The O_2 concentration measured at the SCR outlet was 4.8% (calculated to be 3.3%) and the measured CO_2 concentration was 9.1% (calculated to be 9.9%). Comparison between measured and calculated values indicated an 8% in-leakage based on CO_2 and a 7% in-leakage based on O_2 (insignificant). There was no in-leakage between the inlet and the outlet of the SCR. The total flow (including the in-leakage) was calculated to be 31.1 scfm (0.883 scmm). NO_x and SO_2 were doped at 600- and 700-ppm levels, respectively. NH_3 and HCl were doped at 500- and 50-ppm levels (calculated), respectively. SCR catalyst was conditioned for 4 hours prior to the test with SO_2 and HCl. A 94% NO_x removal across the SCR catalyst was observed. Furthermore, the IGS probe was bias-checked by connecting combustion gas monitors to the sampling arm of the IGS probe. No difference in measurement of O_2 , CO_2 , SO_2 , and NO_x was observed, indicating no leak in the IGS probe. This is the standard operating procedure for the bias-check of the IGS probe.

SCR temperatures (top, middle, and bottom) are shown in Figure 3. As mentioned before, the new and thorough insulation of the SCR reactor caused a decrease in the axial temperature gradient from 80 to 50 $^\circ\text{C}$ (377 $^\circ\text{C}$ at the top and 327 $^\circ\text{C}$ at the bottom). The axial temperature gradients in the first and the second sections were 15 and 35 $^\circ\text{C}$, respectively.

The mercury CEM, PSA, was connected to the IFR duct downstream of the SCR catalyst and just upstream of the baghouse. Since no particulate matter (PM) was present during the natural gas combustion, the PSA was not connected to the IGS probe. PSA results are shown in Figure 4. NH_3 was added around the 13:20 time frame. Addition of ammonia caused an increase in Hg^0 and, thus, total Hg. This was observed before during the bench-scale reactor shake down tests. Probably, addition of NH_3 caused desorption of previously adsorbed Hg^0 . The PSA data before the addition of NH_3 (excluding the

outlier) can be compared to the OH tests performed on 6/5/03. PSA results as compared to the simultaneous isokinetic OH samplings performed at the inlet and the outlet of SCR catalyst are shown in Table 6.

Table 6: Hg speciation across the SCR catalyst for the doped natural gas combustion test days, comparison between OH and PSA (results are in $\mu\text{g}/\text{dscm}$)

| Location | Hg^0 | Hg^{2+} | Hg_T | % Hg^{2+} |
|-----------------------------------|---------------|------------------|----------------|--------------------|
| SCR inlet, OH sampling, 6/5/03 | 7.22 | 1.86 | 9.08 | 20% |
| SCR outlet, OH sampling, 6/5/03 | 6.35 | 2.48 | 8.83 | 28% |
| SCR outlet, PSA sampling, 6/17/03 | 7.8 ± 1.4 | - | 13.4 ± 3.0 | 12-60% |

Considering the experimental variability and the fact that OH and PSA sampling were performed on two different test days, a relatively good agreement was obtained. As discussed, this shows that the SCR catalyst did not contribute to the oxidation of Hg^0 in natural gas firing tests. Moreover, the PSA showed a large variability in the measurement of Hg even when the outlier result (see Figure 4) was not included. The reason is unknown and may be related to the sample-conditioning unit of the PSA.

Before proceeding to the coal tests, the potential biases of the IGS probe toward changes in Hg speciation needed to be investigated (an objective of natural gas test, Subtask I of Task II). Hg speciation in the IGS probe must be preserved in order to accurately quantify the Hg speciation in the duct of the IFR. A doped natural gas combustion test was performed on test day 6/18/03 to investigate potential Hg speciation biases created by the IGS probe. Combustion conditions were similar to those obtained on test day 6/17/03 (see above). The mercury CEM, PSA, was initially connected to the IFR duct downstream of the SCR catalyst and just upstream of the baghouse. After two hours of sampling from the duct (from 8:38 to 10:46), the PSA system was connected to the IGS probe. The PSA results are shown in Figure 5. As seen in this figure, the Hg speciation was preserved through the IGS probe, indicating that the IGS probe does not affect the Hg speciation in the duct. It should be noted that the IGS probe temperature (measured at the surface) was maintained at 240-250 °F (116-121 °C). Previous unpublished results have shown that, at higher temperatures [390 °F (200 °C)], the IGS probe causes oxidation of Hg^0 to Hg^{2+} . At these high temperatures, the stainless steel surfaces of the IGS probe appear to cause a catalytic conversion of Hg^0 to Hg^{2+} . Thus, it is crucial to maintain the temperature of the IGS probe in the low temperature range mentioned above. During this test, an attempt was also made to assess the effect of increased HCl concentration on Hg^0 oxidation by the SCR reactor. HCl concentration was increased from 50 to 100 ppm. Figure 5 shows that the increase in HCl concentration in the combustor had no effect on speciation of Hg, confirming that in natural gas firing tests the SCR catalyst does not contribute to the oxidation of Hg^0 . Note that the catalyst was conditioned with SO_2 and HCl for more than 24 hours.

6/17/03 run; SCR temperatures with new insulation

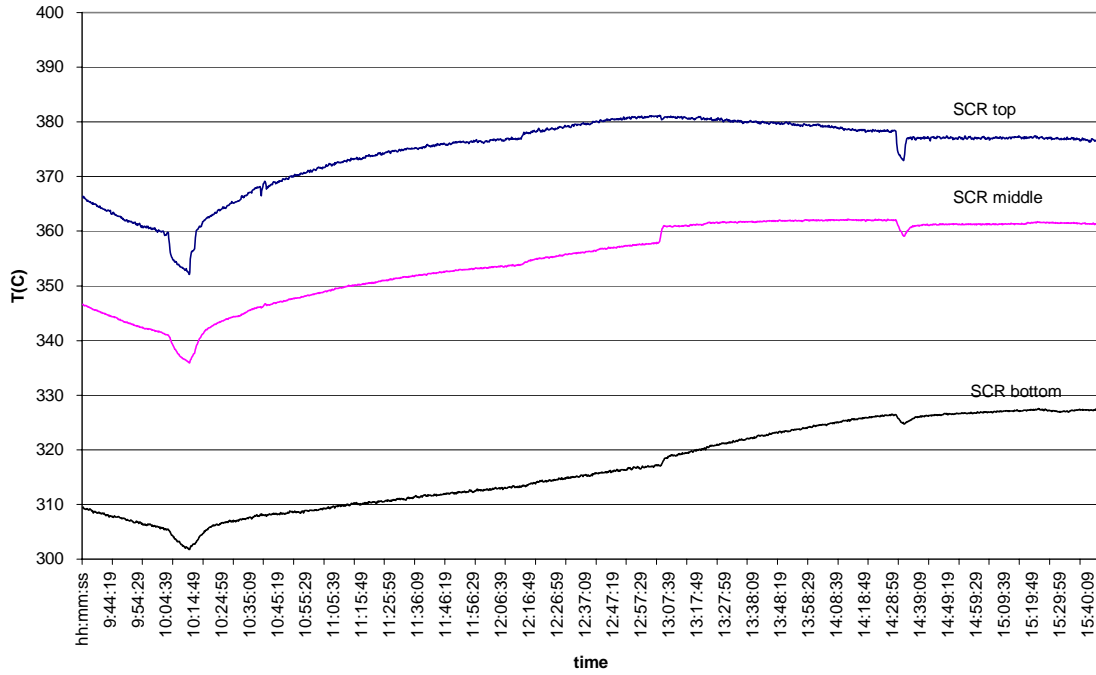


Figure 3: SCR temperature profiles for the doped natural gas test day 6/17/03; Hg CEM (PSA) sampling; new round of insulation across the SCR system

PSA results (downstream of the SCR), 6/17/03, Doped natural gas combustion, catalyst was conditioned with HCl and SO₂ for 4 hours

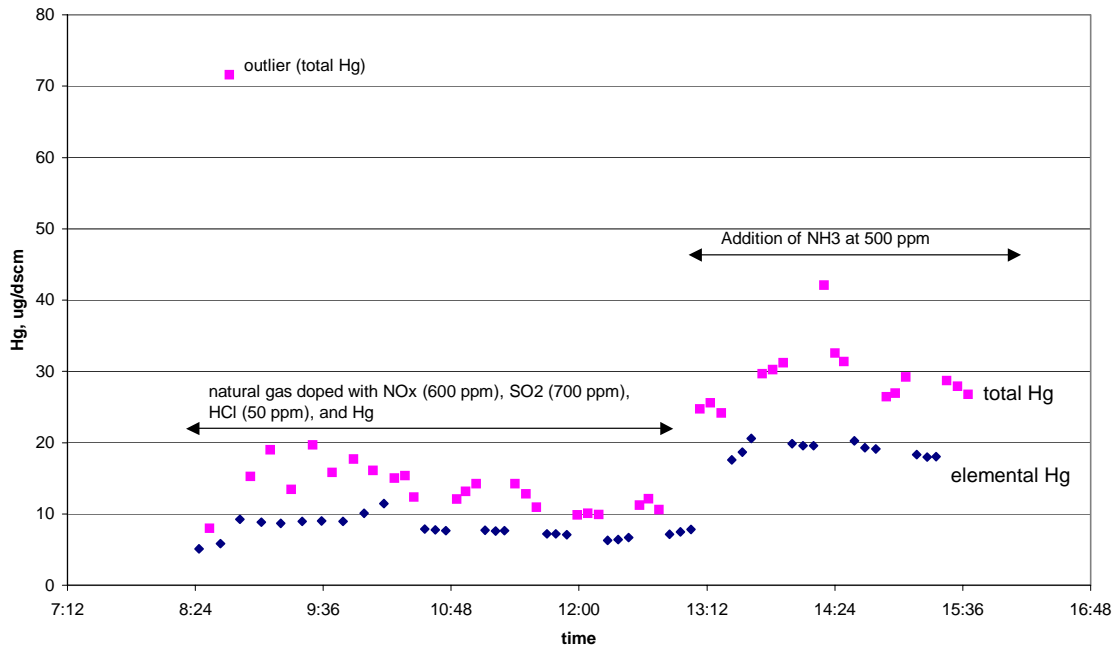


Figure 4: Hg CEM, PSA, results for the doped natural gas test day 6/17/03

6/18/03 run, PSA results, doped Natural gas combustion, catalyst was conditioned with SO₂ and HCl the night before; bias check of the IGS probe and the effect of HCl on Hg oxidation by the SCR catalyst

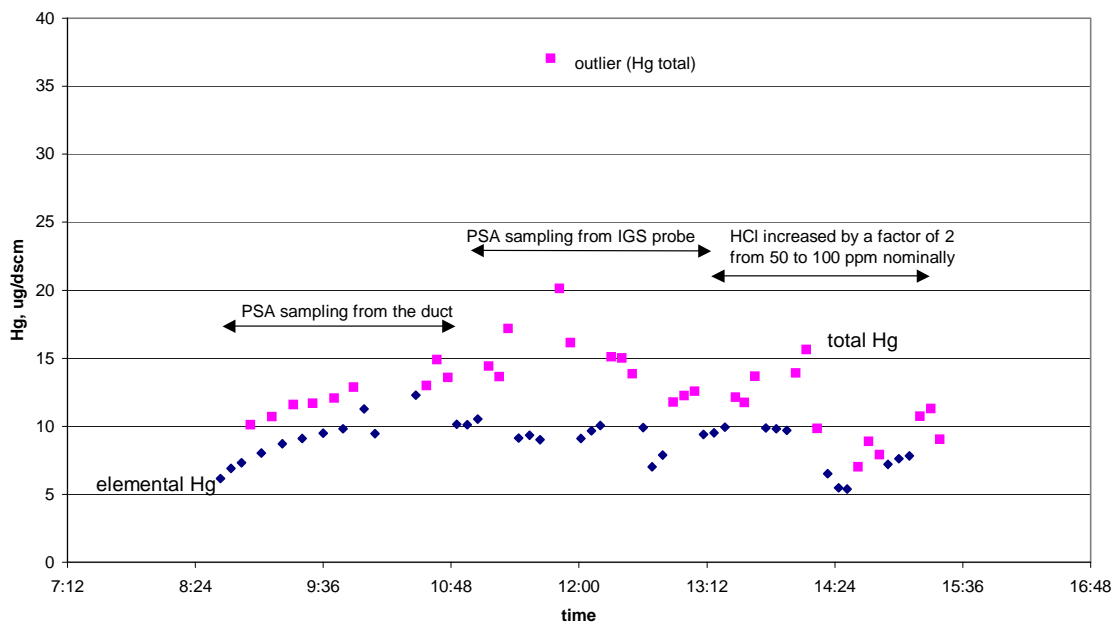


Figure 5: Hg CEM, PSA, results for the doped natural gas test day 6/18/03; IGS probe Hg speciation bias check and the effect of HCl on Hg⁰ oxidation

Task II, Subtask II: SCR Catalyst Evaluation in Illinois and PRB Coal Combustion

Galatia Coal Combustion

Galatia coal was tested under combustion conditions listed in Table 7 (in the Galatia column). Galatia coal was combusted at a firing rate of about 140,000 Btu/hr. Comparison between measured and calculated values of CO₂ and O₂ indicated an insignificant in-leakage of air into the combustor (about 4%). There was no in-leakage between the inlet and the outlet of the SCR. A very good combustion of Galatia coal was observed as indicated by low CO concentration (30±7 ppm) measured in the flue gas. The axial temperature gradient across the SCR catalyst was around 50 °C (change from 354 to 304°C). Uncontrolled levels of NO_x were around 850 ppm. NH₃ was injected at a nominal NH₃/NO_x ratio of 0.9. NO_x conversion around the SCR for the combustion of Galatia coal was about 86%. SO₂ concentration for this low sulfur Illinois coal was measured to be 929±63 ppm. The calculated level of SO₂ under the combustion conditions was 915 ppm, reflecting very good agreement with measured values.

Table 7: Combustion conditions for the Illinois Coals (Turriss, Galatia, and Crown II) and the PRB coal (Black Thunder)

| Parameter | Turriss | Galatia | Crown II | PRB |
|--|---------|----------|----------|----------|
| Coal feed rate (lb/hr) | 14.0 | 11.5 | 13.4 | 15.7 |
| IFR firing rate, Btu/hr | 147540 | 140424 | 145208 | 155873 |
| Total air flow (scfm) | 28.6 | 28.2 | 28.4 | 28.9 |
| Excess air, % | 11 | 18 | 11 | 5 |
| Top SCR temperature (°C) | 373±8 | 354±1 | 363±4 | 380±2 |
| Middle SCR temperature (°C) | 365±5 | 342±1 | 352±3 | 364±1 |
| Bottom SCR temperature (°C) | 323±4 | 304±2 | 315±2 | 325±1 |
| Measured CO, ppm (dry) | 38±6 | 30±7 | 40±4 | 0±1 |
| Uncontrolled NO _x , ppm (dry) | 960 | 850 | 650 | 525 |
| NO _x conversion across SCR, % | 90 | 86 | 90 | 90 |
| Measured SO ₂ , ppm (dry) | 2921±49 | 929±63 | 2739±26 | 222±2 |
| Calculated SO ₂ , ppm (dry) | 3064 | 915 | 3006 | 274 |
| O ₂ , measured (%), dry | 2.7 | 4.0 | 4.4 | 4.1 |
| O ₂ , calculated (%), dry | 1.8 | 3.0 | 1.9 | 0.1 |
| In-leakage based on O ₂ , % | 5 | 5 | 13 | 19 |
| CO ₂ , measured (%), dry | 15.6 | 14.8 | 14.3 | 15.6 |
| CO ₂ , calculated (%), dry | 16.0 | 15.4 | 15.7 | 18.0 |
| In-leakage based on CO ₂ , % | 3 | 3 | 9 | 13 |
| Calculated flow based on O ₂ in-leakage, scm/min | 0.879 | 0.896 | 0.944 | 1.008 |
| Calculated flow based on CO ₂ in-leakage, scm/min | 0.858 | 0.853 | 0.911 | 0.956 |
| Measured HCl, ppm (wet) | NM | 246±23 | NM | NM |
| Calculated HCl, ppm (wet) | 141 | 208 | 96 | 7.9 |
| PM conc., mg/dscm, SCR inlet* | 5863 | 3070±555 | 3946 | 2418±504 |
| PM conc., mg/dscm, SCR outlet* | 3506 | 1735±194 | 2560 | 1606±160 |

NM: Not Measured

* Measured using the filter weight of the isokinetic OH method at the inlet and outlet of the SCR

Simultaneous isokinetic and IGS probe OH sampling were performed at the inlet and outlet of the SCR reactor (i.e., upstream of baghouse) for the Galatia test. The isokinetic trains measure all three forms of mercury, Hg⁰, Hg²⁺, and Hg_p, and Hg_T can be calculated from the isokinetic train measurement. Note all samples were taken at locations upstream of the baghouse. Under these conditions, fly ash collected in the filter of the OH train may cause catalytic reaction or retention of mercury, resulting in a change in mercury speciation. Therefore, the IGS probe can be used for sampling gas-phase Hg⁰ and Hg²⁺, and the difference between Hg_T results obtained by the isokinetic train and the gas-phase Hg⁰ and Hg²⁺ obtained by the IGS probe can be used to estimate the amount of Hg_p. It was found during the initial coal combustion test that the total axial flow for the IGS probe installed at the inlet and the outlet of the SCR reactor dropped from between 8 and 7 to below 2.7 scfm. The total axial flow of the inlet of the IGS probe dropped from 5 to 3 scfm. This drop in the flow might have caused retention of particulate matter (PM, fly ash) in the IGS probes. This might have created an unwanted packed bed in the IGS probe, thus positively biasing the Hg_p results, although the results of the 2nd test (see

below) showed that this was not the case. The problem of drop in sampling flow was corrected by installing an air pump at the outlet for each of the IGS probes. The combination of the pump and the eductor (operated with compressed air) maintained the desired level of axial flow through the IGS probes. Biases created by packed beds of fly ash are not expected, as the high axial flow prevents formation of such packed beds in the probe. The total axial flow in the IGS probes was maintained at around 7-8 scfm.

Results of the simultaneous isokinetic and IGS probe OH sampling for the Galatia test are summarized in Tables 8 and 9. The flow through the IGS probes was maintained at around 8.7 scfm. Relatively good Hg_T recovery was obtained across the SCR catalyst. The Hg_T at the outlet was slightly lower than the inlet (7.2 vs. 9.9 $\mu\text{g}/\text{dscm}$). This could be due to adsorption of Hg by the SCR catalysts.

Comparison of the data obtained by using the isokinetic trains and those obtained by using IGS probes suggests that a considerable portion of the Hg in the Galatia coal combustion may be associated with the PM at the inlet of the SCR catalyst. However, the adsorption of mercury by fly ash is unlikely to be significant at the SCR inlet temperatures (354 °C), which is supported by recent field data that showed very little Hg_p at the inlet of various SCR systems firing different types of bituminous coals (Brickett et al., 2003). Deposition of white powder was observed occurring on the walls of the first impinger connecting to the IGS probes, indicating formation of fine particulate aerosols near the inlet of the impinger. Much more white powder was deposited in the impinger used for measuring the inlet of the SCR reactor than those in the impinger connecting to the outlet train. Formation of ammonium salts near the inlet of the first impinger connected to the IGS probe occurred as the NH_3 -containing flue gas cooled down, which could retain gas-phase Hg and changes in Hg speciation may result. It is well known that the reaction of NH_3 with sulfuric acid (H_2SO_4) condensed from SO_3 as the coal combustion flue gas cooled down produces ammonium salts (sulfates and bisulfate), which could cause severe air pre-heater plugging problem (Burke and Johnson, 1982). The capture of Hg^0 by the aerosols generated by the reactions of NH_3 and SO_3 under flue gas cooling conditions has been demonstrated by a recent bench-scale study (Lee, et al., 2003). The NH_3 concentration in flue gas decreased from about 850 ppm at the inlet of the SCR reactor to a few ppm at the reactor outlet as a result of the NO_x reduction reactions that consumed NH_3 . Thus, the potential for deposition of ammonium salts in the sampling system used at the inlet of the SCR reactor is significantly higher than that used at the reactor outlet. The discrepancies in Hg speciation measured by the isokinetic OH train and the calculated speciation are much larger for the SCR inlet case (Table 8) than those for the outlet case (Table 9). The larger discrepancies for the inlet case are consistent with more white powder deposition found in the first impinger of the inlet IGS probe. It was also observed during the initial shakedown tests of the bench-scale reactor (Task I) that white powders were found deposited near the inlet of an on-line mercury analyzer connecting to the outlet of the SCR reactor for measuring Hg^0 continuously, indicating aerosol formation. The white powder deposits were observed only when NH_3 was added to the simulated flue gases, and the data measured by using the analyzer showed continuous drift as the deposit was accumulating during the test. A decision was made to use the OH sampling method for the Task I tests in order to avoid the measuring

problem created by the aerosol formation when using the mercury analyzer. It appears that the formation of ammonium salts in the Hg speciation sampling/measurement system in the presence of NH_3 and under cooling conditions may cause significant bias of the speciation measurement. Further research and development work is needed for applying the IGS probe to measure Hg speciation in SCR systems. Therefore, it was decided to not use IGS probe in this work.

Table 8: Galatia coal combustion test, OH sampling results measured at the **inlet** of the SCR reactor, Hg in $\mu\text{g}/\text{dscm}$

| Inlet OH Train | Hg^0 | Hg^{2+} | Hg_p | Hg_T | % Hg^0 | % Hg^{2+} | % Hg_p |
|---------------------------------|---------------|------------------|---------------|---------------|-----------------|--------------------|-----------------|
| Isokinetic | 7.2 | 2.1 | 0.61 | 9.9 | 73 | 21 | 6 |
| IGS probe | 1.7 | 3.9 | - | - | - | - | - |
| Calculated inlet Hg speciation* | 1.7 | 3.9 | 4.3 | 9.9 | 17 | 40 | 43 |

* Calculated assuming that isokinetic train is used for measuring Hg_T , and IGS probe train is used for measuring Hg^0 and Hg^{2+}

Table 9: Galatia coal combustion test, OH sampling results measured at the **outlet** of the SCR reactor, Hg in $\mu\text{g}/\text{dscm}$

| Outlet OH Train | Hg^0 | Hg^{2+} | Hg_p | Hg_T | % Hg^0 | % Hg^{2+} | % Hg_p |
|----------------------------------|---------------|------------------|---------------|---------------|-----------------|--------------------|-----------------|
| Isokinetic | 0.32 | 5.1 | 1.8 | 7.2 | 4 | 71 | 25 |
| IGS probe | 0.1 | 5.4 | - | - | - | - | - |
| Calculated outlet Hg speciation* | 0.1 | 5.4 | 1.7 | 7.2 | 1 | 75 | 24 |

* Calculated assuming that isokinetic train is used for measuring Hg_T , and IGS probe train is used for measuring Hg^0 and Hg^{2+}

The OH sampling results show very good oxidation of Hg^0 across the SCR catalyst. Hg^0 was decreased from 73 to 4% and Hg^{2+} was increased from 21 to 71% across the SCR catalyst. The change in Hg speciation from predominantly Hg^0 at the inlet of the catalyst to predominantly Hg^{2+} at the outlet was observed in SCR systems firing bituminous coals (Brickett, et al., 2003). The isokinetic OH filter weights can be used to determine the PM concentration in the IFR. The average PM concentrations for the Galatia coal combustion at the inlet and the outlet of SCR reactor are shown in Table 7. The lower PM concentration at the outlet may be due to the operation of the IGS probe at the inlet. This probe draws a high flow from the duct and, thus, may preferentially take higher amounts of PM from the flue gas.

Turrís Coal combustion

Turrís coal was combusted at a firing rate of about 148,000 Btu/hr. In-leakage of air into the combustor was about 4%. There was no in-leakage between the inlet and the outlet of the SCR. A very good combustion of Turrís coal was observed as indicated by the low CO emissions (38 ppm). The axial temperature gradient across the SCR catalyst was around 50 °C (change from 373 to 323°C). Slightly higher temperatures were obtained around the SCR catalyst as compared to the Galatia coal combustion test. Uncontrolled levels of NO_x were around 950 (higher than Galatia). NH_3 was injected at an NH_3/NO_x

ratio of about 0.9. NO_x conversion across the SCR was about 90%. SO_2 concentration for this high sulfur Illinois coal was measured to be 2921 ± 49 ppm. The calculated level of SO_2 under the combustion conditions was 3046 ppm, reflecting good agreement with measured values.

Table 10 summarizes the results of the OH sampling for the Turriss coal test at the inlet and the outlet of the SCR reactor. Only about half of the inlet Hg_T was recovered at the outlet. This could be due to the adsorption of mercury by the catalyst. Very little Hg was associated with the particulate matter. The SCR catalyst exhibited a strong Hg^0 oxidation capability, dropping the amount of Hg^0 from 68% at the inlet to 9% at the outlet. The Hg^{2+} was increased from 9 to 79% at the two sampling locations. Such significant changes in Hg speciation are in agreement with those observed in the field for bituminous coal-fired SCR systems (Brickett, et al., 2003).

Table 10: Turriss coal combustion test, OH sampling results measured at the inlet and outlet of the SCR reactor, Hg in $\mu\text{g}/\text{dscm}$

| Hg Speciation | Hg^0 | Hg^{2+} | Hg_P | Hg_T | % Hg^0 | % Hg^{2+} | % Hg_P |
|---------------|---------------|------------------|---------------|---------------|-----------------|--------------------|-----------------|
| Inlet | 4.7 | 0.6 | 1.5 | 6.9 | 68 | 9 | 23 |
| Outlet | 0.3 | 2.6 | 0.4 | 3.3 | 9 | 79 | 12 |

Crown II Coal combustion

Crown II coal was combusted at a firing rate of about 145,000 Btu/hr. In-leakage of air into the combustor was about 11%. It appears that as the combustor operated for a longer period of time, air in-leakage increased. This level of air in-leakage is considered insignificant. There was no air in-leakage between the inlet and the outlet of the SCR. A very good combustion of Crown II coal was indicated by low measured CO emissions (40 ppm). The axial temperature gradient across the SCR catalyst was around 48 °C (change from 363 to 315°C). SCR temperatures were within the range for those obtained during the other two Illinois coal tests. Uncontrolled levels of NO_x were about 650 ppm, lower than those for the Galatia and Turriss tests. NH_3 was injected at a calculated NH_3/NO_x ratio of about 0.8. NO_x conversion across the SCR was about 90%. SO_2 emissions for this high sulfur Illinois coal tests measured 2739 ± 49 ppm. The calculated level of SO_2 emission under these test conditions was 3006 ppm.

Table 11 summarizes the results of the OH sampling for the Crown II coal combustion test at the inlet and the outlet of the SCR reactor. Relatively low Hg_T recovery was obtained across the SCR catalyst. The Hg_T concentration measured at the outlet was lower than that measured at the inlet (4.1 vs. 5.5 $\mu\text{g}/\text{dscm}$). The lower Hg_T measured at the outlet of the SCR catalyst may be due to the adsorption of Hg by the catalyst. Very little Hg was associated with the PM. This was also found during the combustion of Turriss coal. Crown II and Turriss coals are very similar in their properties (see Table 1). Galatia was the only Illinois coal that generated relatively high Hg_P during its combustion. The SCR catalyst exhibited a very good Hg^0 oxidation capability. Hg^0 was reduced from 84% at the inlet down to 12% at the outlet, with the increase in Hg^{2+} from 15 to 85% across these two sampling locations. The changes in Hg speciation across the

SCR catalyst observed for the Crown II coal combustion are consistent with those of the two other Illinois coal tests and also those of the bituminous coal-fired SCR systems observed in field (Brickett, et al., 2003).

Table 11: Crown II coal combustion test, OH sampling results measured at the inlet and outlet of the SCR reactor, Hg in $\mu\text{g}/\text{dscm}$

| Hg Speciation | Hg ⁰ | Hg ²⁺ | Hg _P | Hg _T | %Hg ⁰ | % Hg ²⁺ | % Hg _P |
|---------------|-----------------|------------------|-----------------|-----------------|------------------|--------------------|-------------------|
| Inlet | 4.6 | 0.8 | 0.1 | 5.5 | 84 | 15 | 1 |
| Outlet | 0.5 | 3.5 | 0.1 | 4.1 | 12 | 85 | 3 |

PRB (Black Thunder) coal combustion

The Black Thunder PRB coal was combusted in a duplicate run. The combustion conditions were identical and are listed in Table 7 under the PRB column. PRB coal was combusted at a firing rate of about 156,000 Btu/hr. This firing rate was slightly higher than those used for the Illinois coal tests. In-leakage of air into the combustor was about 16%. Similar to the Crown II test, as the combustor operated for a longer period of time, in-leakage seemed to increase. The levels of air in-leakage are considered to be acceptable. There was no in-leakage between the inlet and the outlet of the SCR. A very good combustion of the PRB coal was observed; no CO emissions were measured during the two combustion tests. The axial temperature gradient across the SCR catalyst was about 55 °C (change from 380 to 325°C). SCR temperatures were slightly higher than those observed during Illinois coal tests, possibly due to slightly higher firing rate. Uncontrolled levels of NO_x emissions were about 525 ppm, which are lower than those measured for the Illinois coal tests. NH₃ was injected at a calculated NH₃/NO_x ratio of about 0.8. NO_x conversion across the SCR was about 90%. SO₂ emissions for the combustion of this low sulfur coal was measured to be 222±2 ppm. The calculated level of SO₂ under the combustion conditions was 274 ppm, indicating good agreement with the measured values.

Tables 12 and 13 summarize the results of the OH sampling for the first and second PRB coal combustion tests, respectively. A good Hg_T recovery was obtained across the SCR catalyst during the second PRB test (5.1 $\mu\text{g}/\text{dscm}$ at the inlet vs. 4.6 $\mu\text{g}/\text{dscm}$ at the outlet), which was better than those obtained during the first test (7.2 $\mu\text{g}/\text{dscm}$ at the inlet vs. 5.1 $\mu\text{g}/\text{dscm}$ at the outlet). The inlet Hg_T obtained during the first test (7.1 $\mu\text{g}/\text{dscm}$) is also significantly higher than that measured at the same location for the second test, which may be an outlier. It appears that, at the inlet, almost all Hg was in elemental form with no appreciable Hg²⁺ or Hg_P. The comparison of data measured between inlet and outlet of the catalyst shows only a very small drop in Hg⁰, from 96% to 76% for the second test and from 97% to 88% for the first test. This is equivalent to only a 20-percent oxidation of Hg⁰ (9-percent oxidation of Hg⁰ for the first test). Field sampling conducted to date indicates the same Hg speciation profile as shown by this study, mostly Hg⁰ at the inlet and very low Hg⁰ oxidation. PRB coals in general have significantly lower chlorine and higher alkaline contents than those of the bituminous coals. The low oxidation of Hg⁰ across the SCR catalyst observed for the two PRB coal tests supports the hypothesis that HCl, which serves as the chlorine source, is critical in Hg⁰ oxidation across the SCR

catalyst. The importance of HCl in Hg oxidation across the SCR catalyst had been demonstrated in Task I.

Table 12: First PRB coal combustion test, OH sampling results measured at the inlet and outlet of the SCR reactor, Hg in $\mu\text{g}/\text{dscm}$

| Hg Speciation | Hg^0 | Hg^{2+} | Hg_p | Hg_T | % Hg^0 | % Hg^{2+} | % Hg_p |
|---------------|---------------|------------------|---------------|---------------|-----------------|--------------------|-----------------|
| Inlet | 7.0 | 0.2 | 0.0 | 7.2 | 97 | 3 | 0 |
| Outlet | 4.5 | 0.5 | 0.1 | 5.1 | 88 | 10 | 2 |

Table 13. Second PRB coal combustion test, OH sampling results measured at the inlet and outlet of the SCR reactor, Hg in $\mu\text{g}/\text{dscm}$

| Hg Speciation | Hg^0 | Hg^{2+} | Hg_p | Hg_T | % Hg^0 | % Hg^{2+} | % Hg_p |
|---------------|---------------|------------------|---------------|---------------|-----------------|--------------------|-----------------|
| Inlet | 4.9 | 0.2 | 0.0 | 5.1 | 96 | 4 | 0 |
| Outlet | 3.5 | 1.0 | 0.1 | 4.6 | 76 | 22 | 2 |

CONCLUSIONS AND RECOMMENDATIONS

A two-task, bench- and pilot-scale research study was conducted to evaluate the viability of SCR-based mercury control technology utilizing oxidation of Hg^0 to Hg^{2+} and subsequent removal by conventional FGD systems. Mercury speciation modification was implemented through utilization of SCR catalysts, currently used for NO_x control. The effect of SCR catalysts on mercury speciation was investigated in Illinois and PRB coal combustion flue gases. This research was designed to demonstrate the achievable Hg^{2+} levels in Illinois and PRB coal combustion processes utilizing SCR systems.

Task I involved bench-scale testing of a SCR catalyst formulation to characterize oxidation of Hg^0 to Hg^{2+} in the presence of coal combustion/SCR flue gas species such as NO_x , NH_3 , SO_2 , and HCl. The NO_x , NH_3 , HCl, and SO_2 concentration ranges employed during Task I were based on those expected to be encountered in flue gases resulting from firing of three representative Illinois coals (low to high sulfur and chlorine) and one PRB coal (very low sulfur/chlorine). Task I results showed that HCl (with concentrations as low as 8 ppm) is the 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 all the simulated Illinois and PRB coal tests. Task I tests established the Hg^0 oxidation activity of the formulated SCR catalyst.

Pilot-scale Task II tests were performed using a SCR catalyst installed in a vertical downward flow configuration in the post-combustion region of a pilot-scale 34.9 kW (150,000 Btu/hr), refractory lined, down-fired cylindrical furnace. Three different Illinois coals and one PRB coal were combusted in the pilot-scale facility. For the high sulfur/low chlorine Illinois coals, about 84-68 % Hg^0 , and 15-9% Hg^{2+} were measured at the inlet of the pilot-scale SCR reactor. The SCR catalyst induced high oxidation of Hg^0 , dropping the percentage of Hg^0 at the outlet of the SCR to values below 16%. The low sulfur/high chlorine Illinois coal had a relative high amount (21%) of Hg^{2+} at the inlet of SCR. The Hg^0 content of this coal flue gas was decreased from about 73% at the inlet to about 4%

at the outlet of SCR reactor. These results indicate that installation of SCR systems in Illinois coal-fired boilers that are equipped with a wet FGD system may achieve about 85 to greater than 90% control of mercury.

The PRB coal tests indicated a low oxidation of Hg^0 by the SCR catalyst. Based on the measurements conducted in this study, it appears that SCR applications on PRB (Black Thunder) coal-fired boilers may not result in significant increase in Hg^{2+} content of flue gas. Therefore, such boilers equipped with wet FGD systems may not achieve increased mercury removal resulting from SCR applications.

The following recommendations are made based on the results of this one-year study:

- For most typical Illinois coals (high sulfur and medium to low chlorine), installation of SCR catalysts prior to FGD systems should be very beneficial. In addition to achieving a 90% reduction in NO_x , about 85 to greater than 90% reduction in mercury may be possible.
- A particular low sulfur/high chlorine Illinois coal (Galatia) showed relatively high Hg^{2+} content at the SCR inlet (21%); the rest was predominantly Hg^0 (73%) and very little Hg_p . However, the Hg contents at the SCR outlet were changed to 71% Hg^{2+} , 25% Hg_p , and very little Hg^0 . Based on these data, installation of an SCR catalyst may result in greater than 90% reduction of Hg across the PM control and the wet FGD systems.
- The benefit of installing a SCR catalyst for the tested PRB coal (Black Thunder) appears to be very limited. Installation of an SCR catalyst could only increase the mercury removal to a maximum of 20%.
- Further tests are required to evaluate the long-term activity of the tested SCR catalysts in both Illinois and PRB coal combustion flue gases.
- Tests should be conducted to evaluate the effect of SCR catalyst space velocity on Hg^0 oxidation in Illinois and PRB coal combustion flue gas. Higher space velocities ($>3000 \text{ hr}^{-1}$) should be tested to demonstrate further cost-effectiveness of this technology.

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