

“Airborne Process™” - Advancement of Multi-pollutant Emissions Control Technology and By-product Utilization

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ABSTRACT

Airborne has undertaken an extensive development program for the ultra-high removal of emissions from coal fueled generating stations called the Airborne Process™. Airborne completed a 5 MW demonstration near Ghent, Kentucky in 2003 (Airborne Facility) and has completed additional advanced development for achieving high capture of NO_x and Mercury (Hg) at the Energy and Environmental Research Center (EERC) of North Dakota University. The technology described herein combines the use of dry sodium bicarbonate injection, coupled with enhanced wet sodium scrubbing and employing advanced staging with oxidants to control SO_x, NO_x, and Hg emissions. This paper details the results from the development of the Airborne Process™ including recent results from testing of oxidant solutions that removed both NO_x and Hg at levels near 100% in both bench and pilot scale tests. In addition, the paper also describes the future commercial demonstration of the Airborne Process™ at Peabody's Mustang Energy project, a new 300MW generating station in Milan, New Mexico--a program which would demonstrate a fully-integrated process with ultra-high removal of SO_x, NO_x, and Hg with emission levels similar to IGCC proposed levels. Additionally, a secondary system would be installed for the capture of carbon dioxide. The resulting byproducts are an ammonium sulfate (AS) fertilizer wet cake and a carbon dioxide stream. The ammonium sulfate stream would be further processed by Airborne's patented granulation process. This paper concludes by recognizing the success of the novel regeneration system, as a testament to the viability of this multi-pollutant control system process that produces a high-value useful product.

INTRODUCTION

Peabody Energy's-Mustang Energy Project has been selected to receive a grant from the Department of Energy under the second round of competition in President Bush's Clean Coal Power Initiative (CCPI). The technologies developed under CCPI would help maintain abundant coal resources as a cornerstone of the Nation's future domestic energy portfolio, particularly for power generation. The priorities for this round of competition were technology advancements for gasification-based electricity production, advanced mercury control, and sequestration or sequestration-readiness.

The Peabody Mustang Clean Coal Project teams Peabody Energy and co-sponsor Airborne Clean Energy (ACE), with ACE Partners: HPD LLC - Veolia Water North America, and Icon Construction. This project will demonstrate a commercial-scale of Airborne's emission control process, regeneration system, and fertilizer production systems at the Mustang Energy Company LLC's 300 megawatt coal-fired Mustang Generating Station in Milan, New Mexico. The \$79 million project, for which the U. S. Department of Energy (DOE) would provide \$19.7 million, will develop an innovative and cost-competitive multi-pollutant control process including energy

conservation techniques. Outstanding emissions performance will be achieved using technologies that would be commercially deployable in the near term. As such, there are four equally weighted objectives for this demonstration: achieving 99.5 percent removal of sulfur dioxide (SO₂), 98 percent removal of SO₃ (sulfuric acid mist precursor), 98 percent removal of nitrogen oxides (NO_x), and 90 percent total system removal of mercury from plant emissions, while turning the byproducts into a high-quality, impurity-free, high-value granular fertilizer.

The proposed project is a commercial scale demonstration of advanced emission control technologies integrated with existing emissions control equipment with the goal of simultaneously achieving lower combined emissions of NO_x, SO_x (and other acid gases), sulfuric acid mist and mercury than other operating coal-fired plants elsewhere in the Nation. While demonstrated as a new unit at the Mustang Generating Station, the technologies are also applicable to other new plant projects, conversions of existing wet scrubbing systems and repowering (i.e. replacement of older pulverized coal-fired units with more efficient contemporary pulverized coal units). Therefore, the deployment of these technologies can contribute to higher U.S. energy reliability in a cost-effective and environmentally responsible manner.

The Airborne Process™ commercialization team includes:

- Airborne Clean Energy, based in Terrace Park, Ohio, has the license for the proprietary Airborne Process™ for reducing emissions from coal-fueled generating plants. Airborne Clean Energy holds the exclusive rights to the patented Airborne Technology throughout North America.
- Peabody Energy (NYSE: BTU) is the world's largest private-sector coal company, with 2004 sales of 227 million tons of coal and \$3.6 billion in revenues. Its coal products fuel more than 10 percent of all U.S. electricity and 3 percent of worldwide electricity.
- HPD LLC is a subsidiary of Veolia Environment, which is the largest environmental services company in the world with more than 295,000 employees in about 100 countries and annual revenues of more than \$25.6 billion.
- Icon, Inc., headquartered in Dayton, Ohio, is a private minority owned corporation that specializes in providing engineering, construction and facility operating services for industrial applications. Icon's extensive experience with pan granulation, agglomeration and material handling is directly applicable to Airborne's fertilizer production facility.

TECHNOLOGY DEVELOPMENT

The technology combines the use of dry sodium bicarbonate injection, coupled with enhanced wet sodium bicarbonate scrubbing which is highly effective at capturing sulfur dioxide (SO₂), sulfur trioxide (SO₃), nitrogen oxide (NO), nitrogen oxides (NO_x), mercury (Hg), and other acid gas and heavy metal compounds from flue gas emissions. Prior to development of the Airborne Process™, sodium bicarbonate scrubbing had three serious drawbacks: 1) The cost of sodium bicarbonate is excessive; 2) The resulting by-product from the sodium bicarbonate SO_x reaction (sodium sulfate)

has limited economic value; and 3) Sodium sulfate disposal is expensive and poses significant environmental challenges.

As a result of these prohibitive operating issues, flue gas scrubbing with sodium bicarbonate has not realized significant market share despite its recognition as a superior scrubbing technology. Airborne's patented regeneration process eliminates the financial and disposal barriers of the highly effective sodium bicarbonate scrubbing process by converting the SO_x and NO_x pollutants into valuable sulfate and nitrate based fertilizers.

Airborne developed a process to regenerate sodium sulfate back into sodium bicarbonate and convert SO_x and NO_x pollutants into valuable sulfate and nitrate based fertilizers. Airborne's patented regeneration process eliminates the financial and disposal barriers of the highly effective sodium bicarbonate scrubbing process. The resulting byproduct was an ammonium sulfate (AS) fertilizer wet cake. This can be further processed by Airborne's patented granulation process to a hard, BB sized pellets. The ammonium sulfate byproduct can be enhanced into a wide variety of fertilizer products.

In any emissions control system, scrubbing effectiveness is the key technical issue. To evaluate SO_x and NO_x reductions using sodium bicarbonate, a series of bench and small combustor tests were completed at the CANMET Energy Technology Center in Ottawa. The results of this research were incorporated into the design of Airborne's 5 MW Facility in Ghent, Kentucky. At the Airborne Facility the SO₂ removal results were definitively proven. The promising results for NO_x and Hg removal from the Airborne Facility formed the basis for further research. Therefore, in 2004, Airborne undertook an advanced development program for capture of NO_x and Hg at the Energy and Environmental Research Center (EERC) of North Dakota University with cooperation of the Department of Energy.

AIRBORNE PROCESS™ DEMONSTRATION FACILITY

Airborne's facility was located near Ghent, Kentucky (see Figures 1 and 2). The flow schematic for the Airborne Process™ is shown on Figure 3. Construction of the facility started in August of 2002 and concluded in late March 2003. The Airborne Facility provided for the evaluation of the following key factors:

- Emission Reduction
 - Sulfur dioxide (SO₂) and sulfur trioxide (SO₃) reduction.
 - Nitrogen Oxides (NO_x) reduction.
 - Mercury (Hg) reduction.
- Regeneration Process
 - Sodium bicarbonate regeneration technology.
 - Sodium losses
 - Automation of the process.
 - Fertilizer purity.



FIGURE 1: AIRBORNE DEMONSTRATION ABSORBER, EMISSION CONTROL ISLAND AND ELECTRICAL BUILDING



FIGURE 2: AIRBORNE DEMONSTRATION PLANT REGENERATION BUILDING

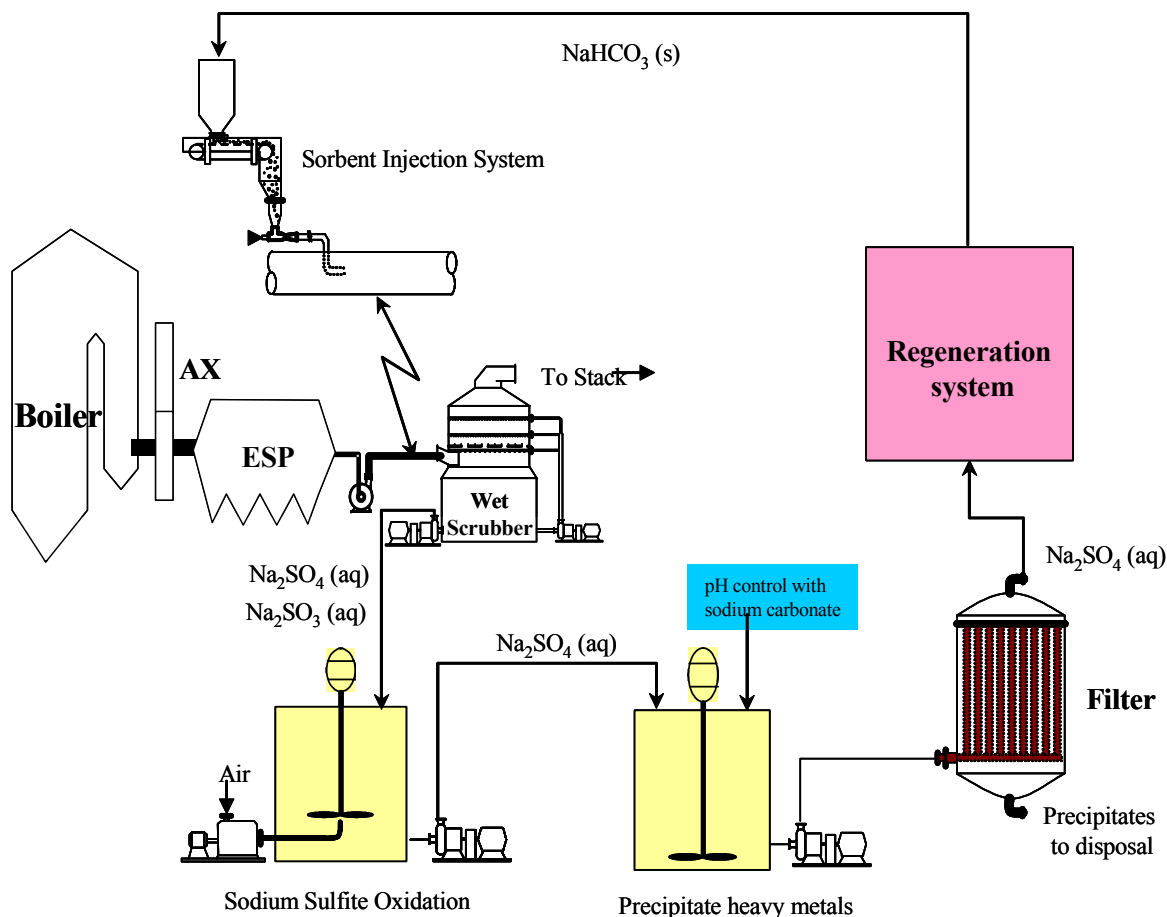


FIGURE 3: SIMPLIFIED AIRBORNE PROCESS FLOW DIAGRAM

EMISSION REDUCTION

SO₂ REDUCTION

The SO₂ reduction goals at the Airborne Ghent Facility and during Canmet testing were quite easily met. At the Airborne demo, SO₂ removal was consistently higher than 99.9% with outlet SO₂ levels below the range detectable by the instruments. The results for the July 8 Runs (Figure 4) show that the SO₂ removal was recorded as 100% regardless of the levels of the rest of the test parameters. These results are typical of the SO₂ response for the duration of the Airborne Ghent Facility testing. Near 100% SO₂ removal can be achieved easily at a Normalized Stoichiometry Ratio (NSR) of 1.0 or greater (the system was operated at NSR 1.2 or higher for NO_x control). Near 100% SO₂ removal could also be achieved easily with both dry injection of sodium bicarbonate and wet injection of sodium bicarbonate slurry directly into the absorber.

The absorber pH was not controlled, but was important. Normally, pH was about 8.0 to 8.3 during steady state operation. When the pH was low, the SO₂ removal was lower than 99%, as

expected. This only happened during the first several minutes of starting the absorber, especially after the absorber had been turned off for several hours.

The overall reactions for SO₂ removal are:

- 1) $2 \text{NaHCO}_3 + \text{SO}_2 + \frac{1}{2} \text{O}_2 \rightarrow \text{Na}_2\text{SO}_4 + 2 \text{CO}_2\uparrow + \text{H}_2\text{O}$
- 2) $\text{Na}_2\text{CO}_3 + \text{SO}_2 + \frac{1}{2} \text{O}_2 \rightarrow \text{Na}_2\text{SO}_4 + \text{CO}_2\uparrow + \text{H}_2\text{O}$

These reactions take place both in the gas stream (dry response) and in the absorber. With the two stage system for SO₂ removal, the results achieved were as expected because of the high stoichiometry (greater than 1.2 NSR) and resulting high pH of the absorber scrubbing solution described above.

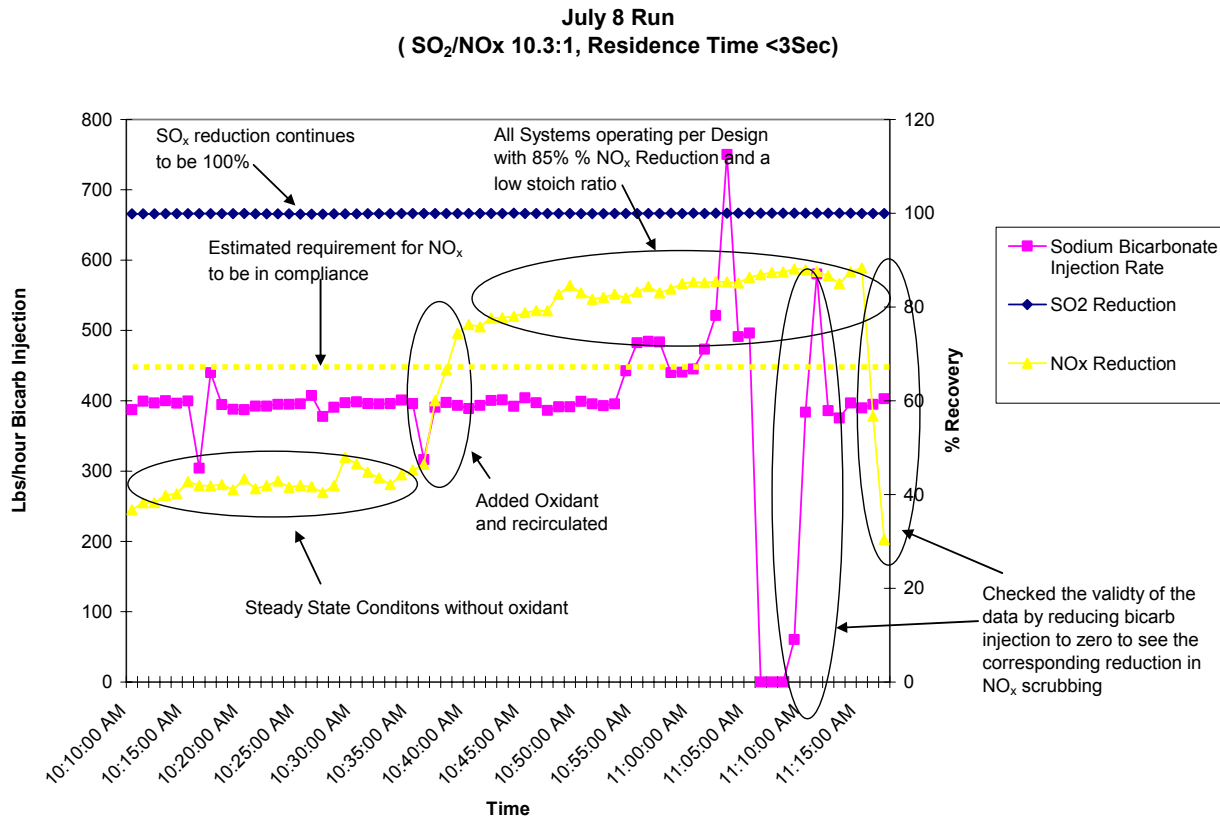
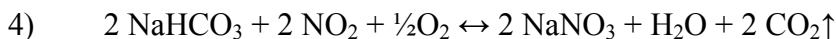
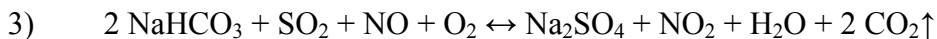


FIGURE 4: SO₂/NO_x REMOVAL AIRBORNE FACILITY, JULY 8, 2003

NO_x REDUCTION

The reduction of NO_x by sodium bicarbonate injection is clearly a function of the amount of SO_x removed. The following have been observed during the NO_x reduction process:

- 1) NO_x removal from flue gas with dry sodium bicarbonate sorbent occurs via a two-step process. Firstly, NO is converted to NO₂, and secondly, a portion of the NO₂ reacts to form NaNO₃. There is not a complete conversion to NO₂ as some of the NO is theorized to be converted to N₂.
- 2) The NO to NO₂ conversion step is closely connected with the SO_x removal, since the NO to NO₂ conversion happens during the reaction of Na₂CO₃ to Na₂SO₄. NO conversion to NO₂ requires O₂ to be present. When both NO and SO₂ are present, the dry sodium bicarbonate reaction products are Na₂SO₄ and NaNO₃. Na₂SO₃ is not found. The NO₂ not converted to NaNO₃ is then captured in the wet scrubber solution.
- 3) The ideal particle size for the sodium bicarbonate injection is in the order of 5 – 10 microns average since particles much smaller than 5 microns may pass through the wet scrubber without being captured. Larger particles do not react sufficiently for two primary reasons both related to surface area. The first is the reaction associated with liberation of CO₂ gas, know as calcination. The rate of calcination is temperature related, but would slow as the particle becomes larger because of the time required to heat the particle.
- 4) Calcination alone is only a minor effect; more important is that calcination results in a porous particle which creates surface area. Surface area is the most important parameter in NO_x reduction with sodium bicarbonate. Thus surface area is created in two ways, by producing fine particles, and through calcination. The particle size effect can be further evaluated by examining the work that has been done previously with sodium bicarbonate injection by several researchers and that has been applied commercially. For example, one researcher showed that SO₂ removal ranged from 50% with coarse ground bicarbonate (65 micron) to 99.3% for finely ground material (90% less than 13.8 micron). With 32 and 9 microns at NSR 1.0, the SO₂ removal increased from about 77% to 92%.
- 5) The overall dry response reactions for NO_x is:



The target for NO_x reduction is 60% or more in the combined response of dry injection and wet scrubbing and 98% or higher overall. The NO_x removal achieved during testing at the Canmet pilot was up to 60% while at the Ghent facility the NO_x removal ranged typically above 40% and higher depending upon test conditions. The highest NO_x reduction achieved at the Airborne Ghent Facility by dry injection and wet scrubbing was 92%.

One process condition modeled was the dry injection, coupled to the wet scrubbing with the addition of an oxidant to provide the final NO_x reduction. A typical run with this configuration is included earlier as the July 8th graph (Figure 4). The notes describing the various elements of the test are included on the graph. The NO_x removal increased from about 45% to almost 90% when the oxidant was added.

MERCURY REMOVAL

The level of mercury control in a wet scrubber is dependent upon the speciation of mercury emitted from the air heater. Particulate mercury is collected in the ESP and oxidized mercury was captured in the wet scrubber and retained. Elemental mercury, without the use of a capturing mechanism such as an oxidant, will not be effectively captured and could escape to the stack. Oxidized mercury in the form of mercuric chloride is readily soluble in aqueous solution, either alkaline or acidic. Elemental mercury is only sparingly soluble in aqueous solution. Therefore, no special provision is necessary to capture oxidized mercury in a wet scrubber. However, once captured, the oxidized mercury can react with constituents in the aqueous solution that can reduce the oxidized mercury back to elemental mercury. If that happens, the elemental mercury would rapidly evolve from the aqueous solution back to the flue gas.

Background mercury removal at the Airborne Facility was measured in a typical range of about 30 – 60% during testing at Ghent. Oxidants that will be tested in 2005 at EERC, in the pilot scale, removed both NO_x and Hg at levels near 100% in bench scale testing.

OXIDANTS

Initial tests with oxidants for post scrubbing of NO_x and Hg began at the Airborne Facility at Ghent (see Figure 4). In 2004, Airborne took the early success of the NO_x reduction with oxidants at the Airborne Facility into an extensive program at the Energy and Environmental Research Center (EERC) of North Dakota University. In this program several potential NO_x and elemental mercury (Hg⁰) absorption solutions were evaluated using bench- and pilot-scale wet scrubber systems and a gaseous mixture representative of that occurring downstream of Airborne Process™, dry/wet sodium bicarbonate scrubbing technology. Approximately 20 screening tests were performed in 2004 using an impinger train assembly similar to that used in American Society for Testing and Materials Method D6784-02 (Ontario Hydro method).

A 7-kW combustion system equipped with a wet scrubber (see Figure 5) containing packing was used to test the most promising absorbent solutions identified during the bench-scale testing. NO_x concentrations were measured at the inlet and outlet of the impinger train and pilot-scale wet scrubber using ion selective potentiometry (Testo 350) or chemiluminescence (Rosemount Analytical NGA 2000). Hg⁰ and total gaseous mercury (Hg^[tot]) concentrations were measured using an Hg^[tot]-to-Hg⁰ conversion system and atomic fluorescence spectroscopy (PS Analytical Sir Galahad or Tekran). Two solutions that effectively absorbed both NO_x and Hg⁰ were identified during the bench-scale screening tests, thus warranting additional testing in the pilot-scale wet scrubber. These pilot-scale testing are scheduled for spring 2005 and will further characterize the NO_x and Hg⁰ removal effectiveness of the two candidate solutions, including testing with coal.

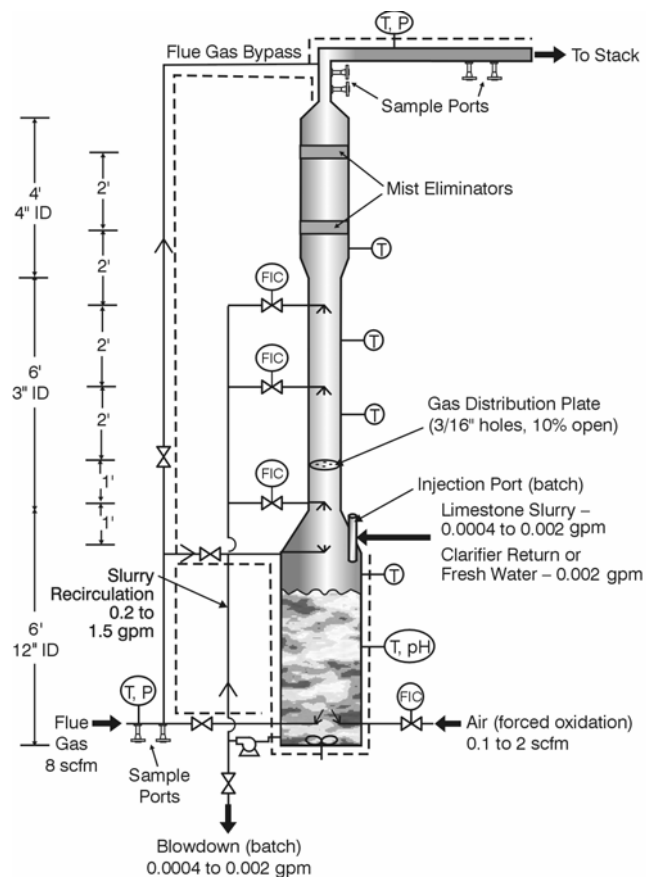


FIGURE 5: EERC WET SCRUBBER PILOT

Some of the results of the bench-scale testing are shown on Table 1. The results were quite interesting. Certain oxidants performed better in acidic or alkaline conditions. Most oxidant solutions that worked reasonably well performed better for Hg removal than NO_x removal. However, the best solutions that were selected for further testing in the pilot scale removed both NO_x and Hg at levels near 100%.

Figures 6 – 8 show a portion of the bench scale results. The Hg capture for Oxidant C under acidic conditions was almost 100% as is seen on Figure 6. The capture was almost immediate and continued until the instrument check at about 1.4 hours into the test. On Figure 7, both NO_x and Hg were effectively captured using Oxidant B.

Figure 8 shows very high Hg capture, but some interesting results for NO_x capture using Oxidant C under slightly acidic conditions. Rather than capture the NO, it appeared to be converted to the more soluble form of NO_x, NO₂. But, there was interference in the measurement thought to be due to other gases leaving the solution as the NO₂ level measured in the outlet was much higher than the inlet NO concentration and could not be accounted for otherwise. The NO₂, which would be seen in stack gases as a brown haze and other gases produced from the solution, could be removed by scrubbing with an alkali, but this would add an unnecessary level of complexity to the process.

TABLE 1: EERC BENCH SCALE TEST RESULTS		
Solution	NO _x Removal or NO Conversion to NO ₂	Hg Removal (Hg ^[tot] and Hg ^o)
Oxidant A	Low	Low
Oxidant A acidic	30 - 40%	30 - 40%
Acid A	Low	30 - 40%
Oxidant B acidic	30 - 40%	~100%
Oxidant B alkaline	~98% (about 4 ppm passed through)	~100%
Oxidant C acidic	~100%	~100%
Oxidant C slightly acidic	70 – 95%	~100%

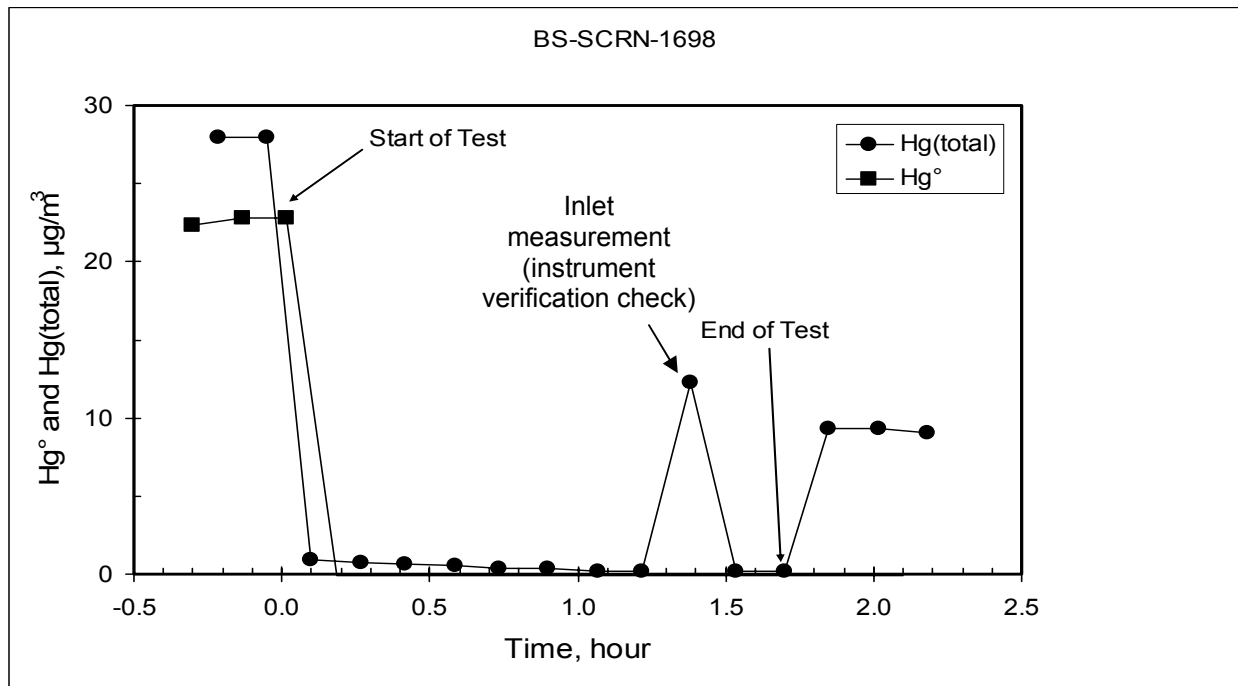


FIGURE 6: Hg REMOVAL BENCH SCALE RESULTS OXIDANT C ACIDIC

Oxidants B and C were selected for additional testing in the pilot. The pilot results for NO_x removal with Oxidant C were similar to the results seen at the bench scale, except the very high increase in apparent NO₂ seen on Figure 8 was not detected. This can be seen on Figure 9 below. There was a slight increase from the baseline of about 250 ppm NO_x to almost 300 ppm NO_x. Then the NO_x levels reduced to slightly less than the baseline value. Figure 9 also indicates that almost 100% of the total mercury (Hg^[tot] and Hg^o) was removed by Oxidant C under slightly acidic conditions.

The best NO_x removal results obtained from the pilot testing are shown on Figure 10. Essentially 100% NO_x removal was achieved using Oxidant B. Additional results from pilot testing of oxidants will be available in later in 2005.

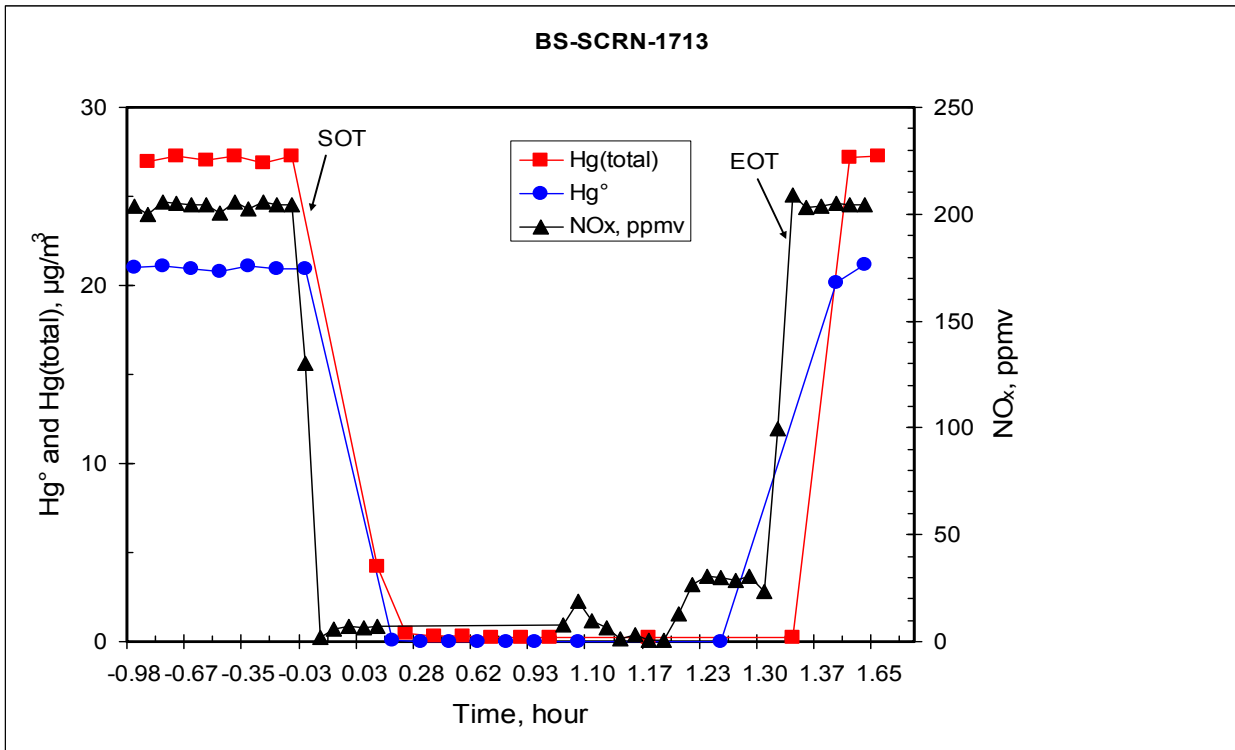


FIGURE 7: Hg & NO_x REMOVAL BENCH SCALE RESULTS OXIDANT B SLIGHTLY ACIDIC

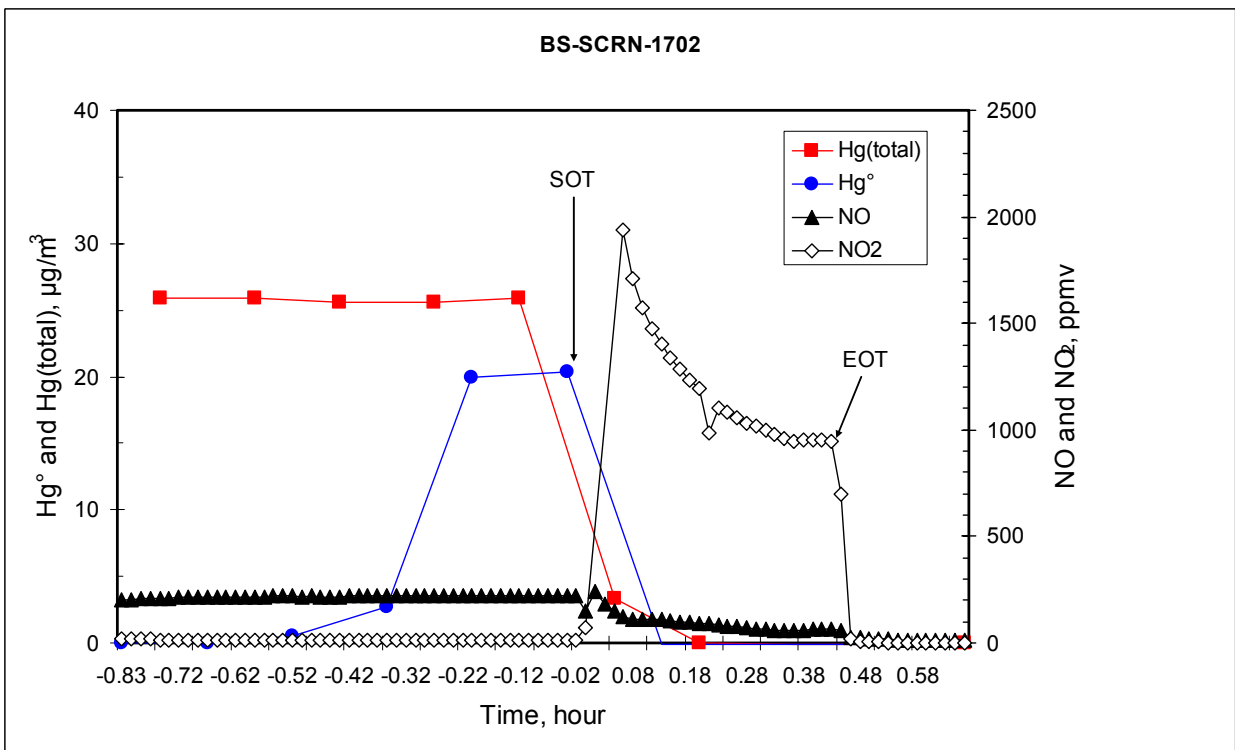


FIGURE 8: Hg & NO_x REMOVAL BENCH SCALE RESULTS OXIDANT C ALKALINE

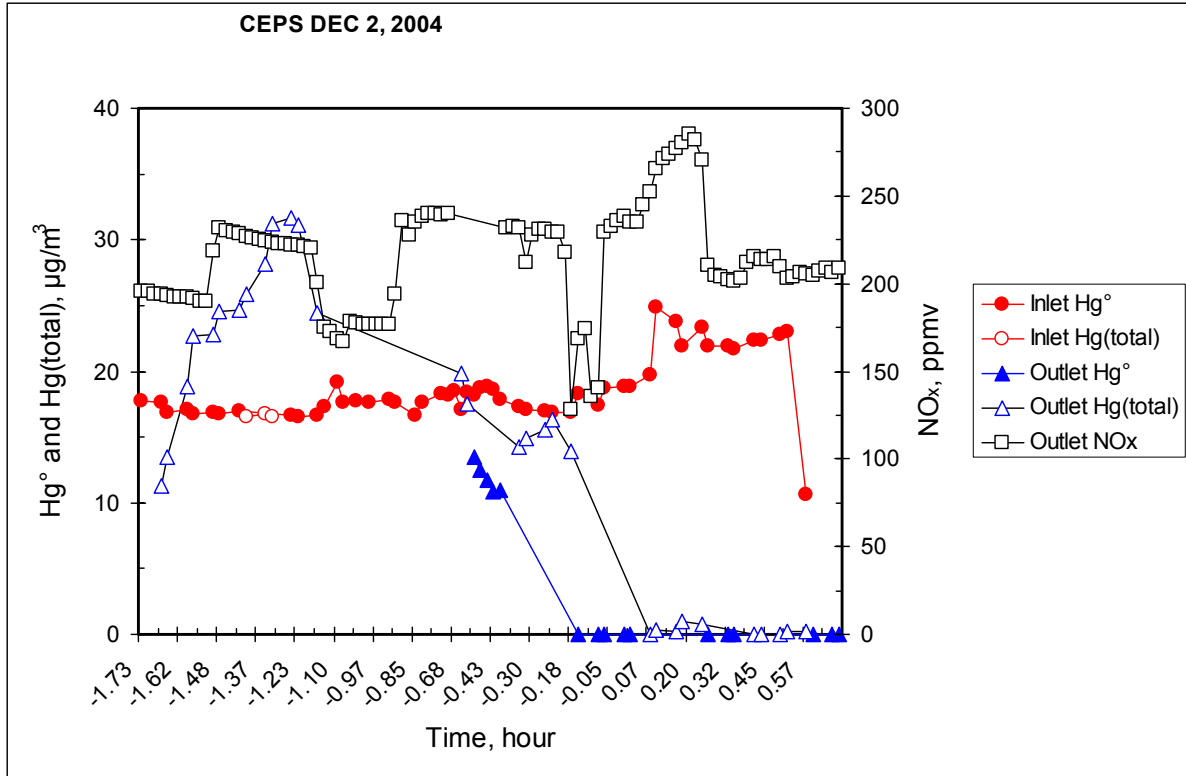


FIGURE 9: Hg & NO_x REMOVAL PILOT SCALE RESULTS OXIDANT C SLIGHTLY ACIDIC

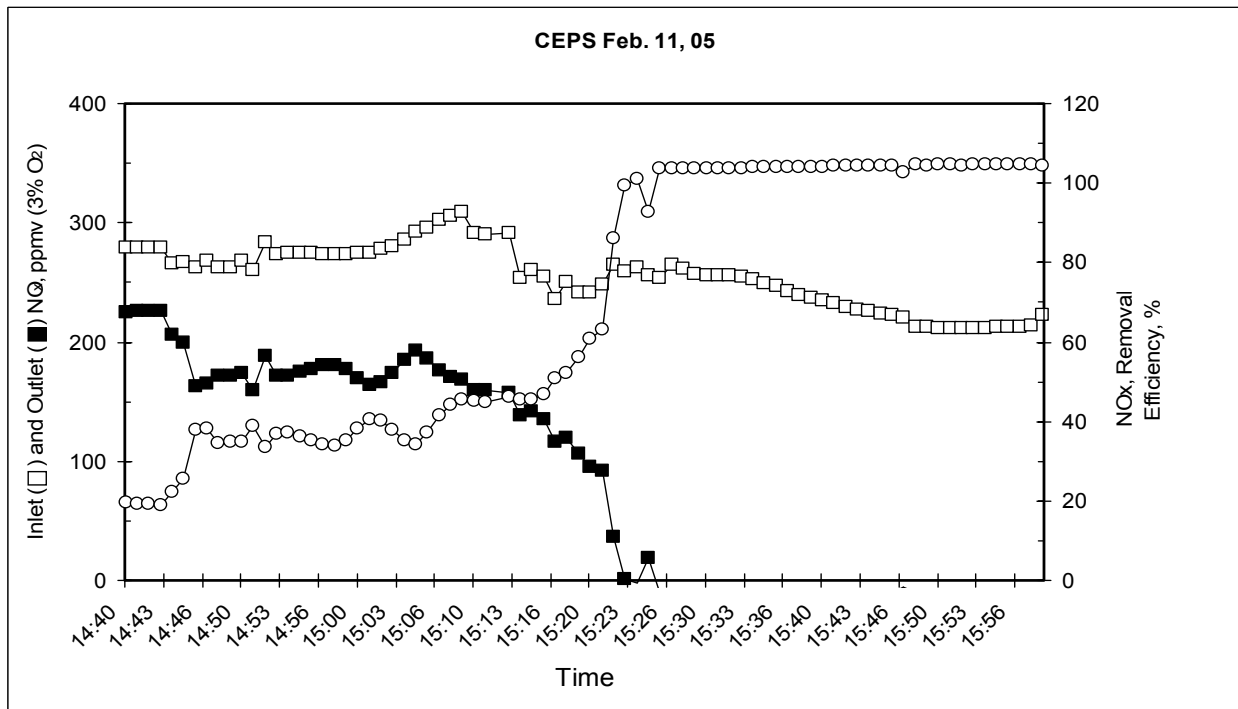
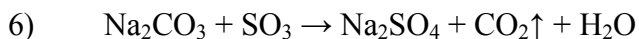
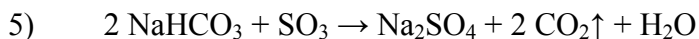


FIGURE 10: NO_x REMOVAL PILOT SCALE RESULTS OXIDANT B ALKALINE

PARTICULATE AND SO₃ REMOVAL

High Particulate Matter (PM) removal from the Airborne Process™ is important. The sodium bicarbonate and reaction products of the dry injection process must be adequately captured to both retain the materials for regeneration of the sodium bicarbonate to produce the ammonium sulfate product and to allow the utility to meet its requirements for particulate emissions. This material was captured effectively in the wet scrubber as was proven at the Airborne Facility. PM removal was measured on two separate days using modified EPA Method 5. All results indicated that the particulate levels exiting the Airborne Process™ were well below the EPA limit requirements of the Ghent plant. In fact, the results were quite impressive with high removal of particulate across the system. The net effect was that employment of the Airborne Process™ will result in no increase in the particulate emissions from a power plant. In fact, it is likely that the multiple stage wet scrubbing system of the Airborne Process™, which will be used when high removal of NO_x and mercury are desired, would result in a decrease in the overall particulate emission from a coal fired boiler.

The upstream removal of SO₃ with dry sodium bicarbonate injection is another major advantage of the Airborne Process™ over competing technologies. The Airborne Process™ removes the SO₃ prior to the conversion to H₂SO₄ mist by the following reactions:



Since SO₃ gas is a very strong acid and the amount of sodium bicarbonate injected (with an NSR relative to NO_x and SO₂ of 1.2) is as high as or higher than 120 times the theoretical amount required for Reactions 5 and 6 above, SO₃ is effectively removed. One problem is that SO₃ is highly acidic. Even more notable, is that with traditional processes such as limestone Flue Gas Desulfurization Systems (FGD), most of the SO₃ becomes a fine aerosol (about 0.3μ) of sulfuric acid (H₂SO₄) mist, which essentially passes untouched through the FGD. The acid mist results in a visible plume and negatively impacts the opacity measurement. Some plants have looked at the use of wet electrostatic precipitators (WESPs) to control acid mist emissions. However, they are very expensive since they are constructed of exotic materials due to the inherent corrosive nature of the acids present, they have a significant added pressure drop and they are difficult to support or integrate into the power plant emission control processes. Also, some plants just do not have the physical space available for a WESP. WESPs are an additional process step not required with the Airborne Process™.

REGENERATION SYSTEM

The Airborne Process™ for regeneration of sodium bicarbonate from sodium sulfate is an innovative, patented process. The Regeneration System at the Airborne Facility as supplied on an EPC (Engineer, Procure, and Construct) basis by of HPD LLC and operated by Icon, Inc. under the direction of engineers from ACE and HPD LLC, performed admirably.

The Regeneration process is a combination of seven main sequential operations:

- 1) Evaporative concentration of the filtrate,
- 2) Reactive crystallization of sodium bicarbonate,
- 3) Cooling crystallization of sodium sulfate decahydrate and ammonium bicarbonate for recycle to the reactive crystallizer,
- 4) Stripping of residual carbon dioxide and ammonia,
- 5) Cooling crystallization of ammonium sulfate-sodium sulfate double salt,
- 6) Evaporative crystallization of ammonium sulfate, and
- 7) Stripping of residual ammonia from the excess water recovered for use in the absorber.

The regeneration system was proven at the Airborne Facility to the point that guarantees on a commercial scale process can be offered. The equipment used in the steps of crystallization and solid/liquid separation performed as expected, and in some cases better than expected. As with any first of a kind project there were valuable lessons learned for future commercial equipment design optimizations.

CONCLUSION

The proposed Airborne Process™ incorporates all of the well recognized advantages of using sodium based sorbents. These practical and novel solutions will allow sodium based scrubbing to be economically viable over a wide segment of the electric generating market. Historically, sodium sorbents have seen limited application commercially, because of the cost of sodium carbonate/bicarbonate compared to lime and limestone, and because the spent reagent was water soluble and therefore problematic for disposal. The waste products from typical sodium based FGD systems are solutions of sodium sulfite and sodium sulfate contaminated with impurities such as fly ash and heavy metals.

The Airborne Process separates the impurities for disposal and converts the sodium sulfite and sulfate to ammonium sulfate and sodium bicarbonate. The ammonium sulfate undergoes additional processing using Airborne's advanced patented granulation techniques to produce a line of superior fertilizers. The sodium bicarbonate is returned to the FGD system for reuse. The Airborne Process employs an efficient regeneration system that keeps sodium losses to a minimum. The Airborne Process will capture NO_x which becomes sodium nitrate in the wet sodium scrubber. This material is ultimately converted to ammonium nitrate fertilizer.

The Airborne Facility has shown a completely integrated sodium bicarbonate scrubbing system for SO_x, NO_x and Hg, whilst demonstrating sodium sulfate purity, sodium bicarbonate regeneration and fertilizer production.

The Airborne Ghent Facility was successful. The performance of the system compared remarkably with the simulation results. All of the chemical processes were validated. As with any first of a kind project there were valuable lessons learned for future commercial equipment design optimizations. The success of the Airborne Facility, particularly the novel regeneration system, is a testament to the viability of this process as a multi-pollutant control system that produced a high-value useful product.

Initial tests with oxidants for post scrubbing of NO_x and Hg began at the Airborne Facility at Ghent in 2003 and resulted in about 90% total capture of NO_x. In 2004, Airborne took the early success of the NO_x reduction with oxidants at the Airborne Facility into an extensive program at the Energy and Environmental Research Center (EERC) of North Dakota University. In this program several potential NO_x and elemental mercury (Hg⁰) absorption solutions were evaluated using bench- and pilot-scale wet scrubber systems and a gaseous mixture representative of that occurring downstream of Airborne Process™, dry/wet sodium bicarbonate scrubbing technology.

The results of oxidant testing show that there are several effective solutions. Even some solutions that show medium removal ranges would remove at higher rates with an appropriate modification to the mass transfer means. The oxidant selected for commercial applications, would be based on economics, availability, desired level of capture, and/or desired end product. Most oxidant solutions that worked reasonably well in the EERC test program performed better for Hg removal than NO_x removal. The best solutions removed both NO_x and Hg at levels near 100%.

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KEYWORDS

Sodium Bicarbonate, Sodium Sorbent, SO_x, NO_x, SO₂, Hg, mercury, NO, NO₂, ammonium sulfate, Airborne Clean Energy, multi-pollutant control system, oxidant, Emission Control, Fertilizer, Regeneration, Granulation.