PILOT-SCALE EVALUATION OF ACTIVATED CARBON-BASED MERCURY CONTROL OPTIONS FOR UTILITIES BURNING LIGNITE COAL

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Abstract

Activated carbon sorbents were evaluated for mercury removal potential in bench- and pilot-scale systems at the Energy & Environmental Research Center (EERC). The Ontario Hydro method and continuous mercury monitors were used to measure mercury species concentrations across the control technology devices with and without sorbent injection. Primarily elemental mercury (Hg^0) was measured when lignite coals from the Poplar River Plant and Freedom Mine were combusted. The effects of sorbent injection rate, particle size, and gas temperature on mercury removal were evaluated for four particulate control device options. Increasing injection rates, decreasing gas temperatures, and improving contact between the sorbent and flue gas all generally promoted mercury capture. Relative to eastern bituminous coal combustion flue gases, higher sorbent injection rates were generally required to effectively remove mercury from the lignite coal combustion flue gases. Similar issues of high Hg^0 and slow kinetics apply to other western coals as well. This paper summarizes the effects of chlorine and other additives on the oxidation and removal of mercury from flue gas.

Introduction

In general, lignite coals contain comparable levels of mercury but significantly lower levels of chlorine compared to bituminous coals. Lignite coals are also distinguished by their much higher calcium contents. These compositional differences have important effects on the quantity and form of mercury emitted from a boiler and the effectiveness of different control technologies to remove mercury from flue gas. The high chlorine content (>200 ppm) that is characteristic of many bituminous coals increases the fraction of the more easily removable mercuric compounds (Hg^{2+}), most likely mercuric chloride ($HgCl_2$), in the total mercury emission. Conversely, experimental results and information collection request data indicate that low-chlorine (<200 ppm) coal combustion flue gases contain predominantly Hg^0, which is substantially more difficult to remove than Hg^{2+}.[1] Additionally, the generally high calcium contents of lignite coals may further reduce the oxidizing effect of the already low chlorine content by reactively scavenging chlorine species (Cl, HCl, and Cl_2) from the combustion flue gas.[2]

The most commonly considered strategy for removing mercury from coal combustion flue gas streams is the adsorption of mercury species by a solid sorbent injected upstream of a particulate control device such as a fabric filter (FF) or electrostatic precipitator (ESP). Many potential mercury sorbents have been evaluated.[3] These evaluations have demonstrated that the flue gas composition and the chemical speciation of mercury affect mercury capture and its ultimate environmental fate.

Currently, powdered activated carbon (PAC) injection appears to be the most mature and promising technology available for mercury control. Research and test data suggest that activated carbons can effectively sorb both Hg^0 and Hg^{2+}. However, much of the research to date has been performed in fixed-bed reactors that simulate relatively long residence time (minutes or hours) and intimate gas–solids contact on a FF cake.[4–9] It is equally important to investigate short residence time (seconds) in-flight capture of Hg^0 because most of the coal-burning boilers in the United States and Canada use cold-side ESPs for controlling particulate matter emissions. The projected annual cost for activated carbon adsorption of mercury in a duct injection system is significant. Carbon-to-mercury weight ratios of 3000B18,000 (gram of carbon injected per gram of mercury in flue gas) have been estimated to achieve 90% mercury removal from a coal combustion flue gas containing 10 μg/Nm^3 of mercury.[3] Pilot data from this project suggest that, for lignite-fired plants, the carbon-to-mercury weight ratio required may be toward the upper end of this range. More efficient carbon-based sorbents and contacting systems are required to enable lower carbon-to-mercury weight ratios to be used, thus reducing the operating costs of PAC injection.

Researchers at the Energy & Environmental Research Center (EERC) and elsewhere are striving to attain a more thorough understanding of mercury species reactions on activated carbon surfaces in order to produce more efficient sorbents.[10,11] The removal of mercury from flue gas by activated carbon probably occurs through reactions with surface functional groups. Mercury-reactive surface functional groups may include acidic carboxyl, lactone, hydroxyl, and carbonyl or alkaline pyrone and chromene functionalities.[10–15] The potential role of acidic and alkaline surface functional groups on mercury capture is unknown and needs to be investigated. Functional groups containing inorganic elements such as chlorine or sulfur are also possibilities.[10,11,16–20] Although chlorine- and sulfur-bearing surface functional groups are not well characterized, the beneficial role of chlorine and the often negative impact of SO_2 and SO_3 in capturing mercury species on activated carbons are well established.[15,21]

The EERC recently completed the first phase of a 3-year, two-phase consortium project to develop and demonstrate mercury control technologies for utilities burning lignite coal. Phase I objectives were to develop a better understanding of mercury interactions with flue gas constituents, test a range of sorbent-based technologies targeted at removing mercury dominated by the elemental form (Hg^0) from flue gases, and demonstrate the effectiveness of the most promising technologies at the pilot scale. The Phase II objective is to demonstrate and quantify sorbent technology effectiveness, performance, and cost at a sponsor-owned and/or operated power plant. This paper documents the Phase I results and provides a brief overview of the Phase II plans.

Results and Discussion

Bench-Scale Testing. Fixed-bed tests were completed on a NORIT FGD sorbent and a calcium-based sorbent. Two simulated coal combustion flue gas compositions were used during the tests as presented in Table 1. The simulated lignite flue gas composition is based on flue gas measurements made at a lignite-fired power plant and derived from the relatively low chlorine contents of the Luscar (18.0 ppmw), Beulah–Zap (12.6 ppmw), and Center (14.3 ppmw) coals. The second simulated flue gas composition in Table 1 had been used in previous testing, thus enabling comparisons to be made with other sorbent test results. The baseline flue gas contains much higher SO_2, NO, NO_2, and HCl concentrations but lower H_2O concentrations relative to the lignite flue gas. Test results are presented in Figures 1–3. These figures show the temporal changes in total mercury concentration downstream from the fixed-sorbent beds expressed as a percentage of Hg^0 input (nominally 10 μg/m^3) into the system. Figure 1 shows results for the activated carbons prepared at the baseline conditions, unactivated chars, and a calcium-based sorbent tested in the simulated lignite flue gas (Table 1). Mercury was initially passed through the activated carbons and then effectively captured for about 3 hr. After 3 hr, mercury began to

break through the fixed-sorbent beds at progressively higher levels and was released primarily as Hg^{2+} (>90%).

Table 1. Simulated Coal Combustion Flue Gas Compositions

Compone Component, unit	Lignite	Baseline
O_2, vol%	6	6
CO_2, vol%	12	12
H_2O, vol%	15	8
SO_2, ppmv	580	1600
NO, ppmv	120	400
NO_2, ppmv	6	20
HCl, ppmv	1	50
N_2	Balance	Balance

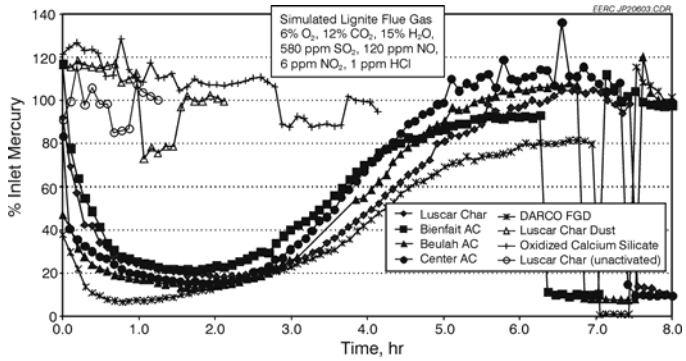

Figure 1. Bench-scale fixed-bed results under simulated lignite flue gas for carbons prepared at baseline conditions.

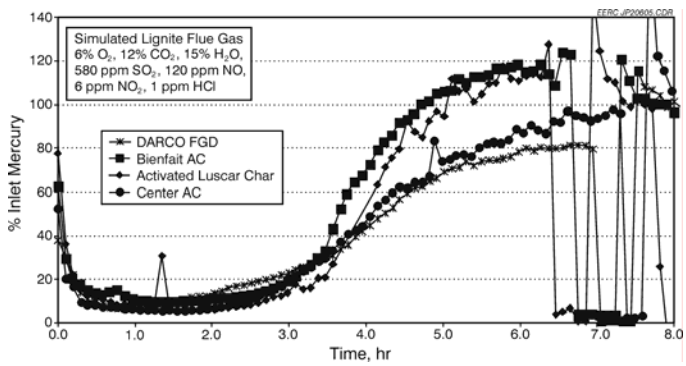

Figure 2. Bench-scale fixed-bed results under simulated lignite flue gas for carbons prepared at an increased temperature.

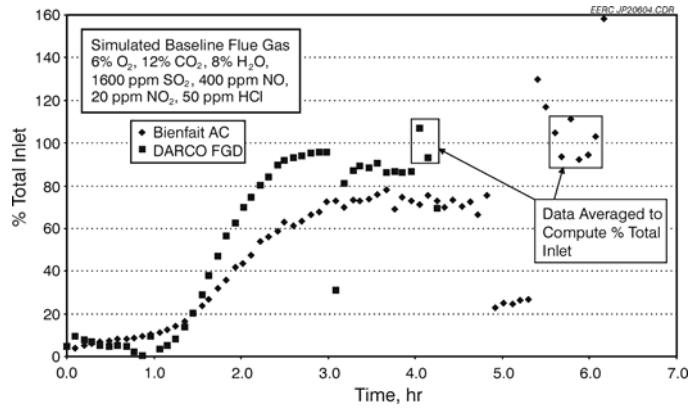

Figure 3. Bench-scale fixed-bed results under simulated baseline flue gas for Luscar coal activated at an increased temperature and NORIT FGD.

The initial breakthrough of mercury during the first 30 to 40 min of testing does not generally occur in flue gases containing higher acid gas concentrations. The unactivated sorbents and calcium-based sorbent were ineffective in capturing mercury; consequently, the tests were discontinued after 2 to 4 hr. Figure 2 compares test results for the carbons activated at an increased temperature and commercial carbon using the simulated lignite flue gas composition. Relative to the carbons activated at baseline conditions (Figure 1), results in Figure 2 indicate improved mercury capture and greater conversions of Hg^0 to Hg^{2+} (>95%). In addition, the initial breakthrough of mercury was not as significant. In order to compare results with a dataset of past test results, the Luscar coal carbon activated at an increased temperature and the NORIT FGD carbon were tested using the baseline flue gas composition in Table 1. Figure 3 test results show better initial mercury capture with no breakthrough. After 1.5 hr, the Luscar coal activated carbon showed less breakthrough compared to the NORIT FGD.

Pilot-Scale Combustor. A 580-MJ/hr (550,000-Btu/hr) pulverized coal-fired unit was used to evaluate mercury sorbent effectiveness in flue gases produced from combustion of lignite coal. The unit, shown schematically in Figure 4, is designed to generate fly ash and flue gas representative of that produced in a full-scale utility boiler. The combustor is oriented vertically to minimize wall deposits. A refractory lining helps to ensure adequate flame temperature for complete combustion. Based on the superficial gas velocity, the mean residence time of a particle in the combustor is approximately 3 seconds. The coal nozzle fires axially upward from the bottom of the combustor, and secondary air is introduced concentrically to the primary air with turbulent mixing. Coal is introduced to the primary air stream via a screw feeder and eductor. An electric air preheater is used for precise control of the combustion air temperature.

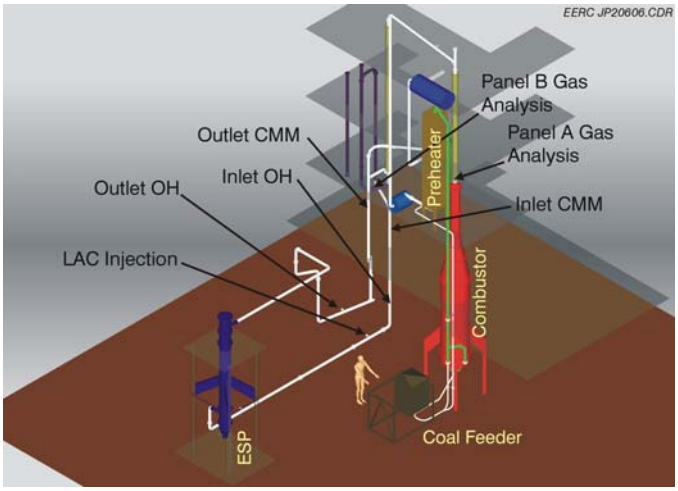

Figure 4. Schematic of the 580-MJ/hr (550,000-Btu/hr) combustion system.

The following particulate control devices were evaluated on the pilot-scale combustor as potential mercury control options: ESP, FF, combined ESPBFF, and *Advanced Hybrid*™ filter technology. Instrumentation enables system temperatures, pressures, flow rates, flue gas constituent concentrations, and particulate control device operating data to be monitored continuously and recorded on a data logger.

Pilot-Scale Tests. The activated (800°C, 1472°F) Luscar char (Bienfait) and DARCO FGD were selected for additional testing in the 580-MJ/hr (550,000-Btu/hr) pulverized coal-fired unit based on sorbent-screening results (reactivity and capacity), physical properties (particle size and surface area), cost, and consensus among project sponsors. The following variables that could potentially affect mercury emission control were tested: lignite coal source (Poplar River or Freedom Mine), control device type (ESP, FF, ESPBFF, or *Advanced Hybrid*™ filter), FF type (Gore-Tex® or Ryton®), sorbent type (steam-activated [800°C, 1472°F] Luscar char or DARCO FGD), particle size (approximate median volume diameters [MVDs] of 20 or 5 µm), sorbent injection rate, and flue gas temperature in the pollution control device. Additional tests were performed to evaluate the effectiveness of chlorine-based additives for enhancing mercury oxidation and removal.

Mercury Speciation. Figure 5 compares the average mercury species distributions, as determined by American Society for Testing and Materials Method D6784-02 (Ontario Hydro [OH]), for the Poplar River and Freedom coal combustion flue gases. The Poplar River coal combustion flue gas contains a higher total mercury concentration; however, the relative proportions of Hg^0, Hg^{2+}, and Hg(p) in both flue gases were very similar at approximately 85%, 15%, and <1%, respectively.

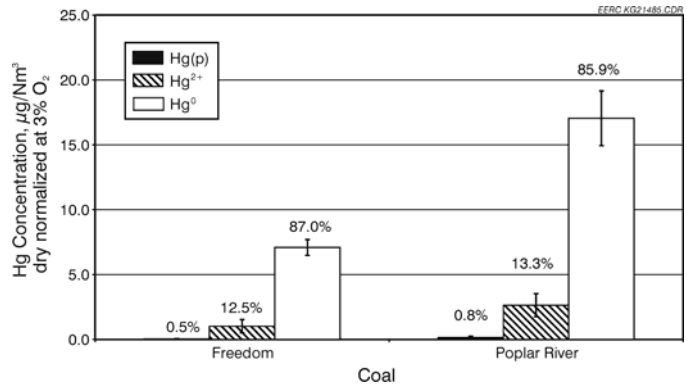

Figure 5. Comparison of average Poplar River and Freedom coal combustion flue gas (149°C, 300°F) mercury speciation results obtained using the OH method.

Effect of Varying Injection Rate on Technology Options. Figures 6 and 7 summarize the impact of varying injection rate for the four particulate technology configurations and the Poplar River and Freedom coals, respectively. For comparison purposes, mercury removal efficiencies for each technology configuration are summarized in Table 2 using the lowest injection rate observed from either coal. In general, the Hg removal efficiencies increased with the carbon injection rate for different particulate control configurations. Clearly, the TOXECON™ configuration (injection downstream of an ESP and upstream of a FF) results in the highest reduction while using the least amount of PAC. However, in batch injection mode, the *Advanced Hybrid*™ filter provides similar performance.

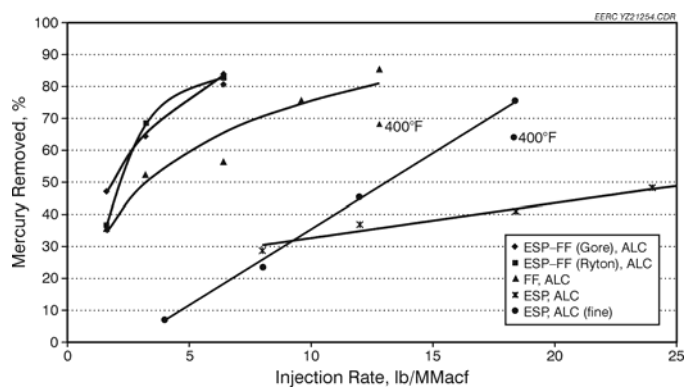

Figure 6. Particulate matter control device efficiencies for removing mercury from 149°C (300°F) Poplar River coal combustion flue gas as a function of activated (800°C, 1472°F) Luscar char injection rate.

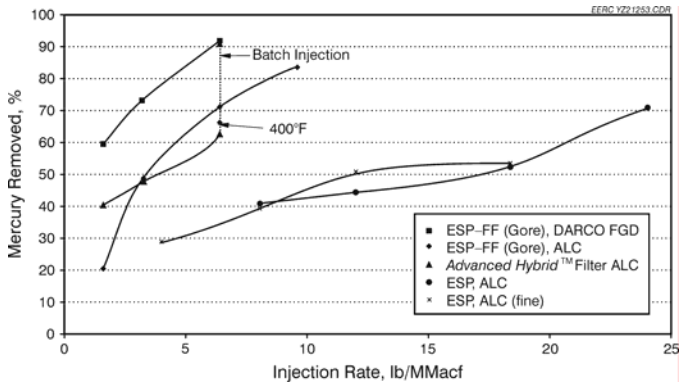

Figure 7. Particulate matter control device efficiencies for removing mercury from 149°C (300°F) Freedom coal combustion flue gas as functions of activated (800°C, 1472°F) Luscar char and DARCO FGD injection rates.

Table 2. Injection Rate (lb/MMmacf)* for Target Removal Rates

Technology Configuration	50%	70%	80%
ESP Only	11.2	17.1	NA
FF Only	3.2	7.8	12.3
ESP–FF	1.9	2.92	4.4
Advanced Hybrid™ Filter	6.0	NA	6.4**

* Lowest observed rate.
** Batch injection at greater than 80% removal.

Effect of Coal. Pilot-scale data showed a higher PAC rate requirement than would be expected based on past results with other coals. Figure 8 compares the mercury removal effectiveness of PAC injection combined with pilot-scale ESP and ESPBFF devices to that obtained by Bustard and others[22] with PAC injection between an ESP–FF (TOXECON™) or ESP installed on full-scale utility boilers. Coal type (i.e., composition) is an important parameter that affects the mercury removal efficiency of a control device. During the pilot-scale lignite tests with TOXECON™ and utility-scale eastern bituminous coal tests, mercury removal efficiency increased with increasing PAC injection rates. Conversely, mercury removal efficiency was never greater than 70%, regardless of the PAC injection rate into the Powder River Basin (PRB) subbituminous coal combustion flue gas. This limitation is probably caused by the low level of chlorine species that can promote mercury–activated carbon adsorption. In addition, the generally abundant lime (CaO) component of PRB subbituminous coal fly ashes reactively scavenges chlorine species (Cl, HCl, and Cl_2) from the flue gas to form $CaCl_2$. Figure 6 indicates that PAC injection combined with the particulate control devices installed on the full-scale boilers generally provided better mercury removal efficiency at a given injection rate relative to PAC injection followed by the pilot-scale ESP. Mercury removal efficiencies for the TOXECON™ configuration shown in Figure 6 are those observed for an eastern bituminous flue gas.

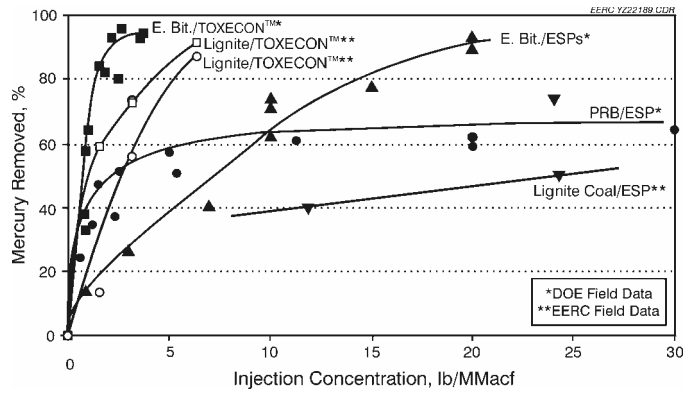

Figure 8. Pilot- and full-scale ESP and TOXECON™ mercury removal efficiencies as a function of PAC injection rate.

Effect of Chlorine. The use of chlorine and carbon-based sorbents for mercury removal has been effective in municipal, hazardous, and hospital waste incineration flue gases. Additionally, chemical kinetic modeling of bench-scale test results indicates that the introduction of HCl or NaCl into the high-temperature furnace region is likely to result in the production of atomic chlorine (Cl) and/or molecular chlorine (Cl_2), which are generally thought to be the dominant Hg^0 reactants in coal combustion flue gases.[23–28] Pilot-scale tests were conducted on two coals to determine if chlorine addition could improve DARCO FGD sorbent reactivity and mercury removal effectiveness from combustion flue gas. Tests were conducted on flue gases from Poplar River and Freedom coals using gaseous HCl and NaCl, respectively. Additional additives were also successfully tested and will be presented in the future.

The first round of testing was conducted on Poplar River coal, using HCl to promote sorbent reactivity. Before tests were conducted with HCl addition, the mercury concentration was measured at the outlet to evaluate the effectiveness of the DARCO FGD sorbent on mercury removal. The mercury concentration dropped from a baseline of approximately 23 µg/Nm^3, prior to sorbent injection, to 5 µg/Nm^3 with increasing sorbent injection rates up to 20 g/hr. At this injection rate, HCl was added at two locations under two separate tests, within the combustion zone and upstream of the ESP and carbon injection location. The flue gas temperature at the first location was approximately 930°C (1700°F) and at the second approximately 149°C (300°F), respectively. The addition of HCl at both locations did not appear to enhance sorbent reactivity and did not significantly improve the mercury removal effectiveness of the tested sorbents. More testing is needed before final conclusions can be drawn.

Additional testing was conducted on Freedom coal combustion flue gas using NaCl for sorbent enhancement. The impacts of NaCl addition with DARCO FGD were evaluated for three different particulate removal technologies: TOXECON™, *Advanced Hybrid*™ filter, and ESP. At baseline conditions, with no sorbent addition, mercury concentrations averaged 10 µg/Nm^3, as illustrated in Figure 9. The addition of sorbent at 20 g/hr resulted in TOXECON™ mercury removal of nearly 72%. The addition of NaCl to the coal feed resulted in an additional 2 µg/Nm^3 and is clearly illustrated in Figure 7. Similar trends were observed with both the *Advanced Hybrid*™ filter and ESP particulate control technologies.

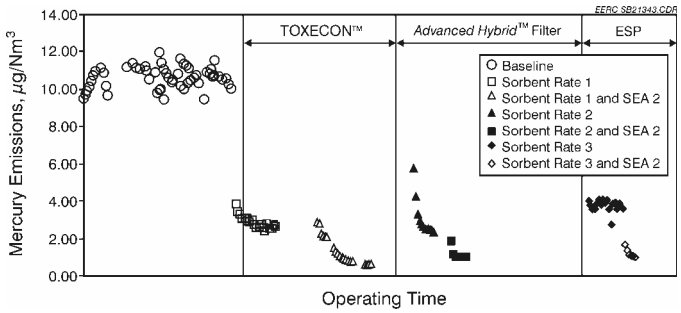

Figure 9. Impact of chlorine addition on mercury removal performance.

The preliminary results from these pilot-scale tests indicate that chlorine additives could be used to enhance mercury removal effectiveness of DARCO FGD, thereby reducing the amount of sorbent needed to achieve removal targets. Further, results suggest that the addition of NaCl to the coal would be more effective in enhancing mercury capture than HCl injection at the furnace outlet; however, differences in the test parameters (i.e., coal type, initial mercury concentration) make direct comparison of HCl and NaCl effectiveness difficult.

Using a chlorine additive has shown the potential to cut carbon rates by more than 50% for a given removal rate by using the additive at a rate below that of the carbon. Current costs for these additives range from $35 to $290/ton, depending on the additive. These values compare to current PAC costs of $1000/ton. Therefore, the $35/ton sorbent enhancement additive (SEA) has the potential to provide greater than 45% reduction in reagent costs (additive + PAC) for Hg control in systems where existing particulate control devices are used. Field testing is needed to better define performance, economics, and balance of plant impacts.

Effects of Temperature, Sorbent Size, Bag Material, and Air-to-Cloth Ratio. The impacts of temperature, sorbent size, filter material type, and air-to-cloth ratio were all presented previously and are summarized here for completeness. Temperature is a very important parameter for effective capture of mercury by PAC. Limited tests were conducted to evaluate temperature effects on sorbent performance in different configurations: ESP only, FF only, and ESP–FF. The flue gas temperature was raised from 149° to 204°C (300° to 400°F). Experimental data in Figures 6 and 7 and data presented previously show that mercury removal efficiencies were dramatically reduced, about 10%–15%, at the elevated flue gas temperature of 204°C (400°F) for all three configurations and for both Poplar River and Freedom coals, as compared to the 149°C (300°F) flue gas temperature.[29] An estimated 14.3% increase of sorbent usage is required to achieve collection efficiencies at a flue gas temperature of 204°C (400°F) similar to those at 149°C (300°F) for both the ESP-only and ESP–FF configurations. However, an estimated 70.7% increase in sorbent usage is needed for the FF configuration when the flue gas temperature was increased from 149° and 204°C (300° and 400°F). One partial explanation for the dramatic increase in carbon usage (70.7%) for the FF only at the flue gas temperature of 204°C (400°F) compared to 149°C (300°F) is the increased pulse rate at higher temperatures (increased A/C ratio) for the FF containing ash and carbon.

Sorbent size is another important parameter for mercury removal in flue gas, especially when mass transfer is an issue. Both the standard and fine MVD (of 20 and 5 µm, respectively) activated Luscar char were injected ahead of the ESP at different rates to examine the corresponding mercury removal efficiencies. The experimental data presented in Figures 3 and 4 show that a reduction in sorbent size did not consistently result in improved mercury capture. At injection rates of >8 lb/Macf, the fine activated Luscar char provides much better ESP mercury removal than that achieved with the more coarse carbon for the Poplar River coal. On the contrary, a reduction in carbon size did not greatly enhance ESP mercury capture in the Freedom coal flue gas. There may be several reasons that caused this inconsistency, such as adhesive agglomeration of the fine sorbent, inconsistency in fineness, poor dispersion of the fine sorbent into the flue gas, etc. More effort is needed before final conclusions can be drawn.

Two different bag materials, Ryton® and Gore-Tex®, were tested in the FF-only case at 149°C (300°F). Based on limited test data as shown in Figure 3, the differences in the FF material, Ryton® versus Gore-Tex®, did not significantly affect mercury capture efficiencies.

The FF was operated at A/C ratios of 6 and 12 with the TOXECON™ configuration to evaluate the impact that increased flows may have on mercury capture. Based on limited tests, the observed differences between 6 and 12 did not appear to be significant with regard to mercury capture. In other words, the increased velocity did not appear to impact mercury removal. Overall, the mercury removal was less when the A/C was 12, mainly due to increased pulsing frequency. This increased frequency translated to poorer utilization of the PAC at a given injection rate. Because the tests were rather short, long-term effects, such as increased blockage, erosion rates, etc., could not be evaluated.

Future Tests. The EERC currently has plans to perform additional tests of additives on bench- and pilot-scale systems, as well as tests at other North Dakota power plants. As part of the second phase of this ongoing project, the applicability of the conclusions from the Phase I bench- and pilot-scale investigations will be evaluated by performing similar PAC injection and flue gas and fly ash measurements at a utility host site equipped with a slipstream FF installed downstream of existing ESPs. Sorbent injection upstream of a pulse-jet-type FF (TOXECON™) is the mercury control technology that will be field-tested at the Poplar River Power Station, which is owned and operated by SaskPower. Activities planned for the field demonstration at the Poplar River Power Station include field-testing a slipstream-scaled version of the TOXECON™ technology, preparing the site and installing the appropriate technology hardware, evaluating sorbent impacts and performance, assessing technology impact on unit operations, assessing ash reuse and disposal impacts, and estimating costs to control mercury. The results from Phase II of the project should provide the lignite industry with a technology option that can be considered by other utilities as they develop their own mercury control strategies. In addition, Phase II data will provide insights into mercury control effectiveness for other coals and particulate control configurations.

Conclusions

The activated (800°C, 1472°F) Luscar char and DARCO FGD were selected for additional testing in a 580-MJ/hr (550,000-Btu/hr) pulverized coal-fired unit, based on sorbent screening results, costs, and consensus among project sponsors. The following variables that could potentially affect mercury emission control were tested: lignite coal source (Poplar River or Freedom Mine), control device type (ESP, FF, ESPBFF, and *Advanced Hybrid*™ filter), FF type (Gore-Tex® or Ryton®), sorbent type (steam-activated [800°C, 1472°F] Luscar char and DARCO FGD) and particle size (approximate MVDs of 20 and 5 µm), sorbent injection rate, SEA use, mercury speciation, and flue gas temperature in the pollution control device. Pilot-scale test results indicated the following:

- The relative proportions of Hg^0, Hg^{2+}, and Hg(p) in the Poplar River and Freedom coal combustion flue gases were very similar at approximately 85%, 15%, and <1%, respectively.
- The relative mercury removal efficiencies of the four control device technologies tested were 1) TOXECON™ and *Advanced Hybrid*™ filter, 2) FF, and 3) ESP, with the performance varying depending on coal and sorbent injection method.
- The pilot-scale results on lignite showed an increase in the required PAC needed for similar removals when compared to full-scale data for eastern bituminous coals.
- Most chlorine additives were effective in enhancing the mercury removal of activated carbon, thereby reducing the amount of sorbent needed to achieve a given level of mercury emission control. However, the use of HCl in the upper furnaces was not found to be effective.

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