MERCURY REMOVAL IN A MULTI-POLLUTANT CONTROL TECHNOLOGY FOR UTILITY BOILERS

Christopher R. McLarnon, Ph.D.
Powerspan Corp.
PO Box 219
New Durham, NH 03855

ABSTRACT

Powerspan is conducting pilot scale testing focused on mercury removal in a multi-pollutant control technology at FirstEnergy's R.E. Burger Plant under a cooperative agreement with the U.S. Department of Energy. The technology, Electro-Catalytic Oxidation (ECO^{TM}), simultaneously removes mercury and other heavy metals, fine particulate matter ($PM_{2.5}$), nitrogen oxides (NO_x), and sulfur dioxide (SO_2) from the exhaust gas of coal-fired power plants.

ECO uses a dielectric barrier discharge (DBD) to convert elemental mercury (Hg) to mercuric oxide (HgO). A scrubber and wet electrostatic precipitator collect the resulting oxidized mercury along with fine particles and the aerosols created in the ECO process.

Testing under the DOE agreement focuses on optimization of mercury removal in the ECO process while maintaining high removal levels of NO_x, SO_2, and particulate matter. The test program includes speciation of mercury entering the ECO process, a determination of the fate of captured mercury, and testing of mercury removal from the process byproduct stream.

The presentation will cover basic principles of ECO technology and the process improvements made over the last two years. Up to date performance results from the pilot test program, including NO_x, SO_2, and mercury removal efficiencies, will be reported. In addition, ECO cost estimates and plans for a commercial demonstration unit will be discussed.

INTRODUCTION

Release of mercury and other air toxic compounds to the environment has gained increasing attention as studies link these compounds to adverse effects on human health. It is expected that regulations controlling Hg emissions will be promulgated in 2004 with reductions required in 2007 to 2008. New regulations, along with rules already in place or anticipated for control of nitrogen oxides, sulfur dioxide and particulate matter, make clear the need for a multi-pollutant control technology—one that keeps coal-fired power generation, the single largest source of electrical energy in the U.S., a viable and economical component of the generating portfolio.

Powerspan Corp. (New Durham, NH) continues pilot testing of a multi-pollutant emissions control technology designed to remove nitrogen oxides (NO_x) and sulfur dioxide (SO_2) from the exhaust of coal-fired utility boilers. In addition to NO_x and SO_2 emissions control, the patented technology, named Electro-Catalytic Oxidation (ECO), substantially reduces emissions of mercury (Hg) and other air toxic compounds such as arsenic and lead, acid gases such as hydrochloric acid (HCl), and fine particulate matter ($PM_{2.5}$). The pilot unit is installed at FirstEnergy's R. E. Burger Plant and was modified in early 2002 to incorporate an ammonia scrubber and redesigned wet electrostatic precipitator fields. The pilot treats 1500 to 3000 scfm of flue gas drawn from the Burger Plant's Unit 4 boiler.

In commercial operation the ECO process is to be installed downstream of a power plant's existing ESP or fabric filter as is shown in Figure 1. It treats flue gas in three steps to achieve multi-pollutant removal. In the first process step a barrier discharge reactor oxidizes gaseous pollutants to higher oxides. For example, nitric oxide is oxidized to nitrogen dioxide and nitric acid, a small portion of the sulfur dioxide is converted to sulfuric acid, and mercury is oxidized to mercuric oxide. Following the barrier discharge reactor is the ammonia scrubber, which removes unconverted sulfur dioxide and nitrogen dioxide produced in the barrier discharge. A wet electrostatic precipitator (WESP) follows the scrubber. It, along with the scrubber, captures acid aerosols produced by the discharge reactor, fine particulate matter and oxidized mercury. The WESP also captures aerosols generated in the ammonia scrubber.

Liquid effluent produced by the ammonia scrubber contains dissolved ammonium sulfate and nitrate salts, along with Hg and captured particulate matter. It is sent to a byproduct recovery system which includes filtration to remove ash and activated carbon adsorption for Hg removal. The treated byproduct stream, free of Hg and ash, can be processed to form ammonium sulfate/nitrate (ASN) fertilizer in crystal, granular or liquid form.

Barrier Discharge Reactor

Oxidation of gaseous pollutants (Hg, NO_x, SO_2) in a barrier discharge reactor is the starting point of the ECO process chemistry. It is accomplished through generation of a non-thermal plasma in which energetic electrons (~5 eV) create radicals leading to pollutant oxidation. The electron energies formed in the barrier discharge are ideal for generating gas-phase radicals, such as hydroxyl (OH) and atomic oxygen (O). Formation occurs through collision of electrons with water and oxygen molecules present in the flue gas.

$$O_2 + e^- \rightarrow O + O + e^- \tag{1}$$

$$H_2O + e^- \rightarrow OH + H + e^- \tag{2}$$

$$O + H_2O \rightarrow 2OH \tag{3}$$

In a flue gas stream, these radicals oxidize NO_x, SO_2, and Hg to form NO_2, nitric acid (HNO_3), sulfuric acid (H_2SO_4), and mercuric oxide (HgO), respectively, as described in Reference [1]. Some reaction paths are presented below:

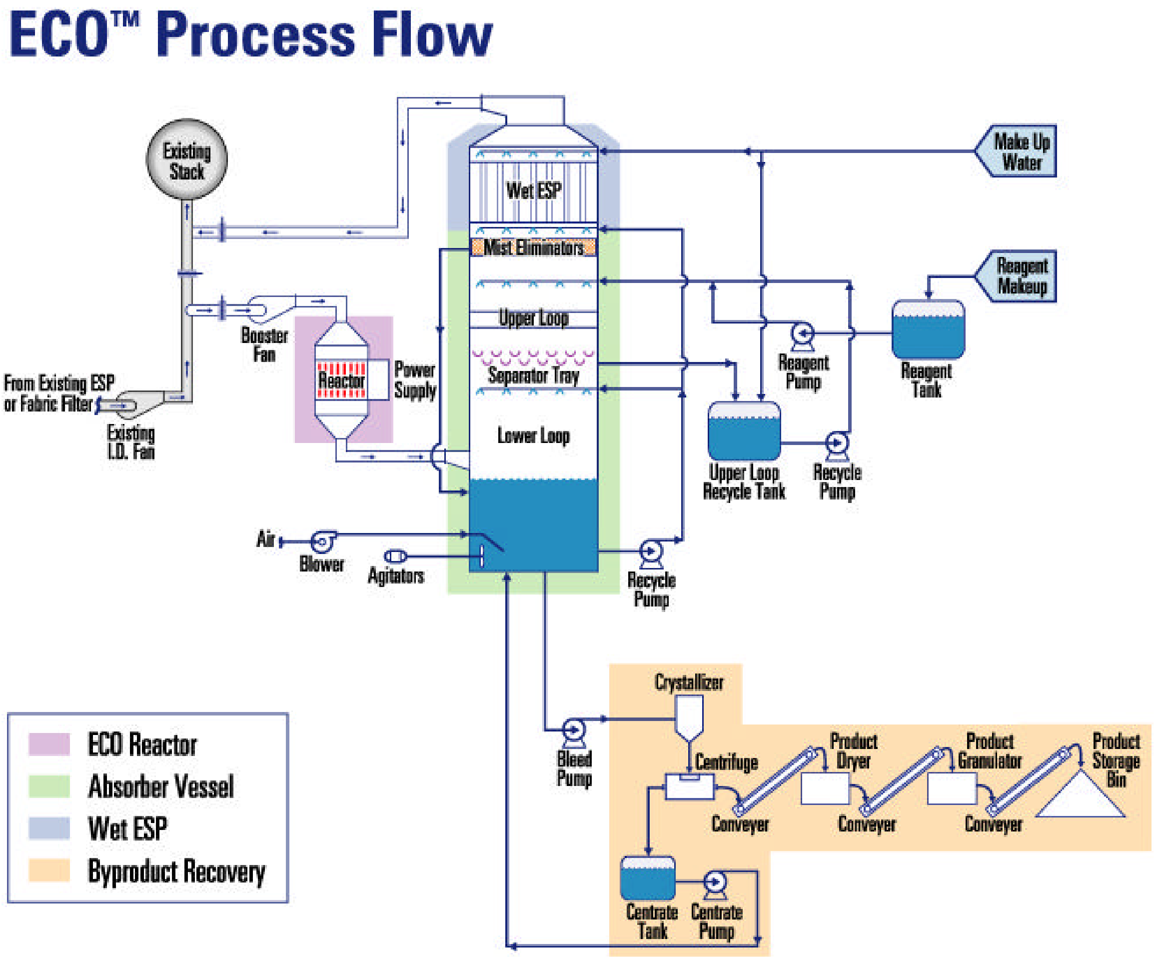

Figure 1: ECO Process Flow Diagram.

$$OH + SO_2 \rightarrow HSO_3 \tag{4}$$

$$HSO_3 + O_2 \rightarrow HO_2 + SO_3 \tag{5}$$

$$SO_3 + H_2O \rightarrow H_2SO_4 \tag{6}$$

$$NO + HO_2 \rightarrow NO_2 + OH \tag{7}$$

$$O + NO \rightarrow NO_2 \tag{8}$$

$$OH + NO_2 \rightarrow HNO_3 \tag{9}$$

$$O + Hg \rightarrow HgO \tag{10}$$

All of the above reactions can be made to occur at low temperature (150-350^{o}F). Additional reactions leading to the production of acids, such as oxidation of SO_2 by O atoms, are also initiated by the discharge. A detailed description of DBD processing of NO_x in a flue gas stream can be found in References [2] through [4]. Combined DBD processing of NO_x and SO_2 is discussed in References [5] through [8]. NO_x removal in the ECO process depends on conversion of NO to NO_2 and/or to HNO_3. Although NO is difficult to remove from a gas stream, once it is converted to NO_2 and HNO_3 by the DBD reactor, ECO's absorber and wet electrostatic precipitator can be operated to capture both.

Efficient oxidation of elemental mercury (reaction 10) in a non-thermal plasma has been demonstrated in Powerspan's laboratories as well as by others [9]. Oxidation of 80 to 90% of the elemental mercury has been reported at inlet concentrations representative of coal-fired flue gas streams with an energy input of 20 watts per standard cubic feet per minute of flue gas.

Ammonia Scrubber

An ammonia scrubber has been incorporated into the ECO process in order to achieve high removal efficiencies of NO_x and SO_2 while reducing the power requirements of the barrier discharge reactor. Flue gas entering the scrubber is first quenched to saturation. The gas then enters a scrubbing stage, which captures both SO_2 and NO_2. Ammonia is utilized in order to scrub SO_2 at a high rate and to produce compounds capable of efficiently reducing NO_2 to nitrogen. Ammonia will also neutralize the acids (HNO_3 and H_2SO_4) created in the barrier discharge reactor and produce ammonium sulfate and nitrate, a useful fertilizer byproduct, from the acids and scrubbed SO_2. The synergy between SO_2 scrubbing and capture of NO_2 produced from NO_x in the barrier discharge reactor results in a system with the ability to achieve high levels of NO_x and SO_2 removal.

The scrubbing chemistry starts with ammonia in aqueous solutions producing ammonium and hydroxyl (base) ions as shown in reaction (13). Reactions (14) through (16) show the absorption of SO_2 into aqueous solution producing sulfurous acid (H_2SO_3), bisulfite (HSO_3^-), sulfite (SO_3^{2-}) and hydronium (H_3O^+) ions.

$$NH_3 + H_2O \leftrightarrow NH_4^+ + OH^- \qquad (13)$$

$$SO_2 + H_2O \leftrightarrow H_2SO_3 \qquad (14)$$

$$H_2SO_3 + H_2O \leftrightarrow HSO_3^- + H_3O^+ \qquad (15)$$

$$HSO_3^- + H_2O \leftrightarrow SO_3^{2-} + H_3O^+ \qquad (16)$$

Hydroxyl (OH^-) and hydronium ions react to maintain a neutral pH, driving the SO_2 absorption reactions (14) through (16) to produce SO_3^{2-}.

$$OH^- + H_3O^+ \leftrightarrow H_2O \qquad (17)$$

Combining reactions (13) through (16) yields the overall SO_2 scrubbing reaction (18).

$$2NH_3 + SO_2 + H_2O \rightarrow 2NH_4^+ + SO_3^{2-} \qquad (18)$$

Sulfite produced by equation (18) reacts with NO_2 through oxidation-reduction reaction (19), reducing NO_2 to nitrogen while oxidizing sulfite to sulfate:

$$2SO_3^{2-} + NO_2 \rightarrow SO_4^{2-} + \tfrac{1}{2}N_2 \qquad (19)$$

From equations (13) through (19) the overall reactions for SO_2 and NO_2 scrubbing with ammonia can be written as:

$$2NH_3 + SO_2 + H_2O \rightarrow (NH_4)_2SO_3 \qquad (20)$$

$$2(NH_4)_2SO_3 + NO_2 \rightarrow 2(NH_4)_2SO_4 + ½N_2 \qquad (21)$$

The ratio of SO_3^{2-} to HSO_3^{-} in solution (16) is determined by solution pH. Both compounds are reported to scrub NO_2 but the rate of reaction between NO_2 and SO_3^{2-} is reported to be forty times faster than that of NO_2 and HSO_3^{-} [10]. Therefore, pH control of the scrubbing solution through addition of ammonia is essential to ensure that an adequate SO_3^{2-} concentration is maintained for high NO_2 scrubbing rates.

From reactions (20) and (21), it can be seen that a minimum of two moles of SO_2 are required for each mole of NO_2 reduced to N_2. However, the sulfite needed for NO_2 reduction can also be consumed by O_2 present in the flue gas, effectively increasing the ratio of SO_2 to NO_2 required for NO_2 scrubbing:

$$2\,SO_3^{2-} + O_2 \rightarrow 2SO_4^{2-} \qquad (22)$$

Oxidation of sulfite to sulfate results in a minimum ratio of SO_2 to NO_x of 3 in order to maintain a sulfite concentration adequate to scrub NO_2 to acceptable levels.

Compounds produced in the scrubbing of SO_2 and NO_2 can result in the reduction of oxidized mercury (Hg^{2+}) to elemental mercury (Hg^0), reversing the oxidation process accomplished by the barrier discharge reactor. Understanding the extent to which the reduction reactions occur and, if necessary, developing a means to retard the reaction, is a focus of the laboratory investigation and the current test program. Solutions to the problem of mercury reduction, include altering conditions of the scrubber chemistry to retard the rate of reduction and treatment of the scrubber solution to remove mercury. The treatment process must keep the steady state mercury concentration low enough to substantially diminish the rate of mercury reduction.

Wet Electrostatic Precipitator

Flue gas exiting the ammonia scrubber may contain oxidized mercury and fine particulate matter. It will also contain aerosols generated in the barrier discharge and ammonia scrubbing process steps (NH_4HSO_4, NH_4NO_3, NH_4Cl). These materials are captured in a wet electrostatic precipitator (WESP) and returned to the scrubbing solution. A WESP is efficient at collection of aerosols and fine particulate matter since there is no mechanism to cause re-entrainment. In a WESP, liquid flows down the collecting plate to remove captured materials from the plate. The advantages of WESPs include: a) water flow prevents particle re-entrainment, which would limit collection efficiency; b) the water layer does not limit corona current; and c) the improved collection characteristics permit high gas velocities, limiting the equipment size required for collection.

Byproduct Processing

Ammonium sulfate and nitrate created in the ECO process can be treated and used as a commercial fertilizer. Solids in the scrubber bleed stream, consisting of ash and insoluble metal compounds, are removed by filtration. The stream is then pumped through an activated carbon adsorption bed. The activated carbon used in ECO is produced by Nucon International (Columbus, OH) and sold under the name of Mersorb®. It is impregnated with sulfur, which reacts with mercury compounds and is strongly adsorbed to the bed.

Liquid substantially free of mercury and ash is sent to a crystallizer in which moisture is driven off to produce crystals of well-defined size, strength, and composition. The crystals may be usable as fertilizer in the form produced by the crystallizer, or may be processed to further reduce the moisture of the crystals or to agglomerate the crystals into granules. Spent mercury-laden activated carbon is replaced and disposed of as a hazardous waste. It is estimated that the variable cost of mercury removal with activated carbon is $733 per pound of mercury, including the media and disposal.

EXPERIMENTAL

ECO Pilot

The ECO pilot system, constructed at FirstEnergy's R.E. Burger Plant, has been in operation for four years to support development of the technology. It was modified at the beginning of 2002 to incorporate an ammonia scrubber and its associated liquid handling equipment. A decision was made prior to the modification to keep the unit in a horizontal configuration, as opposed to the vertical configuration to be commercially deployed. The decision was based on a desire to minimize the time required to complete modifications and an investigation of the process chemistry.

The pilot, shown in Figure 2, draws a slipstream of gas from the Burger Plant's Unit 4, upstream of the unit's ESP. The gas is returned to the unit at the ESP inlet. Gas flow into the pilot, at a rate of 1500 to 3000 scfm (standard cubic feet per minute), passes through a small cyclone separator and two dry ESP fields, each four feet in length. These two units in series are to reduce the ash content of the flue gas to a level similar to that expected after a plant's ESP or fabric filter in a full-scale installation.

Upon exiting the dry ESP, flue gas enters a multi-tube, coaxial cylinder barrier discharge reactor. High voltage applied to the center electrodes of the discharge reactor creates the non-thermal plasma that forms radicals leading to oxidation of gaseous pollutants. The ECO reactor is capable of delivering up to 100 KW of discharge energy to the gas.

The ammonia scrubber follows the barrier discharge reactor and is in an absorber vessel consisting of three packed sections in a cross flow configuration. The first section cools and saturates the gas. It is four feet deep in the direction of gas flow. Next is a six-foot scrubbing section to remove SO_2

and NO_2. Following the scrubbing section is a six-inch packed section that absorbs gaseous ammonia exiting the scrubbing section.

Gas exiting the absorber vessel enters a horizontal, sectionalized, three-field WESP. Each field is thirty inches deep. The collecting plates are washed periodically, and the liquid effluent is sent to the ammonia scrubber section.

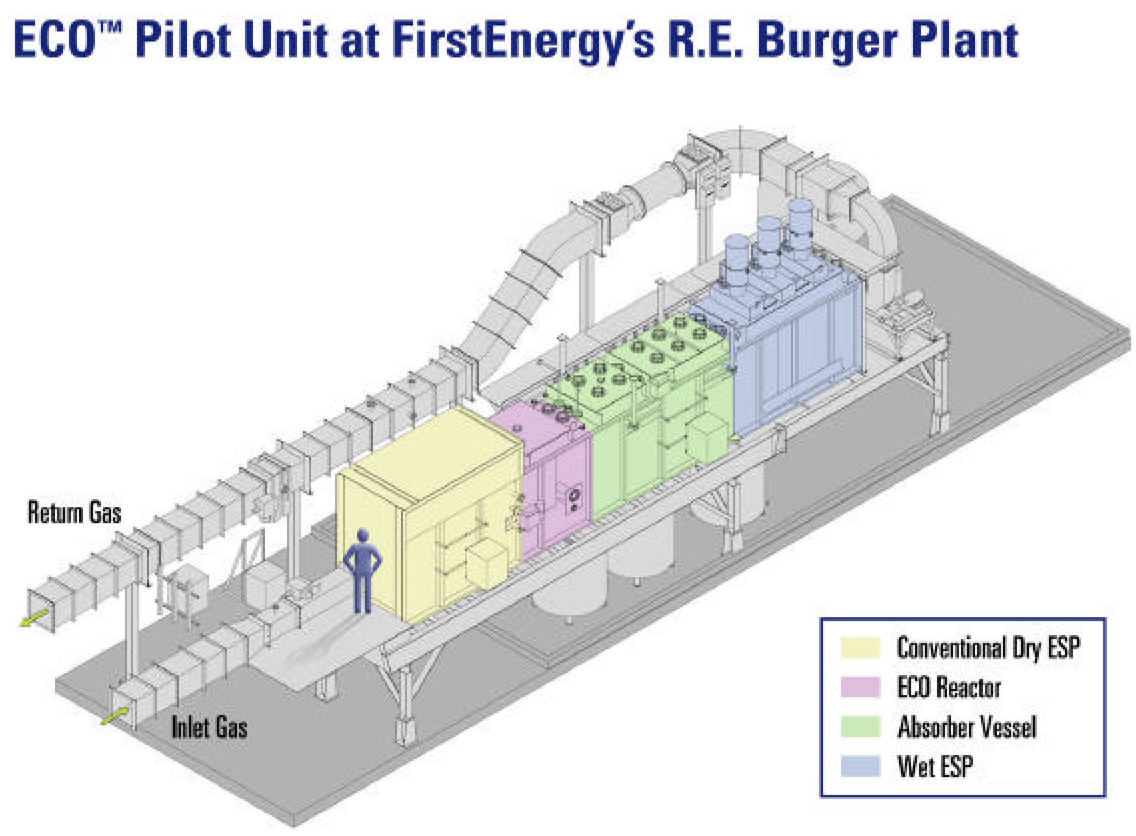

Figure 2: ECO Pilot Isometric Drawing.

A seven-man crew operates the Burger pilot on a three shifts a day basis. Continuous emissions monitoring is accomplished at the flue gas inlet to and exit from the pilot. The system measures the concentration of SO_2, NO_x, O_2, H_2O, CO_2, CO and NH_3.   Outlet flue gas flow and opacity are also measured continuously. Temperatures, flow rates, pH of all liquid flow streams, and pressure drop across all process units are also measured. In all, over 175 parameters are continuously recorded by automatic data logging equipment.

Mercury concentration in the pilot flue gas is measured using PS Analytical's Sir Gallahad semi-continuous mercury CEM systems. Two systems are installed to provide simultaneous, near real time measurement at the pilot inlet and outlet as well as at other selected locations. The systems report elemental and total gas phase mercury concentrations. Since their installation, Powerspan has been working with the supplier of the instrument (PS Analytical) and the sample conditioning system (Baldwin Environmental) to ensure that reliable measurements are made. To date the systems have not run reliably enough to provide reportable results.

An independent stack-testing agency, Air Compliance Testing Inc. (Cleveland, OH), measured particulate bound, elemental, and oxidized mercury in May 2002 [11], using the Ontario-Hydro method. In addition, Air Compliance Testing measured the concentrations of metals (including Hg), sulfur trioxide (SO_3), HCl and HF on the pilot inlet and outlet flue gas streams in June 1999 [12] and again in May 2000 [13]. The testing consisted of EPA methods 1, 2, 3, 4, 26, 29 and the Controlled Condensate Method for determination of sulfuric acid mist. Air Compliance Testing also made particle size distribution measurements in May 2000 [14]. EPA Methods 1, 3, 3A, 4 and 17 (Modified) were used for this testing.

Configuration of the sampling ports in the pilot's gas ducts precluded Air Compliance Testing from simultaneously making measurements at both the inlet and outlet. Instead, sampling was alternated between the inlet and outlet ducts while operation of the pilot was unchanged and "pseudo" removal efficiency calculated. Removal efficiencies were based on average values from at least two sample runs conducted at each sampling location.

Mercury Removal from Scrubbing Liquids

A system for treating synthetic and ECO Pilot effluent liquids has been constructed in Powerspan's New Durham laboratories. The system utilizes Mersorb® Mercury Adsorbent, a sulfur-impregnated activated carbon produced by NUCON International (Columbus, OH), to remove mercury in solution. The system consists of two Mersorb adsorbent beds in series and 0.5 micron filters on the inlet to and outlet of the adsorption beds for particle capture. It can process liquids at flowrates up to 0.1 gpm. A system has also been constructed at the ECO pilot to treat liquid effluent on a continuous basis. It will be operational in September 2002.

RESULTS AND DISCUSSION

Testing of the modified ECO process including the ammonia scrubber began in March 2002 and is continuing. During operation, boiler loads change frequently as Unit 4 of the R.E. Burger Plant is often run at low load (<100 MW) during the evening and cycles as high as 156 MW during peak load. This results in large variation in the flue gas entering the pilot unit. There is also variation in the plant's fuel; for example, sulfur content can vary from about 2% to as high as 4%.

Mercury Removal

The pilot test program requires periodic sampling using the Ontario-Hydro test method. Air Compliance Testing conducted the first test event in May 2002. The testing consisted of three sample runs each on the inlet and outlet flue gas streams. Two of the three sets of sample runs had a sample duration in each location of 4 hours while sampling for the remaining set of runs lasted 3 hours in each location. Averaged results for three fractions and the total Hg concentration measured in each location are shown in Table 1. It can be seen from the table that the overall Hg removal was measured to be 88%.

Table 1 also shows that the average elemental Hg concentration increased from 0.16 µg/dscm (micrograms per dry standard cubic meter) to 0.75 µg/dscm. The increase is likely due to reaction of oxidized mercury with reducing compounds (e.g. SO_3^{2-}) present in the scrubbing solution. Laboratory testing as well as testing at the ECO pilot is in progress to understand the conditions that result in production of elemental mercury from the oxidized form and to alter the process chemistry to eliminate the reduction reaction. To support the test program a system for injecting elemental mercury into the inlet flue gas stream has been installed at the ECO pilot. It is intended to increase the elemental Hg concentration to as high as 20 µg/dscm. Testing of the addition system has begun but verification of its performance is awaiting reliable near real time Hg measurement. Once the system and Hg measurement instruments are functioning reliably a second round of Ontario-Hydro testing, along with Method 29 testing, will be conducted.

Table 1: Ontario-Hydro Measurement Results.

Hg Fraction	**ECO Inlet**	**ECO Outlet**	**Removal**
Particle Bound Hg	0.62	0.016	97.4 %
Oxidized Hg	5.81	0.022	99.6 %
Elemental Hg	0.16	0.75	
Total Hg	6.59	0.79	88.0 %

The total Hg removal levels achieved with the ECO process, after addition of the ammonia scrubber, are consistent with the removal levels measured at the ECO pilot prior to the scrubber installation. Method 29 measurements were made in 2000 by Air Compliance Testing during which the total mercury removal level was measured to be 81.6% [12]. During those same tests the removal of arsenic, barium, chromium, copper, lead, manganese, nickel and phosphorous exceeded 99%. Air Compliance Testing also made particulate matter measurements in 2000 [13]. The results showed that 99.6% of the total particulate matter was captured in the ECO process. The testing also measured 96.7% capture of particles less than 3 microns in size.

NO_x and SO_2 Removal

Presented in Figures 3 and 4 are typical pilot data showing NO_x and SO_2 concentrations and removal levels measured over a 24-hour operating period. Barrier discharge reactor power throughout the test averaged 20 watts per standard cubic foot per minute of flue gas. Figure 3 shows that NO_x removal over the course of the day averaged 90% with an inlet level of 250 ppm. The periodic spikes seen in the outlet NO_x concentration and NO_x removal traces are due to a process in which the barrier discharge reactor is momentarily de-energized and a sonic horn sounded. The process is employed to keep the barrier discharge reactor clear of ash. The rapid rise in outlet NO_x concentration when de-energized clearly shows the barrier discharge reactor's effect on NO_x removal.

Two other aspects of the data presented in Figure 3 are of note. The high inlet and outlet NO_x concentrations shown at 4:00 am are due to instrument calibration. The figure also shows that NO_x removal increased from approximately 62% to 90% at 1:00 am. This increase is due to a drop in inlet NO_x concentration from 470 ppm to 250 ppm. The barrier discharge reactor removes a relatively fixed mass of NO_x for a given set of operating conditions and reactor power. Therefore, higher removal percentages are achieved at lower inlet NO_x concentrations.

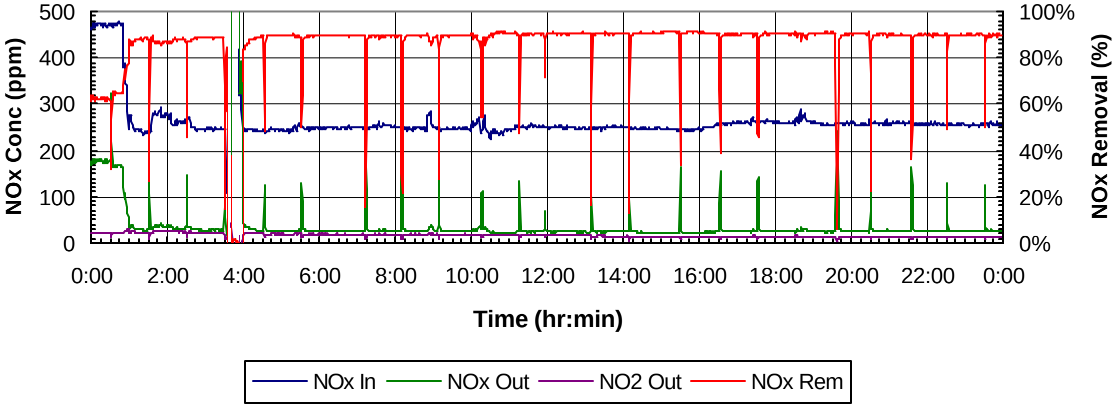

Figure 3: NO_x Concentrations and Removal at ECO Pilot.

Figure 4 presents the inlet and outlet SO_2 concentrations and the removal percentage achieved over the same 24-hour period as that shown in Figure 3. The SO_2 removal was 97% over the course of the day with an average inlet concentration of 1950 ppm. The decrease in removal to 90% that occurred at approximately 19:00 was due to securing scrubbing spray in the packed sections for a short duration to accomplish minor maintenance.

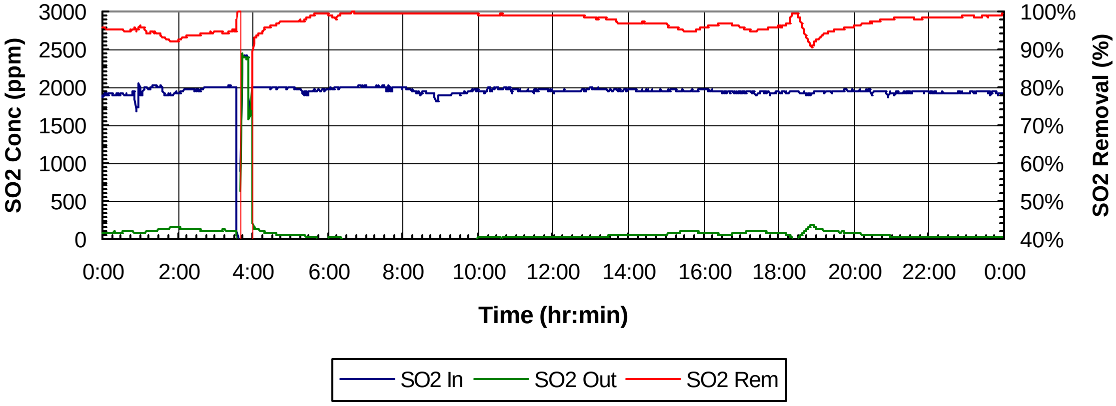

Figure 4: SO_2 Concentrations and Removal at ECO Pilot.

Conclusions

The pilot and laboratory test results to date have shown ECO to be a multi-pollutant control process capable of achieving high levels of removal for Hg, NO_x, SO_2 and particulate matter. An economic analysis of the process has shown the installed cost of ECO to be approximately $150/KW, exclusive of balance of plant modifications. The operating costs associated with an ECO system are estimated to be 1.5mils/kWh. It includes the capture of mercury, estimated to be $733 per pound of mercury removed for adsorbent and its disposal.

Continued pilot testing is needed and will continue under the DOE Cooperative Agreement to fully demonstrate ECO's ability to capture high levels of elemental mercury, while maintaining high SO_2, NO_x and particulate matter removal, from flue gas streams.

Pilot testing of the ECO process and its economics point out several advantages to the ECO system that make it attractive for pollutant control in coal combustion produced flue gas streams. They include:

1. Performance of the ECO technology at the pilot scale shows its ability to remove 90% of NO_x based on 0.4 lb/MMBtu inlet NO_x, 97% of SO_2, 80% of Hg, and 99.9% of fine particles that are less than 10 µm in diameter.
2. The ECO system significantly reduces emissions of NO_x, SO_2, $PM_{2.5}$, and Hg in an integrated system, thereby minimizing the need for additional capital investment in other pollution control equipment. More specifically, a combination of flue gas desulfurization for SO_2, selective catalytic reduction for NO_x, and activated carbon injection for Hg would be required to obtain comparable performance.
3. Capital cost for the ECO system is estimated to be about $150/kW (excluding balance of plant modifications) with operating costs at approximately 1.5 mils/kWh.
4. The ECO system produces a commercially salable, ammonium sulfate nitrate fertilizer byproduct that reduces operating costs and avoids landfill disposal of waste.
5. The ECO system minimizes the amount of plant retrofit required for installation, since it provides multi-pollutant control with a single installation.
6. The ECO equipment has a much smaller footprint than conventional control technologies, facilitating its installation on space-constrained sites that are typical of the existing coal-fired electricity generating fleet.

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