

# **LATEST DEVELOPMENTS OF THE PLASMA-ENHANCED ELECTROSTATIC PRECIPITATOR FOR MERCURY REMOVAL IN COAL-FIRED BOILER FLUE GAS**

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## **ABSTRACT**

The plasma-enhanced electrostatic precipitator (PEESP) is an innovative coal-fired boiler flue gas mercury removal system that combines existing electrostatic precipitator technology with low-energy plasma technology. This new technology is energy efficient and produces very little secondary waste as compared to baseline absorbent mercury removal technologies. In April 2002, MSE Technology Applications, Inc. (MSE), and Croll-Reynolds Clean Air Technologies (CRCAT) were awarded a joint patent on the PEESP technology based on bench-scale experimental work conducted by MSE in February 2000. The original PEESP bench-scale qualitative experiments performed at MSE during the summer of 2000 indicated high mercury removal efficiencies while treating a simulated coal-fired boiler gas stream containing 250 micrograms per dry standard cubic meter ( $\mu\text{g}/\text{dscm}$ ) of elemental mercury. Steam plus oxygen were used as the reagent gas while applying a constant negative high-voltage source to the electrode in a single-element reactor.<sup>1</sup> The latest bench-scale testing results show a comparable removal efficiency, and further work will continue at larger process scale, with multiple PEESP electrodes, and a resulting more uniform and complete plasma treatment zone.

## **INTRODUCTION**

### **Environmental Issue**

Coal is our nation's most abundant source of fossil fuel. Mercury, a toxic metal regulated under the Resource Conservation and Recovery Act, exists naturally in coal. During combustion, mercury compounds are volatilized and become part of the boiler flue gas. In December 2000, under the authority of the 1990 Clean Air Act Amendments, the U.S. Environmental Protection Agency (EPA) announced plans to regulate mercury emissions from coal-fired power plants in an effort to reduce the health risks posed on humans and the

environment. The proposed regulatory limits will be promulgated in December 2003, with final compliance expected by December 2007. Anticipated regulations will require a 90% reduction on the current mercury emission rate.

A major problem in controlling mercury emissions is the low ratio of oxidized mercury to elemental mercury within the flue gas. Wet desulfurization systems used to abate acid gas emissions can efficiently abate water-soluble mercury compounds. However, the elemental mercury removal efficiency of these systems is temperature dependent and varies from less than 10% to more than 90%. Power plant studies have shown mercury emissions average a 60/40 split in elemental versus oxidized mercury and are largely dependent on the type of coal being burned in the boiler. Typically, mercury emissions generated by burning eastern bituminous coal are 60% to 80% in the oxidized state. Comparably, Powder River Basin coals generate mercury emissions that are only 10% to 20% in the oxidized state.<sup>2</sup>

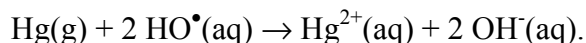
### **Technology Solution**

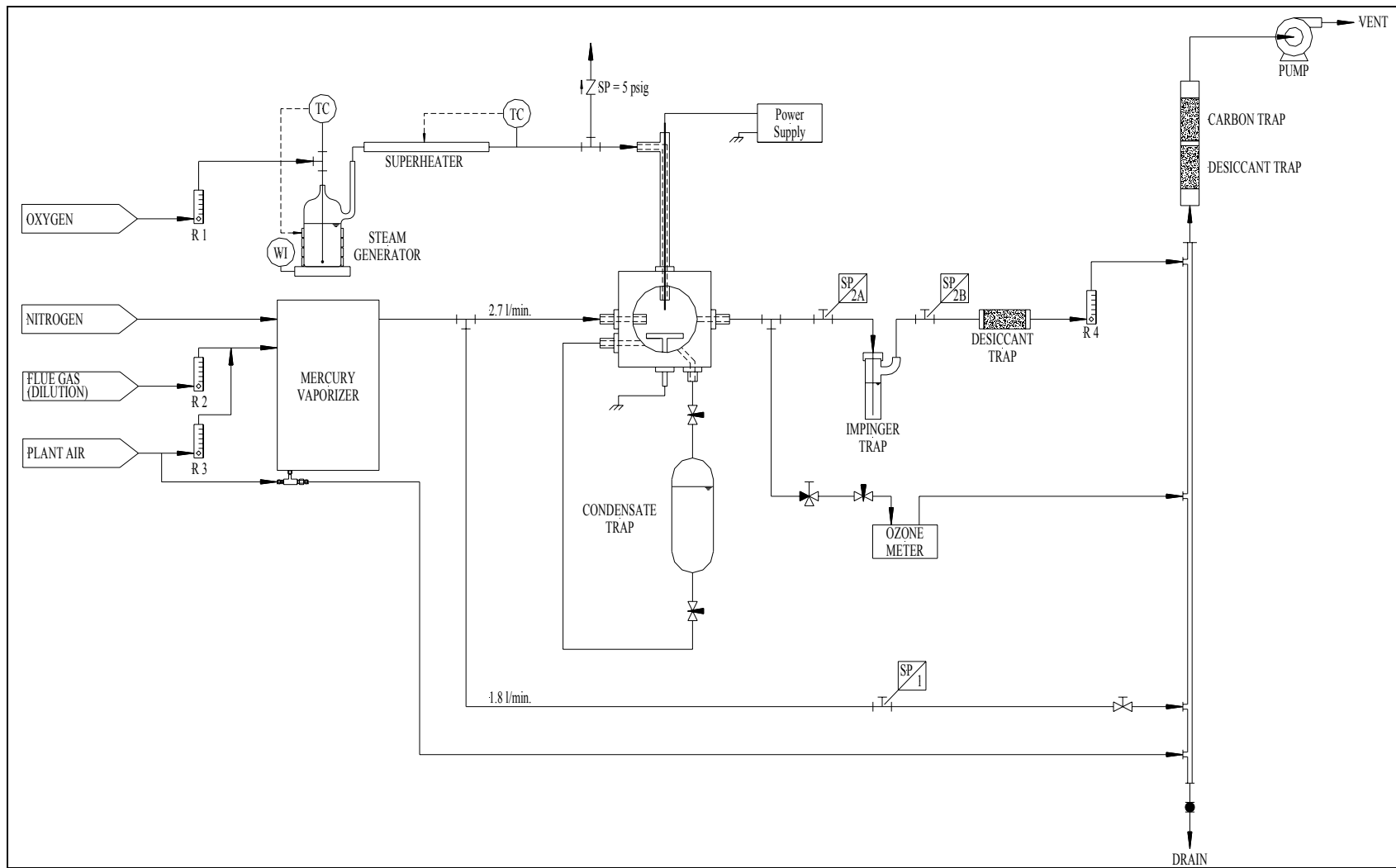
To address the problem of controlling mercury emissions, MSE and CRCAT teamed to develop an innovative technology that applies plasma physics to a wet electrostatic precipitator (WESP) creating a PEESP. The purpose of the latest bench-scale testing was to replicate, to the extent possible, and verify the original bench-scale qualitative test results. The original test results indicated that the PEESP removed more than 90% of elemental mercury in a simulated flue gas. Additional verification data was needed, as not all quality control procedures were followed and mercury measurements were extrapolated by indirect means.

An injector electrode-type corona discharge to standard air pollution control equipment is the basis of the PEESP. Conventional WESPs are quite effective at reducing fine particulate emissions, but are not capable of removing gaseous pollutants, such as elemental mercury vapor, oxides of nitrogen (NO<sub>x</sub>), and oxides of sulfur (SO<sub>x</sub>). However, by modifying the central electrode to inject a reagent gas through the corona discharge, a standard WESP (coaxial cylinders design) can be effective at removing elemental mercury vapor and, potentially, other trace contaminants.

### **EXPERIMENTAL**

Figure 1 is a schematic diagram of the PEESP bench-scale test bed.<sup>3</sup> The PEESP technology injects a reagent gas mixture, steam and oxygen, through a corona discharge needle that is attached to the central electrode within an electrostatic field. By adapting the electrode for reagent gas injection directly into the active zone, the area surrounding the sharp discharge point, hydroxyl radicals are immediately formed by the absorption of ozone in the corona, onto reagent droplets. Because these droplets are also electrically charged, they are attracted to the positively charged collecting electrode wall. As they travel from the discharge electrode to the collecting electrode, they adsorb and react with the elemental mercury vapor to form a mercuric ion according to the following simplified reaction:





**Fig. 1. PEESP Bench-Scale Reactor Test Bed Schematic Diagram.**

The aqueous mercuric ions, thus formed, can be scrubbed, or if operated within a WESP, collected on the electrode walls and removed with the liquid slurry.

Process streams supplying the test bed include oxygen, nitrogen, plant air, and a simulated flue gas composed of a mixture of nitrogen, carbon dioxide (CO<sub>2</sub>), carbon monoxide (CO), nitrogen oxide (NO), and sulfur dioxide (SO<sub>2</sub>). Because of long-term stability issues and safety concerns, oxygen cannot be added to the high-pressure cylinder containing the simulated flue gas. Therefore, to obtain a 7% oxygen concentration typical of a coal-fired generator flue gas, the simulated flue gas will be diluted with plant air at a ratio of 2:1. The final stream composition of the simulated flue gas is 13.3% CO<sub>2</sub>, 7% oxygen, 665 parts per million (ppm) NO, 665 ppm SO<sub>2</sub>, 7 ppm CO, and the balance nitrogen.

Elemental mercury vapor is added to the simulated flue gas using a vaporizing system designed and fabricated by MSE. The vaporizer uses a sonic orifice and dilution eductor to provide a reliable and consistent inlet mercury concentration to the PEESP bench-scale reactor. A hot cell within the mercury vaporizer contains a commercially available mercury permeation tube. By accurately controlling the temperature of the hot cell and permeation tube, a known and repeatable concentration of mercury vapor is eluted. The mercury vapor is removed from the hot cell using a 20-milliliter per minute (mL/min) nitrogen purge controlled by a sonic orifice connected to the suction side of a dilution eductor. The motive fluid used in the dilution eductor can be either air, simulated flue gas, or other gas stream. By adjusting the eductor inlet pressure to maintain a motive fluid flow rate of 4.5 liters per minute (L/min), the vacuum produced on the suction side of the eductor is approximately -5 inches mercury, which draws the mercury vapor and nitrogen purge gas from the hot cell into the eductor. The motive fluid and mercury-vapor mixture exiting the dilution eductor is then fed directly to the PEESP bench-scale reactor. To minimize the potential of amalgamating or condensing mercury vapor within the dilution eductor, the motive fluid is preheated to 150 °C and the dilution eductor is maintained at the system operating temperature. By varying the operating temperature of the hot cell, the mercury concentration in the vaporizer effluent gas can be varied from 9 to 1,000 µg/dscm. As a safety feature, the components comprising the mercury vaporizer are contained in a sealed enclosure. A second eductor is used to maintain a negative pressure within the enclosure. Any mercury fumes resulting from a leak are vented from the enclosure and removed from the work area in a controlled manner.

The simulated flue gas containing elemental mercury vapor enters the PEESP bench-scale reactor at a nominal flow rate of 2.5 standard liters per minute (slpm). Flow indication is monitored by a rotometer located downstream of the reactor. Excess flue gas is purged from the system through a vent line. To determine the mercury concentration entering the PEESP bench-scale reactor, the flue gas is sampled from the purge stream through Sampling Port 1 (SP-1).

Oxygen mixed with steam, generated by boiling deionized water, formulates the reagent gas that enhances the mercury removal efficiency of the PEESP technology. A superheater located downstream of the boiler dries the saturated gas mixture before injection through a high-voltage electrode. As the reagent gas passes through an active zone of the corona discharge, ozone along with hydroxyl radicals are produced. Within the PEESP reactor, these reactive species selectively oxidize the elemental mercury forming either water-soluble compounds or fine particulate that are absorbed by the condensing water droplets. In a full-

scale PEESP reactor, the negatively charged water droplets are expected to be attracted to the grounded electrode and removed from the system during normal wash-down cycles. However, in the bench-scale test bed, liquid condensate is drained from the reactor under gravity and is collected in the condensate trap.

Mercury compounds and fine particulate, which passes through the bench-scale reactor, are removed from the gas stream by bubbling the gas through an impinger trap containing deionized water. To determine the amount of mercury removed by the PEESP reactor, the flue gas exiting the impinger trap will be sampled [Sampling Port 2B (SP-2B)] and analyzed for mercury. Sampling Port 2A (SP-2A), installed between the PEESP bench-scale reactor outlet and the impinger trap, is periodically sampled to measure the mercury concentration in the reactor effluent gas. To determine a mass balance on the system, aqueous samples collected from the condensate trap and the impinger trap will also be analyzed to determine the total mercury concentration.

The process gases exiting the PEESP bench-scale test bed are collected in a venting manifold. A diaphragm pump is used to draw the gas through a desiccant trap to remove moisture and a carbon trap to remove mercury vapor before venting. The discharge of the diaphragm pump is vented to the atmosphere outside the building.

## **RESULTS AND DISCUSSION**

### **Test Objectives**

The primary test objective for the PEESP bench-scale tests was to repeat the proof-of-concept (POC) testing conducted in February 2000, to validate the test results. In this series of testing, special emphasis was placed on obtaining quality inlet and outlet concentration measurements to determine the mercury removal efficiency of the PEESP technology. In addition to sampling the outlet gas stream, the condensate and downstream impinger liquids were sampled and analyzed for mercury compounds. By measuring the mass of mercury entering the system versus the mass of mercury leaving the system, a mass balance on the bench-scale reactor was determined. When necessary, the walls of the reactor cell and electrodes were rinsed with an acidic solution to remove mercury residue that may have accumulated in the system. The rinsing solutions were collected and analyzed for mercury.

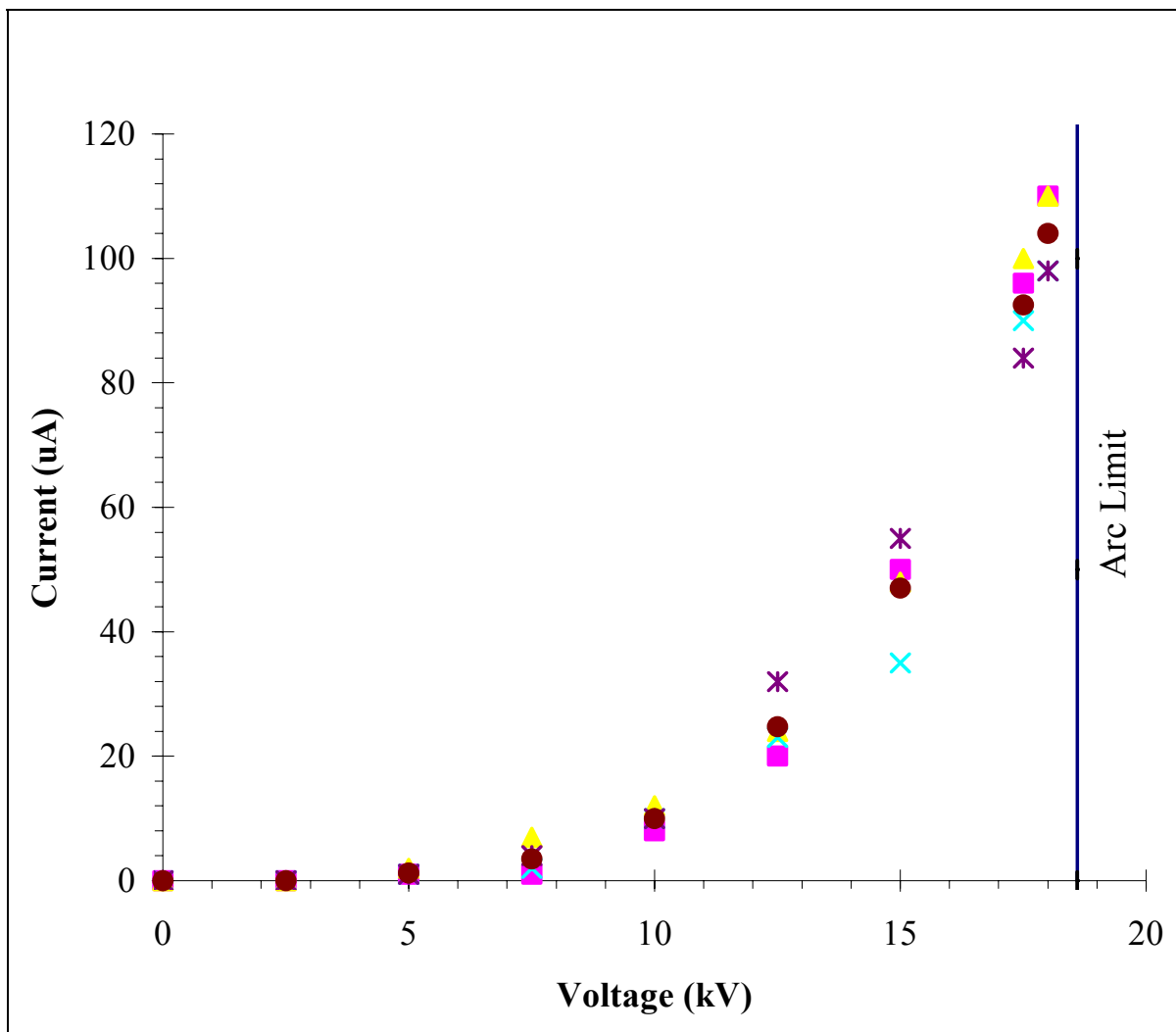
The second test objective was to determine the optimum operating conditions for the bench-scale reactor and the minimum acceptable mercury removal efficiency of the PEESP technology at minimum power consumption.

The final test objective was to obtain operational data to facilitate the PEESP technology scale-up to 5-standard cubic feet per minute (scfm), and then to a 50-scfm pilot-scale reactor that can be demonstrated in the MSE slip-loop test bed.

### **Current-Voltage Characterization of the PEESP Bench-Scale Reactor**

In this test sequence, the PEESP bench-scale reactor current was measured at various voltages to characterize the current-voltage (I-V) relationship of the bench-scale reactor. In addition to determining the onset of corona discharge and the upper voltage limit where

arcing occurs, I-V characteristics of the bench-scale reactor established the optimum power density to scale-up the PEESP reactor. Figure 2 represents a typical I-V characterization curve.



**Fig. 2: I-V Characterization Curve for the Bench-Scale Reactor Processing a Typical Flue Gas Stream while Injecting a Reagent Gas Comprised as a Steam-Oxygen Mixture.**

### Ozone Monitoring

The concept of the PEESP technology is based on the oxidation of elemental mercury vapor. Key reactants in the mercury oxidation reaction are ozone and hydroxyl radicals, which are formed as the oxygen-steam reagent gas passes through the corona discharge. Since the measurement of concentration of hydroxyl radicals is impractical, the ozone generation rate is measured as a function of input power with an online ozone analyzer as a gross indicator of hydroxyl radical production.

## **Calibration of the Mercury Vaporizer**

The mercury concentration in the vaporizer effluent gas can be varied from 15 to 475  $\mu\text{g/dscm}$  depending upon the hot cell operating temperature and the flow rate of dilution gas to the eductor. The mercury concentration in the vaporizer effluent gas was quantified at two selected operating conditions. Initial operating parameters were based on design calculations for the target concentrations of 200 and 35  $\mu\text{g/dscm}$ . Once the mercury vaporizer achieved thermal stability, samples were collected from the effluent stream to determine the actual mercury concentration.

## **Gas Sampling of Mercury**

Two mercury gas sampling methods were employed during testing. The Dräger™ tube method, developed by MSE, provided quantitative results without sending the sample to an offsite laboratory. As the vaporizer effluent gas was drawn through the Dräger™ tube at a sampling rate of 100 mL/min, the indicating layer changed from a pale yellow-gray color to a yellow-orange color. Once the indicating layer was fully discolored up to the printed marker, the sampling pump was turned off and the Dräger™ tube removed from the sampling port. By relating the volume of gas sampled to the concentration curve provided with the Dräger™ tubes, the mercury vapor concentration was quantified. However, because the quality assurance acceptance criteria for this method was not established, the results were treated as a gross approximation, estimated to be  $\pm 20\%$  of the actual concentration.

The second sampling method was the Sorbent Total Mercury (STM) method developed by Frontier Geosciences, Inc., of Portland, Oregon. This method was designed to provide defensible results that are comparable to EPA Method 101A. The accuracy of the STM method is estimated at  $\pm 2\%$ .<sup>4,5</sup>

## **Test Design**

For the test conditions specified in Table 1, the independent variables were applied electrode voltage, distance between the capillary electrode and the grounding electrode, process flow rates, inlet mercury concentration, composition of the flue gas, and composition of the reagent gas. Dependent variables were outlet mercury concentration, reactor current, and concentration of the reactive species (ozone and hydroxyl radicals) generated in the corona discharge.

Based on experience obtained from the POC tests, certain independent variables remained fixed throughout this series of bench-scale testing. For example, the distance between the capillary electrode and the configuration of the reactor and electrodes were changed for this series of tests. Likewise, the reagent gas, consisting of a mixture of oxygen and steam, was used in this series of tests.

**Table 1. Bench-scale test matrix (dry test conditions).**

Test Condition	1	2	3	4	5	6	7	8	9
Flue Gas Composition	Air			Simulated Flue Gas <sup>1</sup>			Air		
Flue Gas Flow Rate (R4) <sup>2</sup>	2.5 L/min			2.5 L/min			2.5 L/min		
Flue Gas Inlet Temperature	40 °C			40 °C			40 °C		
Reagent Gas Composition	Oxygen and Steam			Oxygen and Steam			Oxygen and Steam		
Oxygen Flow Rate (R1) <sup>2</sup>	5 mL/min			5 mL/min			5 mL/min		
Steam Flow Rate <sup>3</sup>	2.0 slpm			2.0 slpm			2.0 slpm		
Inlet Mercury Concentration	200 µg/dscm			200 µg/dscm			35 µg/dscm		
Electrode Voltage <sup>4</sup>	-10 kilovolts (kV)	-15 kV	-17.5 kV	-10 kV	-15 kV	-17.5 kV	-10 kV	-15 kV	-17.5 kV
Interelectrode Distance	2.5 centimeters (cm)			2.5 cm			2.5 cm		
Reactor Stabilizing Time <sup>5</sup>	1 hour (hr)			1 hr			1 hr		
<b>Sample Location</b>	<b>Number of Samples</b>								
Bench-Scale Reactor Inlet (SP-1) <sup>6,7</sup>	1	1	1	1	1	1	1	1	1
Bench-Scale Reactor Outlet (SP-2B <sup>7</sup> )	3	3	3	3	3	3	3	3	3
Impinger Trap Outlet (SP-2A <sup>7</sup> )	---	3	---	---	---	---	---	---	---
Ozone Concentration <sup>8</sup>	1	1	1	1	1	1	1	1	1

<sup>1</sup> Simulated flue gas for the bench-scale experiments was supplied from a compressed gas cylinder and blended with plant air to provide a stream composition of 13.3% CO<sub>2</sub>, 7% oxygen, 665 ppm NO, 665 ppm SO<sub>2</sub>, 7 ppm CO, and balance nitrogen (by volume).

<sup>2</sup> All process flow rates are given at standard conditions, which are 25 °C and 1 atmosphere.

<sup>3</sup> Steam flow to the reactor electrode was determined and quantified during testing by measuring the volume of liquid evaporated versus time. During POC testing, this parameter was not quantified.

<sup>4</sup> Electrode voltages selected for testing were based on the operating limits of the bench-scale reactor cell.

<sup>5</sup> Before sampling for mercury emissions, the bench-scale reactor was brought up to test conditions and allowed to stabilize for the specified time. After collecting the required samples, testing parameters were adjusted to the next test condition.

<sup>6</sup> Only one inlet sample per test condition was taken during the bench-scale test campaign.

<sup>7</sup> The STM method, developed by Frontier Geosciences, Inc., was used to measure the reactor inlet and outlet gaseous mercury concentrations. The STM method is a two-step procedure where elemental mercury and mercury compounds are adsorbed onto an iodine-impregnated activated carbon trap. The STM trap is then sent to an offsite laboratory where the activated carbon is digested and the solution analyzed for mercury using cold vapor-atomic fluorescence spectrometry.

<sup>8</sup> The concentration of ozone generated by the corona discharge was measured at the start of each test condition before injecting mercury vapor into the flue gas.

## Post Testing

Post-test activities included removing the mercury permeation tube from the vaporizer and recording the final weight. The bench-scale reactor was dismantled and cleaned with an acidic solution to remove residual mercury that accumulated during testing. The hole diameter in the negative electrode was measured and recorded. This information will be fed into the design requirements for the pilot-scale electrodes.

## Test Results

Table 2 shows the mercury concentration at the inlet and outlet of the reactor, at the impinger outlet, and at the ozone outlet.

Test Conditions 1 through 3 were baseline tests that demonstrated the mercury abatement of the PEESP bench-scale reactor without interference from other pollutants that may have been present in a typical flue gas. During this testing, a gas, consisting of air and mercury vapor at a concentration of 200  $\mu\text{g}/\text{dscm}$ , was supplied to the bench-scale reactor at a nominal flow rate of 1 slpm. The reagent gas, supplied to the negative electrode, was a mixture of oxygen and steam. The oxygen flow rate was controlled to 0.5 mL/min and, the steam flow rate was determined experimentally by measuring the evaporation rate of the deionized water contained in the boiler. Upon exiting the steam boiler, the oxygen-steam mixture was superheated to dry the steam.

In the initial POC tests, the interelectrode distance was set at 2.5 cm and remained fixed for this test series. The three electrode voltages selected for testing were based on the operating limitations of the bench-scale reactor. Selection of the minimum operating voltage was based on characterization experiments to determine the onset of the corona discharge.

Once the bench-scale reactor operating parameters were set to specified test conditions, the system was stabilized for 3 hr before sampling. To determine the mercury removal efficiency of the bench-scale reactor and the downstream impinger trap, one inlet sample and three outlet samples were collected for each test condition. During this test series, only one inlet sample per test condition was taken.

In addition to the gas samples, liquid samples from the condensate trap and the impinger trap were collected at the end of each test condition to determine the mercury mass balance. Before starting the mercury vaporizer, the ozone concentration of the gas steam exiting the bench-scale reactor was sampled and the results were compared to data collected in the previous test sequence.

In Test Conditions 4 through 6, the gas entering the bench-scale reactor was changed to a dry simulated flue gas. Except for the noted changes, all other operating conditions, including sampling requirements, remained unchanged. The results from this part of the test are still being evaluated and will not be included in this paper. The next tests will be performed on a 5-scfm unit and will include more realistic  $\text{NO}_x$ ,  $\text{SO}_x$ , and CO levels.

**Table 2. Test results for Conditions 1 through 9.**

<b>Test Condition</b>	<b>1</b>	<b>2</b>	<b>3</b>	<b>4</b>	<b>5</b>	<b>6</b>	<b>7</b>	<b>8</b>	<b>9</b>
<b>Sampling Results (Micrograms Mercury Per Dry Standard Cubic Meter)</b> <b>(Averages Shown for Multiple Samples)</b>									
Bench-Scale Reactor Inlet (SP-1)	251.2	212.8	241.1	TBD	TBD	TBD	43.0	76.2	43.6
Bench-Scale Reactor Outlet (SP-2A)	---	---	---	---	---	---	---	---	---
Impinger Trap Outlet (SP-2B)	168.6	90.8	46.4	TBD	TBD	TBD	33.2	38.0	16.4
Ozone Concentration	---	---	---	---	---	---	---	---	---
Mercury Removal Efficiency (%)	32.9	57.3	82.9	TBD	TBD	TBD	22.8	50.1	62.3

In Test Conditions 7 through 9, the mercury concentration in the dry flue gas entering the bench-scale reactor was fed at 35 µg/dscm. The lower mercury concentration was selected based on the past experimental work, where the highest reduction in mercury emissions was observed, and is more typical of actual flue gas mercury concentrations. Air was used as a process gas for this testing, as the bottled simulated flue gas was depleted during aborted testing due to system process leaks.

## **CONCLUSIONS**

### **Summary/Lessons Learned**

The recent PEESP bench-scale test data shows comparable results to the original POC test data, with air as the treatment gas, both at high and low concentrations of injected elemental mercury. The latest testing employed the STM mercury sampling method and extensive mercury mass balance was performed, and the results can be considered a reasonable verification of the original POC testing. Nearly 100% closure was achieved on mercury mass balance. Early in the experiment, dilution leaks were detected in the system and were corrected. An additional flow meter was added directly downstream of the reactor.

The original POC mercury data was based upon atomic absorption (AA) instrument readings. The AA instrument, normally intended to measure ozone, was calibrated qualitatively with a Jerome 431X mercury vapor analyzer. The accuracy of this calibration is unknown. A variance to the POC data noted was that the reactor current in the POC experiment was approximately one-half of that in the latest testing, approximately 17 kV. This could be attributed to the difference in reagent gas (steam) mass flow, thus the difference in reactor electrode and gas conductivities, and ultimate difference in mercury removal efficiencies. The effective treatment zone may have decreased with increased total current in the latest testing.

The latest results show that substantial elemental mercury is indeed being removed, and with optimal multiple reactor electrodes orientated to maximize homogeneous plasma density, the desired 90% or better mercury removal efficiency will be achievable. The latest data indicates that the mercury removal efficiency increases exponentially with increased reactor power. This attribute will be used in the design of the next PEESP reactor to be tested. The reactor will have a flow rate of 5 scfm.

### **Benefits**

The benefits are summarized as follows:

- The PEESP is easily retrofitted to existing dry/WESPs, keeping capital costs of mercury control low.
- The PEESP meets the anticipated EPA-required mercury reduction efficiencies.
- The PEESP does not require any additional real estate, as it fits within an existing dry/WESP.

- The PEESP does not increase pressure drop.
- The PEESP uses steam and oxygen as a reagent gas, removing any storage, handling, or disposal issues.
- The PEESP has no moving parts, minimizing maintenance.
- The PEESP extends the collection capabilities of a dry electrostatic precipitator/flue gas desulfurization (FGD) scrubber or a FGD/WESP system to include mercury removal as a collateral benefit.

### **Future Development**

As mentioned above, the next step in the PEESP development is to design, install, and test an up-scaled 5-scfm version at the MSE Test Facility. This work is now ongoing. The 5-scfm version will be tested stand alone with a dedicated blower, and a simulated flue gas will be mixed from gas bottles similar to the bench-scale tests. Controllability on process and reagent gases will be much easier than with the bench-scale experiment. The test results from the 5-scfm PEESP will be used to design, install, and test a 50-scfm PEESP test bed in MSE's existing simulated flue gas test facility. The flue gas source is a variable oxygen-enriched natural gas burner. The 50-scfm PEESP unit will, again, be tested to similar criteria as the previous smaller PEESP versions. The PEESP upscale designs, single tubes with multiple electrodes, will be aided by test data from the replicate bench-scale testing.

After completion of testing at MSE, a 5,000-scfm version will be designed, installed, and tested in an existing CRCAT experimental WESP at the First Energy Mansfield Plant in Shippingport, Pennsylvania.

### **ACKNOWLEDGMENT**

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