

An Update on DOE/NETL's Mercury Control Technology Field Testing Program

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The U.S. Department of Energy's National Energy Technology Laboratory (NETL), under the Office of Fossil Energy's Innovations for Existing Plants (IEP) Program, manages the premier mercury (Hg) research and development (R&D) program for coal-fired power generation facilities in the world. Working collaboratively with the U.S. Environmental Protection Agency (EPA), the Electric Power Research Institute (EPRI), power plant operators, state and local agencies, and a host of research organizations and academic institutions, the IEP Program has identified the major factors that affect Hg speciation and capture in coal combustion flue gas and funneled this knowledge into the development of a suite of Hg-specific control technologies for the diverse fleet of U.S. coal-fired power plants. The high performance observed during many of these field tests has given coal-fired power plant operators the confidence to begin deploying technology. As of April 2008, over 44 gigawatts (GW) of coal-based electric generating capacity have placed orders for full-scale activated carbon injection (ACI) systems.

BACKGROUND

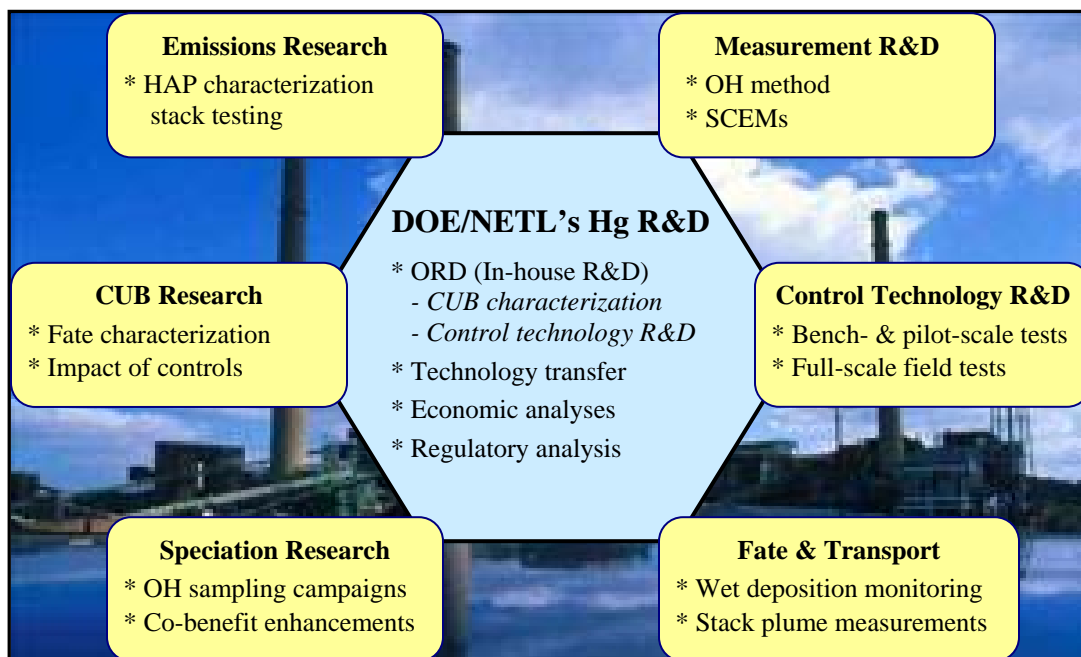
Since first being identified for potential regulation in the 1990 Clean Air Act Amendments, there has been concern within the industry whether it would be possible to develop cost-effective emission control technologies for Hg because of its low concentration and reactivity during coal combustion. However, while technical issues remain, the U.S. Department of Energy's NETL has been successful, through public-private partnerships, in significantly improving both the cost and performance of Hg control technology.

NETL initiated comprehensive Hg research under the Office of Fossil Energy's IEP Program in the early 1990s to ensure that cost-effective and reliable pollution control technologies are available for the existing fleet of coal-fired utility boilers.¹ To comprehensively address the life-cycle of Hg released during coal combustion, NETL has directed over \$80 million in Federal funding over the last decade toward external (extramural) and in-house research projects focusing on six inter-related research areas (see Figure 1):

- Emissions characterization;
- Development and testing of total and speciated measurement devices;
- Speciation research;
- Development and testing of control technologies;

- Coal utilization by-products (CUB) characterization; and
- Fate and transport of emissions.

Figure 1 – Overview of Hg R&D Conducted under NETL’s IEP Program



Emissions characterization performed by NETL and others in the early 1990s showed that Hg was not effectively captured across existing air pollution control device (APCD) configurations. Further conclusions were limited by the lack of a reliable method to measure the different chemical forms of Hg in coal combustion flue gas. To overcome this hurdle, NETL co-funded the development and validation of the Ontario Hydro (OH) method through a Jointly Sponsored Research Program with the University of North Dakota’s Energy and Environmental Research Center (UNDEERC).

Analysis of OH method sampling campaigns revealed that the trace amount of Hg present in coal is volatilized during combustion and converted to gaseous elemental mercury (Hg^0). Subsequent cooling of the coal combustion flue gas and interaction of the gaseous Hg^0 with other flue gas constituents, such as chlorine (Cl) and unburned carbon (UBC), result in a portion of the Hg^0 being converted to gaseous oxidized forms of mercury (Hg^{2+}) and particulate-bound mercury (Hg^P). As a result, coal combustion flue gas contains varying percentages of Hg^P , Hg^{2+} , and Hg^0 and the exact speciation has a profound effect on the Hg capture efficiency of existing APCD configurations, which has been found to range from 0 to over 90%.² The Hg^P fraction is typically removed by a particulate control device such as an electrostatic precipitator (ESP) or fabric filter (FF). The Hg^{2+} portion is water-soluble and therefore a relatively high percent can be captured in wet flue gas desulfurization (FGD) systems, while the Hg^0 fraction is generally not captured by existing APCD.

Insight into the factors that influence Hg speciation and capture in coal combustion flue gas allowed NETL to focus its R&D efforts on the most promising technologies for controlling Hg emissions from coal-based power systems. One obvious cost-effective strategy for power plants that have or plan to install FGD would be to take advantage of the “co-benefit” capture of Hg. However, wet scrubbers do not achieve 100% capture of Hg²⁺, nor as noted above is all the Hg in an oxidized form, even for bituminous coal. Moreover, this approach is not a viable Hg control option for power generation facilities not equipped with FGD systems. As such, NETL initiated an R&D program in the mid-1990s directed at two general approaches for controlling Hg -- (1) Hg-specific control technology such as sorbent injection and (2) Hg⁰ oxidation concepts.

In 2000, following laboratory through pilot-scale development of these technology approaches, NETL launched a three-phase field testing program. This program called for the installation and full-scale and slip-stream testing of the most promising Hg control technologies at operating coal-fired power plants. The initial field testing (Phase I) focused on untreated ACI and improving the capture of Hg across wet FGD systems, while Phase II, which began in 2003, was expanded to include testing of chemically-treated ACI, sorbent (i.e., activated carbon) enhancement additives, and Hg⁰ oxidation catalysts. The goal of Phases I and II was to develop Hg control technologies (available for commercial demonstration by year-end 2007 for all coal ranks) that could achieve 50 to 70% Hg capture at costs 25 to 50% less than the baseline (1999) estimate of about \$60,000 per pound of Hg removed (\$/lb Hg removed). Phase III of the field testing program was initiated in 2006 and directed at continued field testing of advanced Hg control technologies that could achieve 90% or greater capture at a 50 to 75% cost reduction and that would be available for commercial demonstration by 2010. In addition, the Phase III field testing emphasized the assessment of potential balance-of-plant impacts associated with continuously operating a Hg-specific control technology.

This paper focuses on results from the Phase II Hg control technology field testing program with the data segregated by technology. In addition, the results of NETL’s economic analysis of Hg control via ACI are presented, along with a discussion of potential coal utilization by-product (CUB) impacts. Preliminary results from NETL’s Phase III Hg field testing program are also presented.

Field Testing of Advanced Mercury Control Technologies

Building on promising advances from the Phase I field testing program, NETL selected eight new full-scale projects in September 2003 to test and evaluate Hg control technologies under a Phase II, Round 1 (Phase II-1) field testing solicitation. The Phase II-1 projects shown in Table 1 were initiated in 2004 and completed in 2007. An additional six projects – representing seven technologies^a - were subsequently awarded in October 2004 under a Phase II, Round 2 (Phase II-2) solicitation that were also

^a The seven Phase II-2 Hg control technologies are: TOXECON™, TOXECON™ II, high-temperature Hg sorbents, brominated PAC injection, chemically-treated PAC injection via the Mer-Cure™ process, wet FGD chemical additives, and an integrated approach to Hg control that includes combustion modifications.

completed in 2007 (Table 2). Total funding for the Phase II program was about \$45 million with the various research teams providing nearly \$13 million in cost sharing.

The Phase II projects focus on longer-term (~ 1 month at optimized conditions), large-scale field testing on plants burning primarily low-rank coals or blends (with some units burning bituminous coal) and equipped with a variety of APCD configurations. Most of the 14 projects fall under two general categories of Hg control – sorbent injection or oxidation enhancements. Sorbent injection generically describes conventional (untreated) ACI, chemically-treated ACI, or the injection of non-carbon sorbents into the flue gas for Hg control. Mercury oxidation enhancements are intended to improve the Hg capture efficiency of conventional ACI or downstream APCD by converting Hg^0 to a more reactive oxidized state. For instance, coal or flue gas additives are being investigated both alone, and in conjunction with conventional ACI. Additional Hg control technologies are being tested to enhance Hg capture at coal-fired units equipped with wet FGD systems. These FGD-related technologies include coal and flue gas chemical additives and fixed-bed catalysts to increase levels of Hg^{2+} in the combustion flue gas, and wet FGD chemical additives to promote Hg capture and prevent re-emission of Hg^0 from the FGD absorber vessel.

Table 1 -- NETL's Phase II-1 Field Testing Projects

Project Title	Lead Company	Test Location	Coal Rank	APCD Configuration
Evaluation of Sorbent Injection for Mercury Control	ADA-ES	Sunflower Electric's Holcomb Unit 1	PRB	SDA/FF
		AmerenUE's Meramec Unit 2	PRB	CS-ESP (320 SCA)
		Missouri Basin Power Project's Laramie River Unit 3	PRB	SDA & CS-ESP (599 SCA)
		DTE Energy's Monroe Unit 4	PRB/Bit. Blend	SCR & CS-ESP (258 SCA)
		American Electric Power's Conesville Unit 6	High-Sulfur Bituminous	CS-ESP (301 SCA) & Wet FGD
		AmerenUE's Labadie Unit 2	PRB	CS-ESP (279 SCA)
Sorbent Injection for Small ESP Mercury Control	URS Group	Southern Company's Plant Yates Unit 1	Low-Sulfur Bituminous	CS-ESP (173 SCA) & Wet FGD
		Southern Company's Plant Yates Unit 2	Low-Sulfur Bituminous	CS-ESP (144 SCA)
		Reliant Energy's Shawville Unit 3	Mid-Sulfur Bituminous	Two CS-ESPs (82 & 230 SCA)
Enhancing Carbon Reactivity in Mercury Control in Lignite-Fired Systems	UNDEERC	Basin Electric's Leland Olds Unit 1	ND Lignite	CS-ESP (320 SCA)
		Great River Energy's Stanton Unit 10	ND Lignite	SDA/FF
		Basin Electric's Antelope Valley Unit 1	ND Lignite	SDA/FF
		Great River Energy's Stanton Unit 1	PRB	CS-ESP (470 SCA)
		Montana-Dakota Utilities Co. Lewis & Clark Station	ND Lignite	Mechanical Collector & Wet Venturi Scrubber
Advanced Utility Mercury Sorbent Field-Testing Program	Sorbent Technologies	DTE Energy's St. Clair Unit 1	PRB/Bit. Blend	CS-ESP (SCA 467)
		Duke Energy's Buck Unit 6	Low-Sulfur Bituminous	HS-ESP (240 SCA)
Demonstration of Amended Silicates for Mercury Control	Amended Silicates	Duke Energy's Miami Fort Unit 6	Mid-Sulfur Bituminous	CS-ESP (353 SCA)
Pilot Testing of Mercury Oxidation Catalysts for Upstream of Wet FGD Systems	URS Group	Luminant Power's Monticello Unit 3	TX Lignite/PRB blend	CS-ESP (452 SCA) & Wet FGD
		Southern Company's Plant Yates Unit 1	Low-Sulfur Bituminous	CS-ESP (173 SCA) & Wet FGD
Evaluation of MerCAP™ for Power Plant Mercury Control	URS Group	Great River Energy's Stanton Unit 10	ND Lignite	SDA/FF
		Southern Company's Plant Yates Unit 1	Low-Sulfur Bituminous	CS-ESP (173 SCA) & Wet FGD
Mercury Oxidation Upstream of an ESP and Wet FGD	UNDEERC	Minnkota Power's Milton R. Young Unit 2	ND Lignite	CS-ESP (375 SCA) & Wet FGD
		Luminant Power's Monticello Unit 3	TX Lignite/PRB blend	CS-ESP (452 SCA) & Wet FGD

Table 2 -- NETL's Phase II-2 Field Testing Projects

Project Title	Lead Company	Test Location	Coal Rank	APCD Configuration
Field Testing of Activated Carbon Injection Options for Mercury Control	UNDEERC	Luminant Power's Big Brown Unit 2	TX Lignite/PRB Blend	CS-ESP (162 SCA) & COHPAC® FF
Field Demonstration of Enhanced Sorbent Injection for Mercury Control	ALSTOM-PPL	PacifiCorp's Dave Johnston Unit 3	PRB	CS-ESP (600 SCA)
		Basin Electric's Leland Olds Unit 1	ND Lignite	CS-ESP (320 SCA)
		Reliant Energy's Portland Unit 1	Mid-Sulfur Bituminous	CS-ESP (284 SCA)
Low Cost Options for Moderate Levels of Mercury Control	ADA-ES	Entergy's Independence Unit 1	PRB	CS-ESP (542 SCA)
		MidAmerican's Louisa Unit 1	PRB	HS-ESP (459 SCA)
		MidAmerican's Council Bluffs Unit 2	PRB	HS-ESP (224 SCA)
Brominated Sorbents for Small Cold-Side ESPs, Hot-Side ESPs, and Fly Ash use in Concrete	Sorbent Technologies	Progress Energy's Lee Unit 1	Low-Sulfur Bituminous	CS-ESP (300 SCA)
		Midwestern Generation's Crawford Unit 7	PRB	CS-ESP (112 SCA)
		Midwestern Generation's Will County Unit 3	PRB	HS-ESP (233 SCA)
Field Testing of a Wet FGD Additive for Enhanced Mercury Control	URS Group	Luminant Power's Monticello Unit 3	TX Lignite/PRB blend	CS-ESP (452 SCA) & Wet FGD
		Southern Company's Plant Yates Unit 1	Low-Sulfur Bituminous	CS-ESP (173 SCA) & Wet FGD
		Indianapolis Power & Light's Petersburg Unit 2	High-Sulfur Bituminous	CS-ESP (430 SCA) & Wet FGD
Demonstration of Integrated Approach to Mercury Control	GE-EER	Progress Energy's Lee Unit 3	Low-Sulfur Bituminous	CS-ESP (300 SCA)

The intent of NETL's Phase I and II Hg control technology field testing programs was to work with industry to evaluate the most promising Hg control technologies at full-scale in a variety of configurations. Although 30-day long-term tests were conducted in Phase II, the test period was not sufficient to answer many fundamental questions about long-term consistency of Hg removal and reliability of the system when integrated with plant processes. As the technologies move towards commercial implementation, it is critical to accurately define the Hg removal performance and costs so that power companies and policy makers can make informed decisions. NETL awarded nine new projects in 2006 to conduct Hg control tests at full-scale coal-fired units and in the laboratory, under a Phase III Hg control solicitation. Table 3 presents general information about each of the Phase III projects selected by NETL. Total funding for the Phase III program was just over \$27 million with the various research teams providing nearly \$9 million in cost sharing. Building on advances from the Phase I and II mercury projects, Phase III has four topic areas.

- **Topic Area I** – Phase III Field Testing of Mercury Control Technology Capable of Achieving 90% or Greater Removal of Mercury
- **Topic Area II** – Additional (Round 3) Phase II Field Testing of Mercury Control Technology Capable of Achieving 50-70% Mercury Removal
- **Topic Area III** – Novel Combustion and Post-Combustion Control Technologies
- **Topic Area IV** – Novel Pre-combustion Control Technology

Table 3 -- NETL's Phase III Mercury Control Technology Projects

Project Title	Lead Company	Test Location	Coal Rank	APCD Configuration
<i>Topic Area I Projects</i>				
Demonstration of Mer-Cure™ Technology	ALSTOM-PPL	Lower Colorado River Authority's Fayette Unit 3	PRB Blends	CS-ESP (640 SCA) & Wet FGD
Long-Term Demonstration of SEA Technology for Mercury Control	UNDEERC	Kansas City Power & Light's Hawthorne Unit 5	PRB	SCR & SDA/FF
		E.ON America's Mill Creek Unit 4	High-Sulfur Bituminous	SCR, CS-ESP (232 SCA) & Wet FGD
Full-Scale Field Trial of the Low Temperature Mercury Capture Process	CONSOL Energy	TBD	TBD	TBD
Long-Term ACI Field Test for >90% Mercury Removal	ADA-ES	Rocky Mountain Power's Hardin Station	PRB	SDA/FF
<i>Topic Area II Projects</i>				
Full-Scale Testing of Mercury Oxidation Catalyst	URS Group	LCRA's Fayette Unit 3	PRB Blends	CS-ESP (640 SCA) & Wet FGD
Mercury Control for Plants Firing Texas Lignite and Equipped with ESP-Wet FGD	URS Group	NRG Texas Power LLC's Limestone Station Unit 1	TX Lignite/PRB blend	CS-ESP (452 SCA) & Wet FGD
Evaluation of Mercury Control Strategies on a Cyclone Boiler with SCR	ADA-ES	Public Service of New Hampshire Company's Merrimack Unit 2	Mid-Sulfur Bituminous	SCR & (2) CS-ESPs in series (120 & 230 SCAs)
<i>Topic Area III Project</i>				
Utilization of Partially Gasified Coal for Mercury Removal	GE-EER	Boiler Simulator Facility	Multiple	Multiple
<i>Topic Area IV Project</i>				
Pilot Testing of WRI's Novel Mercury Control Technology	WRI	WRI & UNDEERC's PTC	Multiple	Multiple

The following is a discussion of results obtained during NETL's Phase II Hg control technology field testing program. While multiple Hg control technologies were explored at many of the Phase II field testing sites, the results are grouped by the technology

selected for evaluation during the 30-day extended tests. In addition, preliminary results from the Phase III field testing sites are presented.

Elemental Hg Oxidation Enhancements

Oxidation of flue gas Hg^0 followed by absorption of Hg^{2+} across a wet FGD system has the potential to be a reliable and cost-effective Hg control strategy for coal-fired power plants. To optimize Hg capture across FGD systems, NETL is funding the development of technologies that promote Hg^0 oxidation in coal combustion flue gas: retrofit Hg^0 oxidation catalysts and chemical additives. The impact of combustion modifications, such as coal reburn, on flue gas Hg^0 oxidation has also been examined under the IEP Program.³ In addition, NETL field tested FGD additives designed to suppress Hg^0 re-emissions across the scrubber.

Combustion Modifications

General Electric Energy and Environmental Research Corporation (GE EER) evaluated a novel multi-pollutant control technology to reduce Hg, NO_x , and carbon monoxide emissions, while simultaneously improving plant efficiency and reliability, at Progress Energy's bituminous coal-fired Lee Station Unit 3 that is equipped with a CS-ESP and SO_3 flue gas conditioning (FGC) system.⁴ The primary objective of this Phase II project was to achieve at least 70% incremental Hg capture via: (1) combustion modifications designed to enhance "co-benefit" Hg capture by fly ash; (2) duct humidification to reduce ESP temperature; and (3) ACI upstream of the ESP for polishing control. Preliminary results indicate a 38% improvement in "co-benefit" Hg capture following combustion optimization activities. Meanwhile, untreated ACI at about 18 lb/MMacf achieved 80% total Hg removal with SO_3 conditioning idled, but the removal efficiency was limited to approximately 55% with the operation of SO_3 FGC.

Elemental Hg Oxidation Catalysts

URS Corporation (URS), in collaboration with EPRI, Great River Energy (GRE), City Public Service (CPS) of San Antonio, and the North Dakota Industrial Commission, conducted pilot-scale testing of Hg^0 oxidation catalysts at GRE's North Dakota (ND) lignite-fired Coal Creek Station (CCS) and CPS of San Antonio's J.K. Spruce Plant, which burns PRB coal. CCS is equipped with a wet FGD system downstream of a CS-ESP, while Spruce is equipped with a wet FGD downstream of a reverse-gas FF. Pilot catalyst skids installed at both plants were loaded with a palladium (Pd#1) catalyst, a selective catalytic reduction (SCR) catalyst, and an experimental activated carbon (C#6) catalyst. In addition, a subbituminous ash-based (SBA#5) catalyst was installed at CCS, while a gold (Au) catalyst was evaluated at Spruce. In a full-scale application, the fixed-bed catalysts would be installed downstream of an ESP or FF, to: (1) minimize fly ash deposition on the catalysts; (2) prevent or minimize catalyst erosion; and (3) ensure a low flue gas temperature and flow rate, which reduces the catalyst space velocity and minimizes the length of catalyst required.

High levels of baseline Hg^{2+} downstream of the FF at Spruce made it difficult to evaluate the Hg^0 oxidation catalysts. Final catalyst activity measurements, conducted at CCS in June 2004, showed 79% Hg^0 oxidation across the C#6 catalyst, with nearly 13 months of

operation, and about 67% oxidation across the Pd#1 catalyst after more than 20 months of operation.⁵ Significantly lower activity was measured for the SCR and SBA#5 catalysts. In an attempt to extend the effective life of the catalysts, *in-situ* thermal catalyst regeneration tests were performed. Following thermal regeneration, Hg⁰ oxidation across the Pd#1 catalyst increased from 67 to 88% (near the 95% activity of the fresh catalyst), while the C#6 catalyst showed no improvement. It is important to note that these “proof of concept” tests were conducted simply to determine if the catalysts could be thermally regenerated, and conditions were not adjusted to optimize thermal regeneration. Meanwhile, 79% total Hg capture was observed across the pilot-scale wet limestone forced oxidation (LSFO) FGD, with 84% Hg²⁺ at the FGD inlet.

URS has completed pilot-scale testing of Hg⁰ oxidation catalysts at Luminant Power’s Monticello Steam Electric Station (MoSES) Unit 3, which fires a blend of Texas (TX) lignite and PRB coals.⁶ In January 2005, four Hg⁰ oxidation catalysts were installed downstream of the CS-ESP: (1) Au; (2) SCR; (3) regenerated Pd#1 from CCS; and (4) fresh Pd#1. During catalyst inspection, URS observed severe fly ash buildup on the catalyst surfaces, likely caused by frequent pilot unit outages during the test campaign. Following *in-situ* catalyst cleaning in August 2006, Hg⁰ oxidation was approximately 72% across the regenerated Pd#1 catalyst and 66% across the Au catalyst, after 17 months of pilot-scale operation. Tests completed in April 2005 indicated total Hg capture across a pilot-scale wet FGD ranged from 76 to 87%, compared to only 36% removal under baseline conditions. This equates to about 70% incremental Hg capture due to the catalysts.

URS has also completed pilot-scale testing of the catalytic Hg oxidation technology at Southern Company’s Plant Yates Unit 1, which fires a low-sulfur Eastern bituminous coal. The pilot catalyst skid, installed downstream of a CS-ESP, was loaded with fresh Pd#1 and Au catalysts in December 2005, along with regenerated SCR and Au catalysts from J.K. Spruce. After nearly 11 months of operation, Hg⁰ oxidation measured 58% across the fresh Au catalyst, 38% across the fresh Pd#1 catalyst, 32% across the regenerated SCR catalyst, and 26% across the regenerated Au catalyst. The pilot catalyst skid was taken off-line in January 2007 to accommodate an SO₃ injection test plan.

Under a Phase III award, URS began a full-scale field test of a gold-based Hg⁰ oxidation catalyst at Lower Colorado River Authority’s Fayette Power Project Unit 3 in May 2008. The test is intended to confirm the required catalyst quantities and catalyst life for achieving an average of 70% or greater Hg⁰ oxidation in PRB flue gases over a two-year period. The project represents the next logical advancement of the catalytic oxidation technology from its current pilot-scale. It will answer technical questions such as the catalyst quantity required to achieve high Hg oxidation percentages, catalyst life, the efficiency of catalytically-oxidized Hg capture in full-scale wet FGD systems, and the ability to keep the catalysts clean of fly ash buildup at full-scale with sonic horns.

Chemical Additives

NETL is funding Phase II field tests of chemical additives designed to promote flue gas Hg⁰ oxidation and enhance FGD Hg capture.⁷ The additives, collectively referred to as

sorbent enhancement additives (SEA) by UNDEERC and as oxidation additives (OA) by URS, are sprayed onto the coal as an aqueous salt solution. This approach maximizes the residence time available for interactions with Hg^0 . Full-scale field testing of this technology has been completed at Minnkota Power Cooperative's Milton R. Young (MRY) Unit 2 and Luminant Power's MoSES Unit 3.

MRY Unit 2 fires ND lignite coal in a cyclone boiler and is equipped with a CS-ESP and wet FGD. UNDEERC evaluated three additives during short-term parametric tests: SEA1, calcium chloride (CaCl_2); SEA2, a proprietary chemical formulation; and magnesium chloride (MgCl_2). SEA2 yielded the best results, achieving approximately 44% total Hg capture across the ESP/FGD combination with injection at 75 ppm (on a dry coal basis), as compared to less than 20% Hg capture with SEA1 and MgCl_2 injection at 500 ppm. About 60% total Hg capture was observed with SEA2 injection at 50 ppm (on a dry coal basis), coupled with DARCO[®] Hg (an untreated PAC developed by NORIT Americas, Inc.) injection at 1 lb/MMacf. During the 30-day long-term test, total Hg capture across the ESP/FGD configuration ranged from 50 to 65% with SEA2 injection at 60-100 ppm (on a dry coal basis) and DARCO[®] Hg injection at 0.15 lb/MMacf.

During parametric testing at MoSES Unit 3, URS evaluated the performance of OA1 (CaCl_2) and OA2 (calcium bromide - CaBr_2). These trials clearly displayed the superior performance of CaBr_2 as 72% Hg^{2+} was observed at the ESP outlet with an injection rate of 100 ppm Br in the coal (on a dry basis). As a result, long-term testing was conducted with CaBr_2 . The two-week test, at a CaBr_2 injection rate equivalent to 55 ppm Br in the coal, oxidized 67% of the Hg entering the FGD, resulting in an average total Hg capture of 65%. At a CaBr_2 injection rate equivalent to 113 ppm Br in the coal, Hg^0 oxidation reached 85%, resulting in an average total Hg capture of 86% over the subsequent two-week test. In addition, a short-term test conducted with a CaBr_2 injection rate equivalent to 330 ppm Br in the coal resulted in 92% total Hg capture across the ESP/FGD configuration.

Addressing Hg Re-emissions across FGD Systems

Originally thought to be a sampling artifact, Hg^0 re-emissions have been observed at several coal-fired units and occur when Hg^{2+} captured by a wet FGD is chemically-reduced within the vessel and re-emitted as Hg^0 . Through funding provided by NETL, URS is conducting bench-scale testing to determine the mechanisms and kinetics of the aqueous reactions of Hg absorbed by wet FGD systems, and to develop a kinetics model that predicts Hg reduction and re-emission reactions in wet FGD systems over a wide range of chemical conditions.⁸ The underlying hypothesis of this project is that Hg re-emission reactions (apparently driven by SO_2 -derived "sulfite" species in the liquid phase) are limited by kinetics rather than equilibrium. Results indicate that low FGD chloride concentrations can slow Hg^{2+} reduction, while high chloride concentrations may completely inhibit the sulfur reactions, preventing Hg^{2+} reduction within the FGD vessel. Moreover, results show that the rate of Hg^{2+} reduction increases by a factor of about 10 as the pH decreases from 5 to 6 down to about pH 3, in the presence of chloride. These

results suggest that Hg^0 re-emissions in full-scale wet FGD systems could be greatly influenced by factors such as chloride concentration and slurry droplet pH.

A chemical kinetics model has been developed to describe the aqueous Hg reactions in FGD systems. The model predicts the basic trends seen experimentally for pH, sulfite, thiosulfate, and chloride effects, and provides fairly good numerical agreement with experimental data. The model is currently being verified through a series of bench-scale wet FGD tests using simulated flue gas to determine if it accurately predicts the tendency for Hg^0 re-emissions, and the phase in which Hg is found in the FGD by-products. Preliminary results indicate that mercuric mono-chlorosulfite (ClHgSO_3^-) is a key species since the rate constant for decomposition of ClHgSO_3^- to Hg^0 is much smaller than that for the mercuric sulfite complex, thus contributing to the inhibiting effect of chloride on Hg^0 re-emissions. In addition, thiosulfate appears to inhibit Hg^0 re-emissions at low pH, but accelerate this phenomenon at high pH. The validated model will be run over a wide range of potential wet FGD operating conditions with and without the use of FGD additives to determine conditions that: (1) optimize FGD Hg capture; and (2) influence the phase in which Hg leaves in the FGD by-products.

Wet FGD Additives

URS also conducted pilot- and full-scale field tests of a wet FGD additive, Degussa Corporation's TMT-15, to determine whether the additive can precipitate absorbed Hg as a stable salt, thereby minimizing Hg^0 re-emissions and lowering FGD liquor Hg concentrations.⁹ This project is also assessing whether this same additive can be used to minimize Hg in reused FGD solid by-products, through separation of the fine Hg-containing salts from the remainder of the byproduct. The project included pilot-scale testing at MoSES Unit 3 and Plant Yates Unit 1. Full-scale field tests have been completed at Indianapolis Power & Light's Petersburg Station Unit 2, which fires high-sulfur bituminous coal, and Plant Yates.

The effectiveness of Degussa Corporation's TMT-15 additive in suppressing Hg^0 re-emissions was inconclusive at pilot-scale due to: (1) the absence of re-emissions, even without chemical addition, at Monticello Station; and (2) Hg measurement issues at Southern Company's bituminous-fired Plant Yates. However, TMT-15 had the anticipated impact on FGD by-products as the FGD liquor Hg concentrations were significantly reduced during both tests. During a full-scale field test at Indianapolis Power & Light's Petersburg Station, which burns high-sulfur bituminous coal, a modest decline in Hg^0 emissions was observed during an eight-day TMT-15 injection test, but the additive did not impact the partitioning of Hg in FGD by-products at this site. Meanwhile, full-scale results obtained during a 30-day evaluation of Nalco Company's 8034 additive at Plant Yates were confounded by low baseline Hg^0 re-emission levels.

A third wet FGD additive, Babcock & Wilcox's Absorption Plus(Hg)TM, was evaluated at E.ON America's high-sulfur bituminous-fired Mill Creek Station after parametric trials revealed that untreated ACI had little, if any, impact on Hg removal.¹⁰ During long-term testing, total Hg removal averaged about 92% with the addition of Absorption

Plus(Hg)TM. Note that over 80% total Hg removal was observed under baseline conditions.

Sorbent Injection

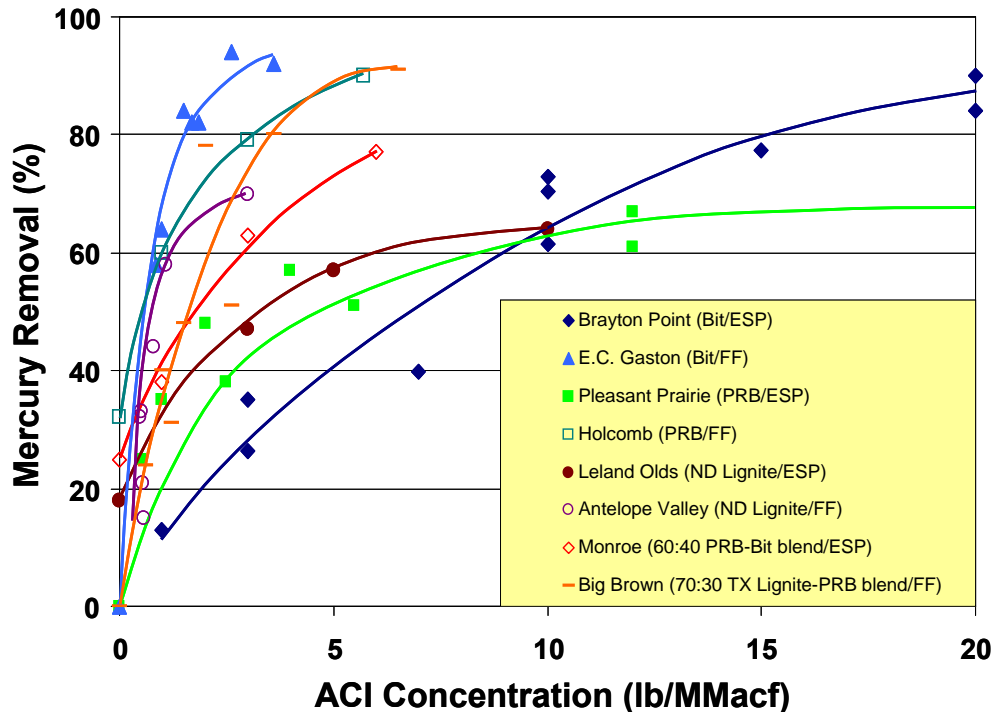
NETL's Phase II portfolio of full-scale field testing projects encompass several variations of sorbent injection technology: untreated ACI, untreated ACI with chemical additives, chemically-treated ACI, EPRI's TOXECONTM configuration, TOXECON IITM, as well as non-carbon and "concrete-friendly" PAC sorbents. The development, and subsequent field testing, of chemically-treated ACI and chemical additives, in conjunction with untreated ACI, represents a concerted effort to enhance Hg capture at units firing low-rank coal after Phase I results at the PRB-fired Pleasant Prairie Unit 2 showed total Hg removal via untreated ACI was limited to about 65%.¹¹ These advanced Hg-specific control technologies are designed to introduce excess halogens into the Cl-deficient flue gas emitted from low-rank coals to promote Hg⁰ oxidation and adsorption. Meanwhile, minimizing the impact of ACI on fly ash utilization is the driving force behind Phase II field testing of the TOXECONTM configurations and non-carbon/"concrete-friendly" sorbent injection.

Untreated PAC

In addition to serving as a benchmark during nearly all of NETL's Phase II full-scale field tests, untreated ACI has also been evaluated during two 30-day long-term tests. At Plant Yates Unit 1, URS selected RWE Rhinebraun's Super HOK sorbent and total Hg capture varied from 50 to 86% with injection rates ranging from 4.5 to 9.5 lb/MMacf.¹² Plant Yates was selected for long-term testing, in part, to gain a better understanding of the effect of ACI on small-SCA ESP (173 SCA) and wet FGD operation. URS observed an increase in the ESP arcing rate during continuous ACI, particularly at high load. While the 30-day long-term injection test caused no visible physical damage to the ESP, it remains unclear what effect the increased arcing rate will have on ESP performance over longer time periods.

ADA-ES chose DARCO[®] Hg for the 30-day long-term test conducted at DTE Energy's Monroe Station Unit 4, which burns a 60% PRB and 40% bituminous coal blend and is equipped with an SCR and CS-ESP.¹³ Total Hg removal averaged 78% with DARCO[®] Hg injection at 4.9 lb/MMacf. The performance of untreated DARCO[®] Hg during select Phase I and II full-scale field tests is presented in Figure 2.

Figure 2 – Phase I and II Performance Curves for Untreated ACI



* All data generated using NORIT Americas' DARCO® Hg

Chemically-Treated PAC

The limited Hg removal achieved by untreated ACI at Pleasant Prairie spurred the development and full-scale field testing of alternatives, such as PAC chemically-treated with bromine. Two brominated PACs – NORIT Americas' DARCO® Hg-LH and Sorbent Technologies' B-PAC™ - have consistently been top performers at Phase II field testing units burning lower-rank coals. In fact, the outstanding performance (see Figure 3) of these brominated sorbents has accelerated the commercialization of Hg-specific controls technologies and drastically reduced the estimated cost of Hg control due to a reduction in the ACI rate required to achieve a given level of control, which offsets the higher cost of these sorbents.

ADA-ES selected brominated DARCO® Hg-LH for 30-day long-term field tests at two PRB-fired units: Sunflower Electric's Holcomb Station Unit 1 and AmerenUE's Meramec Station Unit 2. At Meramec, 93% average total Hg removal was achieved across the CS-ESP with DARCO® Hg-LH injection at 3.3 lb/MMacf.¹⁴ Total Hg capture averaged 93% across the spray dryer absorber and fabric filter (SDA/FF) configuration at Holcomb with DARCO® Hg-LH injection at 1.2 lb/MMacf.¹⁵ UNDEERC also conducted a 30-day evaluation of DARCO® Hg-LH at GRE's ND lignite-fired Stanton Station Unit 10. With DARCO® Hg-LH injection at 0.7 lb/MMacf, total Hg capture across the SDA/FF configuration averaged 59%.¹⁶ However, greater than 90% Hg capture was achieved at this unit during parametric trials with both DARCO® Hg-LH and B-PAC™ injection at 1.5 lb/MMacf.

The brominated B-PAC™ sorbent has also been selected for 30-day long-term trials at three of NETL's Phase II field testing sites. At GRE's PRB-fired Stanton Station Unit 1, URS observed 85% average total Hg removal across the CS-ESP with B-PAC™ injection at 1.7 lb/MMacf.¹⁷ Sorbent Technologies conducted long-term field tests with B-PAC™ at DTE Energy's St. Clair Station Unit 1, which burns an 85% PRB and 15% bituminous coal blend, and Progress Energy's bituminous-fired Lee Station Unit 1. At St. Clair, 94% average total Hg removal was achieved across the CS-ESP with B-PAC™ injection at 3 lb/MMacf.¹⁸ Total Hg capture averaged 85% across the CS-ESP at Lee with B-PAC™ injection at 8 lb/MMacf.¹⁷ Note that the 30-day long-term test at Lee was conducted with the SO₃ FGC system idled and opacity levels remained acceptable.

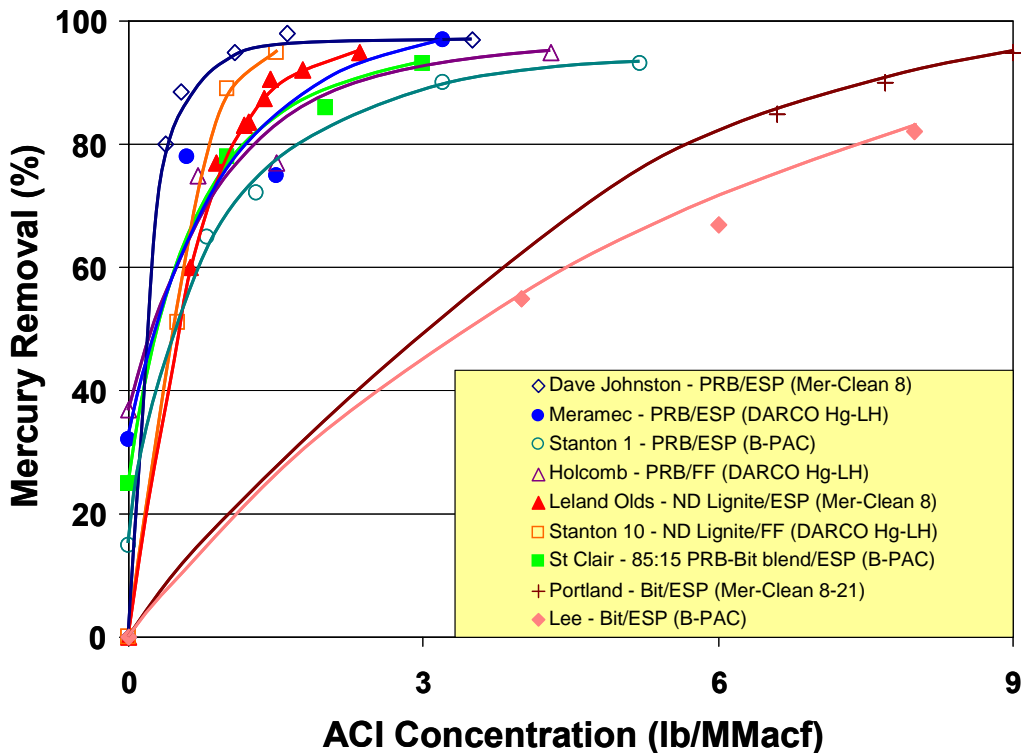
ADA-ES is conducting Phase III testing at Rocky Mountain Power's PRB-fired Hardin Generating Station to evaluate the Hg removal performance, long-term emissions variability, and associated operating and maintenance costs of PAC injection for greater than 90% Hg control during a ten- to twelve-month full-scale demonstration. Baseline Hg capture at Hardin ranged from 20 to 30% across the SCR and SDA/FF configuration.¹⁹ During parametric testing, an injection rate of about 1 lb/MMacf was required to attain slightly more than 90% total Hg removal with DARCO® Hg-LH and Calgon Carbon's brominated FLUEPAC™-MC Plus. In addition, injection of a DARCO® Hg and FLUEPAC™-MC Plus mixture achieved 90% total Hg at 0.14 lb/MMacf, with a low KNX™ additive rate. The long-term test began in September 2007 and will continue through August 2008 using DARCO® Hg-LH. At low load (80 to 90 MW), a DARCO® Hg-LH injection rate of 1.5 lb/MMacf is required to maintain 90% removal compared to 3.0 lb/MMacf at 120 MW. This is likely due the high fraction of Hg²⁺ produced at low load (up to 50%) that is captured in the SDA.

URS conducted Phase III field testing at NRG Texas Power LLC's Limestone Electric Generating Station Unit 1, which fires a 70:30 blend of TX lignite and PRB coals and is equipped with a CS-ESP and wet FGD. Baseline Hg removal was highly variable ranging from about 5 to 50%. Since this unit markets its fly ash for reuse, two Hg control technologies designed to preserve ash quality were evaluated during parametric tests: low-ash impact sorbent injection and TOXECON II™.²⁰ During injection upstream of the ESP, the brominated B-PAC™ and DARCO® Hg-LH sorbents performed similarly with about 90% ACI Hg removal at 2 to 3 lb/MMacf. Untreated DARCO® Hg also achieved 90% ACI Hg removal with injection at slightly less than 6 lb/MMacf. Injection of the "concrete-friendly" C-PAC™ sorbent at about 1.5 lb/MMacf resulted in approximately 73% ACI Hg removal. During parametric trials with the TOXECON II™ configuration, ACI Hg removal was limited to about 60% with DARCO® Hg and DARCO® Hg-LH injection at about 5 to 6 lb/MMacf. Note that DARCO® Hg-LH injection into the TOXECON II™ configuration took place with the unit firing 100% PRB coal. A two-month continuous injection test was completed with DARCO® Hg-LH injection at 2 lb/MMacf and preliminary results indicate that the project goal of 50 to 70% ACI Hg removal across the ESP was achieved. In addition, URS is confident that the low DARCO® Hg-LH injection rate will not prohibit fly ash reuse, but analysis is ongoing.

Another chemically-treated ACI technology, ALSTOM Power, Inc. – U.S. Power Plant Laboratories’ (ALSTOM-PPL) Mer-Cure™ process, is unique in that injection takes place in the high-temperature region upstream of the air preheater (APH) and the process employs a proprietary “processor” to prevent chemically-treated Mer-Clean™ sorbent agglomeration and ensure uniform sorbent dispersion.²¹ Three 30-day long-term field tests of Mer-Cure™ have been completed at Phase II sites equipped with a CS-ESP: (1) PacifiCorp’s PRB-fired Dave Johnston Unit 3; (2) Leland Olds Unit 1; and (3) Reliant Energy’s medium-sulfur (2%) bituminous-fired Portland Station Unit 1. Chemically-treated Mer-Clean™ 8 injection rates of 0.63 and 1.4 lb/MMacf achieved average total Hg removals of 92 and 90% at Dave Johnston and Leland Olds, respectively. At Portland, about 95% average total Hg capture was observed with chemically-treated Mer-Clean™ 8-21 injection at 8.5 lb/MMacf. The reduced efficiency of the Mer-Clean™ sorbents at Portland may have been caused by elevated levels of flue gas SO₃, resulting from the combustion of medium-sulfur bituminous coal.

A Phase III evaluation of Mer-Cure™ was completed at LCRA’s Fayette Unit 3 in April 2007.²² Baseline Hg capture was approximately 50% across the CS-ESP and wet FGD. Note that all results are based on the incremental (or ACI) level of Hg control. ALSTOM-PPL evaluated three sorbents (eSorb™ 11, eSorb™ 13, and eSorb™ 18) designed to preserve fly ash quality, along with Mer-Clean™ 8, during parametric testing. Excluding eSorb™ 18, 80% ACI Hg capture was achieved with injection at 0.4 to 0.5 lb/MMacf. At an injection at about 0.8 lb/MMacf, eSorb™ 11 and Mer-Clean™ 8 attained 90% ACI Hg capture. Preliminary results indicate that fly ash remains marketable with eSorb™ 13 at about 0.5 lb/MMacf (85% ACI Hg capture). The testing program was halted prematurely due to an unscheduled plant outage.

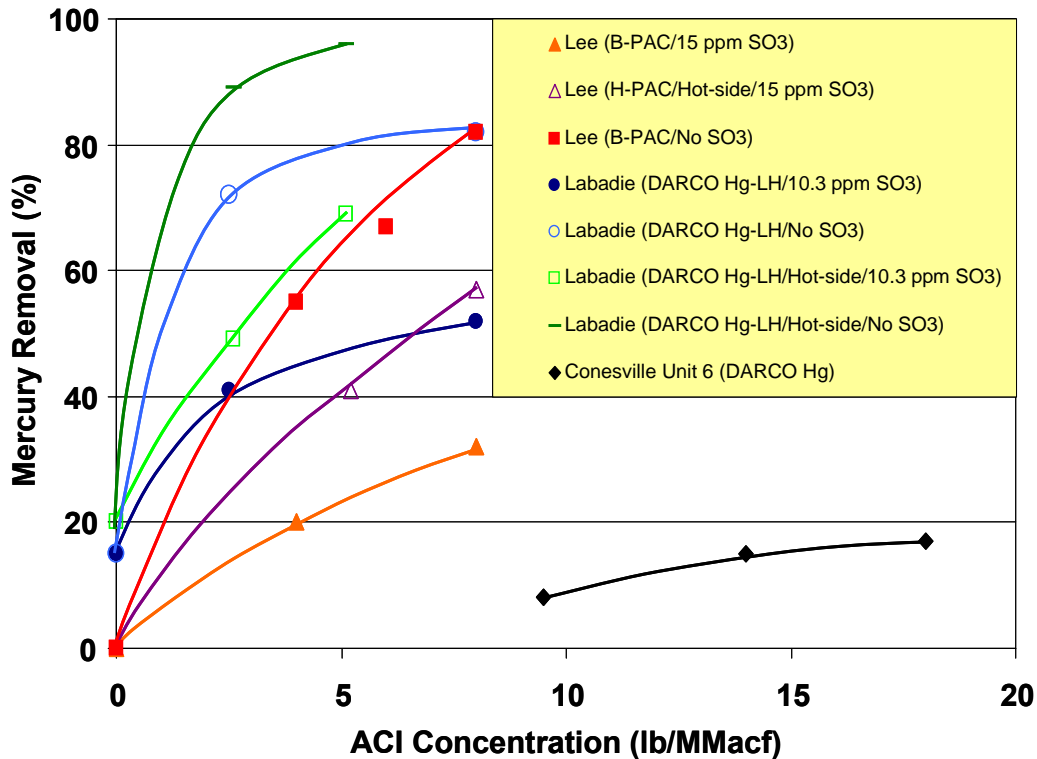
Figure 3 – Phase II Performance Curves for Chemically-treated ACI



SO₃ Impacts

NETL Hg control R&D has shown that flue gas SO₃, even at low concentrations, can interfere with the performance of ACI. SO₃ is generated in coal combustion flue gas via three mechanisms: (1) oxidation of SO₂ within the furnace; (2) further oxidation of SO₂ across SCR catalysts; and (3) SO₃ FGC systems. It appears that SO₃ competes with Hg for adsorption sites on the PAC surface thereby limiting its performance and/or requiring much higher ACI rates to achieve a given level of Hg control (see Figure 4).²³

Figure 4 - Impact of SO₃ on ACI Performance



During Phase II field testing at AEP's high-sulfur (3-4%) bituminous-fired Conesville Station Unit 6, ADA-ES evaluated over 50 candidate sorbents and total Hg removal was limited to approximately 30% with chemically-treated PAC injection at 12 lb/MMacf.²⁴ Consequently, a 30-day long-term field test was not conducted at this unit; instead, NETL funding was used to evaluate the impact of SO₃ FGC on brominated DARCO[®] Hg-LH at AmerenUE's PRB-fired Labadie Station.²⁵ As shown in Figure 4, turning the SO₃ FGC system off at Labadie increased total Hg removal from about 50 to 80% with DARCO[®] Hg-LH injection at 8 lb/MMacf. In addition, greater than 90% Hg removal was observed with no SO₃ injection and DARCO[®] Hg-LH injection upstream of the APH at about 5 lb/MMacf. The performance of brominated B-PAC[™] was also impacted by SO₃ FGC at Lee Station. With B-PAC[™] injection at 8 lb/MMacf, Hg capture increased from 32 to 82% when SO₃ FGC was idled at Lee. One possible solution to this problem is the dual injection of PAC and alkaline materials. Preliminary results from a few Phase II and III field testing sites are encouraging.

ADA-ES is conducting a Phase III field test at Public Service of New Hampshire Company's Merrimack Station Unit 2, which utilizes a cyclone-fired boiler to burn a blend of bituminous coals (1.0 - 1.3% sulfur content) and is equipped with an SCR system followed by two CS-ESPs in series.²⁶ This is a challenging environment for ACI due to elevated SO₃ levels and high flue gas temperature. During parametric testing, several Hg sorbents were evaluated both with and without the injection of magnesium oxide (MgO) or sodium sesquicarbonate (trona) – two potential SO₃ mitigation additives that also permit a reduction in flue gas temperature. Results indicate that trona injection enhanced ACI performance to a greater degree than MgO; however, the sodium content of trona may limit fly ash recycling opportunities.

Without SO₃ mitigation, Hg removal was limited to about 22% with brominated DARCO[®] Hg-LH injection between the two ESPs at 8 lb/MMacf. Untreated DARCO[®] Hg injection at 8 lb/MMacf, coupled with trona injection, resulted in about 65% Hg removal. During a continuous injection test completed in March 2008, 50% Hg removal was achieved with trona injection upstream of the APH at 500 lb/hr and DARCO[®] Hg-LH injection between the two ESPs at about 4 lb/MMacf.

Conventional PAC with Chemical Additives

As an alternative to using chemically-treated PAC, NETL also has sponsored field tests using conventional PAC supplemented with chemical additives applied to the coal and/or flue gas to overcome the poor performance of conventional PAC in low-rank coal applications. Through funding provided by NETL, UNDEERC conducted two 30-day long-term field tests at full-scale units firing ND lignite coal to determine whether SEA coal treatment enhances the performance of untreated ACI at units burning lower-rank coals.¹⁶ Indeed, SEA coal treatment improved the Hg capture efficiency of untreated ACI at both of these Phase II field testing sites. During the 30-day trial at Basin Electric's Leland Olds Station Unit 1, 58% average total Hg capture was observed across the CS-ESP with DARCO[®] Hg injection at 2.7 lb/MMacf, coupled with the addition of an aqueous CaCl₂ solution to the ND lignite coal at a rate of 2.9 lb/MMacf. UNDEERC also evaluated this advanced Hg-specific control technology at Basin Electric's Antelope Valley Station Unit 1. Total Hg removal averaged 92% across the SDA/FF configuration with the addition of SEA2 to the ND lignite coal at a rate of 0.033 lb/MMacf and DARCO[®] Hg injection at 0.81 lb/MMacf.

UNDEERC conducted additional Phase II Hg control field testing at Montana-Dakota Utilities Company's Montana lignite-fired Lewis & Clark Station that is equipped with a mechanical collector and wet venturi scrubber. Parametric tests evaluated the Hg capture efficiency of SEA1 and SEA2 addition to the coal with and without ACI upstream of the wet venturi scrubber.²⁷ With SEA1 injection at 600 ppm coal equivalent, about 90% Hg removal is achieved with untreated DARCO[®] Hg injection at 3 lb/MMacf. Slight improvements in performance were observed with higher SEA1 and DARCO[®] Hg injection rates. Ninety percent Hg removal was also observed with SEA2 injection at 100 ppm coal equivalent and DARCO[®] Hg injection at 1 lb/MMacf. Note that in the absence of PAC injection, Hg removal was limited to about 55% with SEA2 injection at 100 ppm

coal equivalent. A one-week, optimized field test was also conducted, but results are currently unavailable.

Under a Phase III award, UNDEERC is evaluating PAC and SEA injection, as well as, the SEA2 technique 2 (SEA2 T2) technology, which involves co-injection of the proprietary SEA2 additive and PAC upstream of the particulate control device. Full-scale field testing is currently on-going at Kansas City Power & Light's Hawthorn Unit 5 that burns PRB coal and is equipped with an SCR and SDA/FF configuration. During parametric testing at Hawthorne, greater than 90% total Hg capture was achieved with SEA1 added to the coal at 1200 ppm and DARCO[®] Hg-LH injection at 3 lb/MMacf.²⁸ With the SEA2 T2 technology, the co-injection of DARCO[®] Hg at 2.8 lb/MMacf and SEA2 at 0.14 lb/lb of PAC, resulted in about 85% total Hg removal. In addition, >90% Hg removal was achieved with SEA1 added to the coal at ~500 ppm and DARCO[®] Hg injection at 2 lb/MMacf. A 45-day long-term is scheduled for Hawthorne where UNDEERC will further evaluate SEA1 addition to the coal, coupled with untreated DARCO[®] Hg injection upstream of the ESP.

ACI Upstream of a Hot-Side ESP

NETL is also evaluating Hg control technologies designed specifically for hot-side ESP (HS-ESP) applications, where the elevated flue gas temperature limits the Hg capture efficiency of ACI. A four-day trial conducted at Duke Energy's low-sulfur bituminous coal-fired Buck Plant achieved approximately 70% total Hg removal with the injection of Sorbent Technologies' chemically-treated H-PACT[™] at 10 lb/MMacf.¹⁸ Sorbent Technologies conducted additional field testing at Midwestern Generation's PRB-fired Will County Unit 3. A high temperature version of the brominated, "concrete-friendly" C-PACT[™] sorbent was evaluated since fly ash from this unit is marketed for reuse. Using a newly developed X-a-Lance distributing lance design, 73% Hg removal was achieved during a parametric trial with C-PACT[™] injection at 5 lb/MMacf.²⁹ During a six-day continuous test, Hg removal ranged from about 60 to 73% with C-PACT[™] injection at 5 lb/MMacf.

Under a separate Phase II project, ADA-ES evaluated the impact of adding high-temperature liquid sorbents to the pre-combusted coal and/or upstream of the HS-ESP on Hg control at MidAmerican's PRB-fired Louisa Station Unit 1.³⁰ While Alstom's brominated KNX[™] coal additive promoted Hg⁰ oxidation, the lack of a downstream FGD at this unit led to no increase in Hg removal.

Alternative Sorbent Injection Technologies

The typical ACI system is located upstream of a particulate control device to enable simultaneous capture of the spent PAC and fly ash. This Hg control strategy leads to commingling of the PAC and fly ash that can prohibit certain fly ash recycling efforts. In particular, fly ash collected at coal-fired units that employ sorbent injection for Hg control is banned from serving as a feedstock at cement kilns following a December 2006 final rule issued by EPA.³¹ Note that this ruling is based on the potential for increased Hg emissions at cement kilns rather than carbon contamination concerns. In 2006, over 4

million tons of fly ash served as alternative feedstock to shale or clay at about 34 U.S. cement kilns.³²

Meanwhile, one of the highest-value reuse applications for fly ash is use as a substitute for Portland cement in concrete production. The utilization of fly ash in concrete production is particularly sensitive to carbon content as well as the surface area of the carbon present in the fly ash. PAC injection will increase the carbon content of the fly ash with the degree of carbon contamination dependent upon the ACI concentration required to achieve a given level of Hg control. In addition, PAC has a high surface area that is ideal for Hg capture, but also promotes the adsorption of surfactants known as air entraining admixtures (AEA) that are added to the concrete slurries to stabilize an optimum amount of air in the concrete product, thus improving its workability and durability to freeze-thaw cycles.^{33,34} The adsorption of AEA by the injected PAC will lead to an increased Foam Index value, which refers to the quantity of AEA required to saturate the fly ash and cement mixture, resulting in an inferior concrete product. Furthermore, the association of fly ash with Hg capture may influence marketability simply due to a perceived connection with the hazards of Hg. As a result, NETL's Phase II Hg control technology field testing program includes evaluations of alternative sorbent injection technologies designed to minimize, or completely eliminate, fly ash carbon contamination caused by ACI upstream of a particulate control device.

TOXECON™ Configuration

Based on the promising Phase I results at E.C. Gaston (see Figure 5), TOXECON™ was selected for a first-of-a-kind commercial Hg control technology demonstration at WeEnergies' Presque Isle Power Plant in Marquette, Michigan, under DOE's Clean Coal Power Initiative. Currently operational, the TOXECON™ configuration has achieved about 90% total Hg removal with untreated DARCO® Hg and brominated DARCO® Hg-LH injection at about 1.7 and 1.2 lb/MMacf, respectively.³⁵ During an extended testing period, greater than 90% total Hg removal was maintained for 48 consecutive days with both DARCO® Hg and DARCO® Hg-LH injection.

Under the Phase II program, NETL has also funded a full-scale field test of the TOXECON™ configuration at Luminant Energy's Big Brown Unit 2, which fires a 70% TX lignite and 30% PRB coal blend.³⁶ UNDEERC evaluated the performance of untreated ACI, co-injection of SEA and untreated PAC, and UNDEERC's proprietary enhanced PAC during parametric tests. Due to concerns about the cumulative impact of SEA and PAC injection on differential pressure across the relatively small FF (air-to-cloth ratio of 12:1), UNDEERC's enhanced PAC was selected for the 30-day long-term demonstration and total Hg capture average about 74% with an injection rate of 1.5 lb/MMacf.

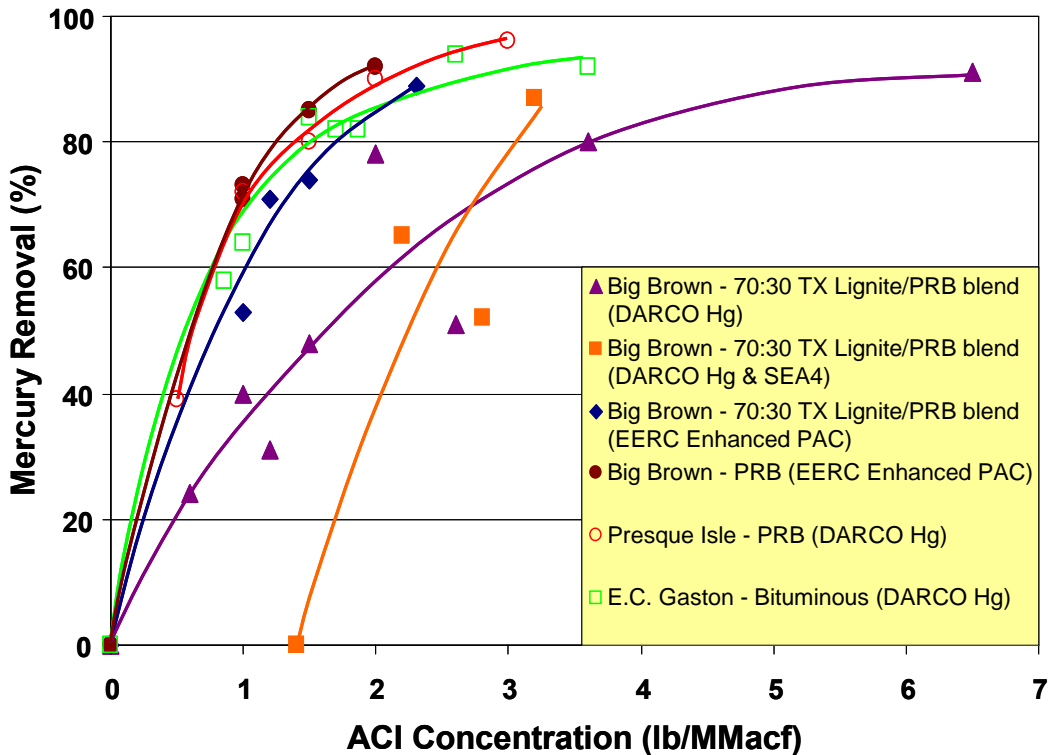
According to an in-depth BOP analysis performed by UNDEERC, enhanced PAC injection at 1.5 lb/MMacf increased the pressure drop across the FF at Big Brown by about 1" H₂O at high load (~600 MW). Handling and storage issues with the PAC/ash mixture have also been observed at both Presque Isle and Big Brown. In particular, a portion of the PAC/ash mixture was found to be very hot and smoldering at each unit and

preliminary results indicate that plant personnel will need to monitor and empty the FF hoppers on a regular basis to avoid self-heating and ignition of the PAC/ash mixture.

TOXECON II™ Configuration

EPRI's TOXECON II™ technology injects sorbents directly into the downstream collecting field(s) of an ESP. Since the majority of fly ash (~90%) is collected in the upstream ESP fields, only a small portion of the total collected ash contains spent sorbent. The technology requires minimal capital investment compared to the TOXECON™ configuration, because a retrofit FF is not required. A full-scale TOXECON II™ field test was conducted by ADA-ES at Entergy's PRB-fired Independence Station Unit 1.³⁷ The CS-ESP at Independence contains four electric fields (542 SCA) and ACI took place between the second and third fields. During a long-term field test in November 2005, about 60% average total Hg removal was observed with DARCO® Hg-LH injection at 4 to 5 lb/MMacf. While the project goal of 50 to 70% Hg capture was attained, ADA-ES felt that performance was limited by the injection lance design. During subsequent full-scale field testing at Independence in February 2007, DARCO® Hg-LH injection at 5.5 lb/MMacf achieved 90% total Hg removal with ADA-ES' new TOXECON II™ lance design. A remaining concern with any Hg control strategy involving sorbent injection, particularly the TOXECON II™ technology that limits the ESP residence time, is the potential for increased particulate emissions that could trigger New Source Review requirements.

Figure 5 – Phase I and II TOXECON™ Performance Curves



Non-carbon & “Concrete-friendly” PAC Sorbents

NETL is funding full-scale field tests of alternative Hg sorbents that would allow coal-fired power generators to continue marketing fly ash commingled with the spent sorbent as a suitable replacement for Portland cement in concrete. Two approaches are discussed below: non-carbon based sorbent injection and injection of PAC that is passivated during production to limit AEA adsorption.

The performance of Amended Silicates’ non-carbon Amended Silicates™ sorbent (comprised of a chemically-amended silicate substrate) was evaluated during a 30-day long-term field test at Duke Energy’s medium-sulfur (~2.3%) bituminous-fired Miami Fort Unit 6.³⁸ Total Hg capture across the CS-ESP averaged 40% with Amended Silicates™ injection at 5 to 6 lb/MMacf. Once again, flue gas SO₃ may have had a detrimental effect on sorbent performance at Miami Fort.

Under a separate Phase II project, Sorbent Technologies conducted a 30-day long-term evaluation of their brominated, “concrete-friendly” C-PAC™ at Midwestern Generation’s PRB-fired Crawford Station Unit 7.³⁹ With C-PAC™ injection at about 4.6 lb/MMacf, total Hg removal across the small CS-ESP averaged 81%. Most importantly, preliminary results indicate that fly ash samples collected during sorbent injection at these units would satisfy the criteria used to determine the suitability for reuse in concrete production.

Novel Hg Control Concepts

Innovative techniques for Hg control that could eventually replace and/or augment the more mature technologies discussed above are also being explored under the IEP Program. The following is a brief discussion of these NETL-funded efforts.

MerCAP™

The Hg control via adsorption process (MerCAP™), developed by EPRI, relies on fixed structure sorbents positioned in the flue gas stream to adsorb Hg and then, as the sorbent becomes saturated, regenerate the sorbent and recover the Hg. An initial retrofit application of the MerCAP™ technology is for “polishing” control of Hg⁰ downstream of FGD systems. During two six-month extended pilot-scale tests, the performance of gold-coated MerCAP™ plates was evaluated downstream of a: (1) spray dryer adsorber and fabric filter (SDA/FF) configuration at Great River Energy’s Stanton Station Unit 10; and (2) wet FGD system at Plant Yates Unit 1.⁴⁰

After more than 6,000 hours of continuous operation at Stanton Station, Hg removal averaged 30 to 35% across the acid-treated MerCAP™ plates and 10 to 30% across the untreated plates. Testing also revealed that regeneration via acid treatment and tighter plate spacing (½-inch vs. 1-inch) improved the Hg capture efficiency of the MerCAP™ technology. At Plant Yates, Hg removal decreased from 15 to 3% during the first three days of pilot-scale MerCAP™ operation. It was believed that limestone slurry carryover from the FGD system was inhibiting Hg reactions. Subsequent use of a water wash system for the plates was able to restore Hg removal to 15%.

Utilization of Partially Gasified Coal for Mercury Removal

GE EER will evaluate the Hg capture efficiency of sorbents produced from coal in an *in-situ* gasification process at coal burning plants. Proposed Phase III work will optimize the gasification process to maximize sorbent reactivity while minimizing sorbent production costs.⁴¹ Optimization will be conducted with respect to coal type, gasification process parameters, and the sorbent injection rate required to achieve at least 70% Hg removal. Among the gasification process parameters to be optimized are: (1) composition of solid fuel/air mixture, (2) temperature, and (3) mixture residence time. GE EER will also evaluate the stability of captured Hg and the potential impact on fly ash marketability.

Low Temperature Mercury Capture Process

CONSOL Energy, Inc. (CONSOL) will conduct a Phase III field test of the Low Temperature Mercury Capture (LTMC) process at a bituminous-fired power plant that is equipped with a CS-ESP. LTMC has the ability to reduce Hg emissions by over 90%, as was recently shown by CONSOL on a slip-stream pilot plant at Allegheny Power's Mitchell Station. The LTMC process controls Hg by cooling the flue gas temperature to about 220°F, which promotes Hg adsorption on the UBC inherent in fly ash. To avoid corrosion at the low-temperature conditions, the SO₃ concentration will be controlled through magnesium hydroxide slurry injection. The project will also demonstrate that water spray humidification can maintain ESP performance under low-SO₃ conditions. A two-month test will be conducted to evaluate long-term performance and any potential balance-of-plant impacts.

Pilot Testing of WRI's Novel Mercury Control Technology

Western Research Institute (WRI) will develop and evaluate pre-combustion Hg removal of raw coal by thermal treatment.⁴² Key process steps in the WRI technology include treating the fuel at two selected temperature windows. In the first stage, the moisture in the fuel is driven-off; in the second stage, coal is heated by nearly inert gas resulting in significant removal of coal-bound Hg. Bench-scale testing has revealed that residence time is an important parameter for Hg release in some coals - an increase of 8 minutes of residence time results in almost 80% of Hg released from coal. The percentage of Hg released from the coals varied from 50 to 87% depending on residence time. In addition, initial results from a fixed-bed test unit indicate that high temperature sorbents will be available to remove Hg from the process recycle sweep gas in the temperature range of 550 to 600°F. Pilot-scale testing (100 lb/hr) is currently being conducted to assess and scale-up results from the bench-scale tests. The pilot unit will examine two different Hg removal configurations: a vibratory fluid bed, and a proprietary vertical reactor.

NETL in-house development of novel control technologies

After studying numerous sorbents for Hg capture in simulated coal-derived gases, scientists at NETL discovered and patented three trace metal capture technologies that are now licensed and in commercial demonstration. The Thief process, licensed to Nalco-Mobotec USA, is a cost-effective alternative to ACI for Hg removal from flue gas as Thief carbon sorbents range from \$90 to \$200 per ton. The Photochemical Oxidation (PCO) process, licensed to Powerspan Corporation, introduces a 254-nm ultraviolet light into the flue gas, leading to enhanced Hg oxidation and capture. NETL researchers

received the 2005 Award for Excellence in Technology Transfer from the Federal Laboratory Consortium (FLC) for the PCO method.

Recognizing the need for a low-cost technique to remove Hg from coal-based Integrated Gasification Combined Cycle power plants, NETL researchers have invented a new sorbent that works on fuel gas at elevated temperatures. The new sorbent is palladium (Pd) on an inert support. Unlike conventional sorbents such as activated carbon, which operate at ambient temperature, high temperature Pd sorbents remove Hg and arsenic at temperatures above 500°F, and have more than twice the capacity of previously existing sorbents, resulting in a major improvement in overall energy efficiency of the power combustion process. NETL researchers received the 2008 Award for Excellence in Technology Transfer from the FLC for developing the Pd-based Hg sorbents licensed to Johnson Matthey.

Economic Analysis of ACI

NETL published an updated economic analysis of Hg control via ACI at 12 of the Phase II ACI field testing sites.⁴³ The May 2007 report provides “study-level”, plant-specific cost estimates and was carried out to provide NETL a gauge in measuring its success in achieving the target of reducing the baseline (1999) Hg control cost estimate of \$60,000/lb Hg removed by 25 to 50%. As shown in Figure 6, 20-year levelized cost estimates for the incremental cost of Hg control (\$/lb Hg removed) were calculated for 90% ACI Hg removal at seven Phase II field testing sites. Note that chemically-treated ACI was the Hg control technology evaluated at each of these units.

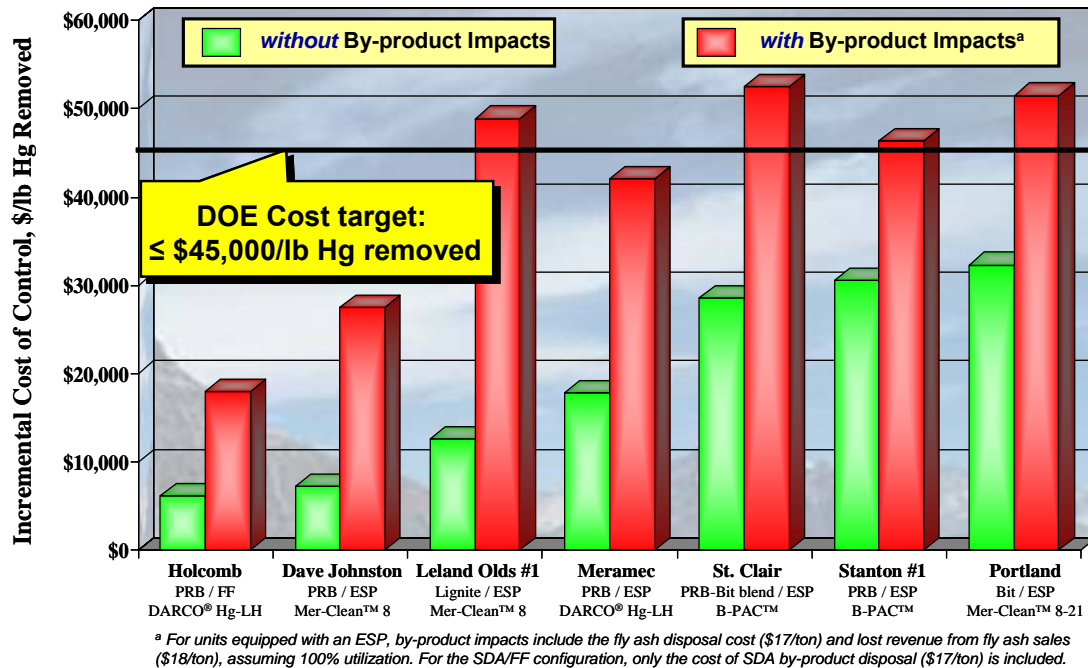
The Phase II field testing results are very encouraging both in terms of the level of Hg removal achieved and the estimated cost of control on a 20-year levelized basis. Specifically, the economics of Hg control via chemically-treated ACI at units burning lower-rank PRB and lignite coals is noteworthy. The 20-year levelized incremental increase in cost of electricity (COE) for 90% ACI Hg removal remains below 1.30 mills per kilowatt-hour (mills/kWh) for the four PRB units (Holcomb, Meramec, Stanton Unit 1, and Dave Johnston), as well as the units firing a PRB/bituminous blend (St. Clair), and ND lignite (Leland Olds), when CUB impacts are excluded.^b For comparison, the increase in COE calculated for 90% ACI Hg removal at the medium-sulfur bituminous-fired Portland Station is over 1.90 mills/kWh, when CUB impacts are excluded. Meanwhile, the incremental cost of 90% ACI Hg removal ranges from less than \$10,000 to about \$30,000/lb Hg removed, when CUB impacts are excluded.

The increase in COE resulting from Hg control via ACI is primarily determined by annual PAC consumption costs that are dependent on the required ACI rate, delivered PAC price, and the volume of flue gas being treated. Chemical composition also affects PAC price since manufacturers charge a higher price for chemically-treated PAC to offset the additional production costs required to alter the sorbent’s molecular structure. The

^b As shown in Figure 6, the economic analysis also included an assessment of the potential for ACI to negatively impact the sale and disposal of CUB (fly ash) and therefore impact the overall cost of Hg control.

ACI rate required to achieve a given level of Hg control can be impacted by a host of plant-specific dynamics, including, but not limited to: chlorine and sulfur contents of the coal being burned, APCD configuration, flue gas temperature, boiler efficiency, UBC content of the fly ash, and ductwork geometry in proximity to the ACI location. The impact of PAC manufacturing location on delivered sorbent price has taken on added significance following the U.S. Department of Commerce’s decision to impose tariffs, ranging from 62 to 228%, on Chinese activated carbon manufacturers.⁴⁴ The February 26, 2007 determination responds to concerns that Chinese manufacturers were dumping activated carbon at less than the fair U.S. market value.

Figure 6 – 20-Year Levelized Incremental Cost of 90% ACI Hg Control



The cost on a \$/lb Hg removed basis is impacted largely by the level of “co-benefit” Hg capture exhibited by the existing APCD configuration and coal Hg content. For example, the incremental cost of Hg control will increase when: (1) “co-benefit” Hg capture is high; and/or (2) coal Hg content is low, because a smaller quantity of Hg is removed from the flue gas for a given level of control.

Additional factors can influence the cost of Hg control, including: economic factors (labor rate, taxes and contingencies, economic life of capital equipment, etc.), process disruptions (unexpected or excessive outages, etc.), proximity to a reliable PAC manufacturer, and modifications to existing equipment. The estimates developed here assume an uncomplicated retrofit and minimal economic impact due to the installation of the ACI system, assuming that the installation occurs during a regularly scheduled plant outage. The estimates are also based on the assumption that Hg control via ACI will not cause any BOP impacts. In addition, the potential demand for a significant quantity of ACI systems within a relatively short timeframe, to ensure nationwide compliance with CAMR and the patchwork of state-level regulations, could place a strain on qualified

engineers, skilled laborers, and the raw materials required to erect both retrofit and new PAC storage and injection systems.

Technology Commercialization

Although the Federal regulatory structure for Hg emissions from coal-fired power plants is once again uncertain following the vacatur of EPA's Clean Air Mercury Rule on February 8, 2008,⁴⁵ NETL's field testing program has successfully brought Hg control technologies to the point of commercial-deployment readiness. As of April 2008, nearly 90 full-scale ACI systems, a signature technology of the IEP Program, have been ordered by U.S. coal-fired power generators.⁴⁶ These contracts represent over 44 gigawatts (GW) of coal-fired electric generating capacity. This includes approximately 33 GW of existing capacity (~10% of total U.S. coal-fired capacity) that will be retrofit with ACI systems to control Hg emissions. The ACI systems have the potential to remove more than 90% of the Hg in most applications, at a cost that can dip below \$10,000/lb Hg removed. Although the results achieved during NETL's field tests met or exceeded program goals, site-specific Hg characterization and testing may be required to evaluate alternative methods and their Hg capture efficiency on individual power plant generating units.

COAL UTILIZATION BY-PRODUCTS R&D PROGRAM

In addition to developing cost-effective control technologies for coal-fired power generation facilities, NETL's Hg research also focuses on CUB characterization.⁴⁷ CUB research is in response to the transfer of Hg and other trace metals from flue gas to the solid and liquid effluent streams as a result of implementing pollution controls on coal-based power systems, and driven by the goal of increasing the overall beneficial use of CUB. At least partly attributable to NETL's research efforts, CUB beneficial utilization has increased from 25% in 1990 to over 43% in 2006, while annual CUB production in the United States has risen from about 80 to nearly 125 million tons over this time period.³² NETL's overall goal is to increase CUB reuse to 50% by 2010, and nearly 100% by 2020.

CUB are currently regulated as non-hazardous under the Resource Conservation and Recovery Act, and it is expected that they will continue to be regulated as such. However, EPA has left the door open for future review of the regulatory status of CUB because of continuing concern over the fate of Hg and other trace metals in by-products. This concern is heightened in light of the deployment of Hg capture technology under CAMR. In response, NETL has sponsored a number of projects that focus on an evaluation of the potential leaching and volatilization of Hg and other trace metals from CUB, particularly FGD by-products, such as synthetic gypsum, and fly ash.

Determining the Fate of Hg in FGD By-products

The use of FGD as a Hg capture technology highlights the importance of better understanding the chemistry of Hg and FGD solids and synthetic gypsum products in order to predict the environmental fate of the CUB-bound Hg. The following NETL-

sponsored research is examining the mobility and ultimate fate of Hg in FGD by-products.

NETL's Office of Research and Development (NETL/ORD) is conducting an in-house research effort directed at determining the fate of Hg in FGD materials.⁴⁸ This activity focuses on Hg stability during FGD gypsum drying, Hg stability during wallboard production using FGD gypsum, Hg leachability from FGD gypsum, and the Hg-binding phase in FGD gypsum. The stability of Hg during FGD gypsum drying was studied by collecting samples before and after a natural gas-fired heating unit, which reduces the moisture content of the FGD solids for ease of handling during the wallboard manufacturing process. Results indicated that within analytical precision, no Hg desorbed during drying of free moisture.

The stability of Hg in FGD gypsum during wallboard production was analyzed by NETL/ORD by collecting samples of FGD gypsum feedstock and the corresponding wallboard products from five wallboard manufacturing plants. The Hg present in the unprocessed FGD gypsum and the finished wallboard product ranged from 0.04 to 1.5 ppm on a dry basis. The quantity of Hg retained in the finished wallboard product varied, with three samples showing nearly complete Hg retention during the wallboard manufacturing process. For the other two samples, Hg losses were 12 and 58%, suggesting that the quantity and thermal stability of Hg in FGD gypsum and wallboard depends on the origin of the gypsum and/or the nature of processing.

NETL/ORD also conducted settling and leaching experiments using FGD slurry samples in an attempt to isolate the Hg-binding phase from bulk gypsum. During settling, Hg partitioned almost exclusively to the top, slower-settling layer of the FGD slurry. Analysis of this residue revealed that both Hg and iron (Fe) were enriched in the top layer by factors of about 20 and 10, respectively. Meanwhile, Hg was not mobilized during FGD slurry leaching experiments using a continuous, stirred-tank extractor. This is indicative of a strong chemisorption rather than physical adsorption of Hg. As a result, it is believed that Hg sequestered in FGD gypsum is primarily bound to an iron-rich phase, such as iron-coated clay materials or iron oxide/hydroxide particles, probably introduced with the limestone used as the FGD reagent. Additional research⁴⁹ has also shown that some Hg retained in FGD gypsum requires hydrogen peroxide oxidation for release, such as a phase containing sulfides or carboneous materials. Mercury not bound in either of these two fractions is likely a precursor to Hg⁰ that could form via mechanisms being investigated through URS' bench-scale kinetics model.

Leach testing is also being used to evaluate the impacts of pH and oxidation-reduction potential (ORP) on Hg mobility in FGD byproducts. Leaching of six FGD materials to pH > 2 under aerobic conditions failed to mobilize appreciable amounts of Hg. While no Hg was released to the leachate during experiments with pH > 4 and ORP > 100 millivolts (mV), dissolution of the major immobilized form of Hg was complete with pH < 1 and an ORP of approximately 350 mV. Consequently, NETL/ORD researchers concluded that the Hg-retaining phase will immobilize Hg in many reuse applications,

with one possible exception being placement in mine-land reclamation areas where the FGD by-products could be exposed to acidic, anaerobic conditions.

The extramural thrust of NETL's CUB research is focused on the fate of Hg in FGD gypsum during wallboard production and ultimate disposal. For instance, the Tennessee Valley Authority (TVA) evaluated Hg retention in FGD gypsum from three sources: a Hg-amended commercial calcium sulfate, a laboratory-scale wet FGD system, and a full-scale wet FGD system.⁵⁰ TVA researchers concluded that no Hg would evolve from disposed FGD byproducts up to a maximum temperature of 140°F, although thermal desorption of Hg from FGD gypsum did occur at approximately 400°F.

USG Corporation is providing data on the extent and location of Hg loss during the wallboard production process, and also providing information on the potential for Hg leaching at the end of the wallboard life-cycle, when it is disposed in municipal landfills.⁵¹ FGD gypsum evaluation tests from six different power plant/FGD feedstock variations have been completed to investigate the impact of different configurations on the stability of Hg during wallboard production. Testing included the use of FGD gypsum produced during TMT-15 injection into a power plant scrubber.

Results to date indicate that use of fines blowdown in wet FGD systems significantly reduced Hg content in the FGD gypsum. For FGD gypsum generated without fines blowdown, Hg loss amounted to less than 8%, while tests using FGD gypsum from power plants employing fines blowdown indicated Hg loss of 46 to 55% during wallboard production.

Determining the Fate of Hg in Fly Ash

Beginning with the Phase I full-scale field testing program, NETL has required that field contractors evaluating Hg control via sorbent injection collect and analyze fly ash samples. Fly ash analyses are focused on determining the stability and ultimate fate of Hg during potential utilization applications and disposal. More recently, NETL awarded a contract to Frontier Geosciences, Inc. (Frontier) to conduct independent laboratory analysis of CUB generated during NETL's Phase II full-scale Hg control technology field testing program.⁵² The purpose of the independent laboratory analysis is to ensure accurate and consistent laboratory procedures are used to determine the environmental fate of Hg in CUB. NETL/ORD has also been conducting in-house leaching experiments with fly ash collected from ACI field testing sites.

The Frontier work includes leaching studies using the Synthetic Precipitation Leaching Procedure (SPLP, EPA Method 1312), low- (40°C for 30 days), medium- (190°C for 1 hour), and high-temperature (900-1200°C for 5 minutes) Hg volatility tests, microbial methylation experiments, and halide analysis. Preliminary SPLP results indicate that little to no Hg would be released under normal disposal conditions. In addition, Hg bound to PAC sorbents, particularly those that have been chemically-treated, appears to be more stable than the UBC-bound Hg. During the low-temperature volatility tests, essentially no Hg was emitted from the fly ash samples. Thermal desorption of Hg has been observed

during the medium- and high-temperature volatility tests conducted by Frontier; however, the extent of release is still under investigation.

Using a pure culture of sulfate reducing bacteria known to methylate Hg, the production of methyl-mercury, over a 30 day period, is being monitored to assess the methylation potential of Hg present in CUB. Preliminary results from this “worst-case-scenario” microbial mobilization study indicate an increase in methyl-mercury production. However, microbial activity has also stabilized a number of target metals.

NETL/ORD researchers have also conducted leaching experiments on the Phase II by-products using the modified SPLP, the NETL Serial Batch Leaching Procedure (SBLP), and NETL Column Leaching on a select number of sample pairs. During a five-month continuous column experiment using four leachants: water (pH=5.7), dilute sulfuric acid (pH=1.2), dilute acetic acid (pH=2.9), and sodium carbonate (pH=11.1),⁵³ the PAC/ash mixtures were generally found to effectively immobilize the captured Hg over a range of laboratory conditions. Overall, very little of the Hg (always below 0.5% and often under 0.1%) contained in the ash samples was solubilized during leaching. Neither the pH nor the nature of the anion had a noticeable effect on the leachate mercury.

Not only have the Hg control technologies demonstrated capture of Hg that would otherwise be released into the environment, but the Hg has generally been shown to be retained in the control technology by-products under conditions of laboratory leaching tests. For some of these materials, the tests performed in this study show these control technology ashes, in spite of their higher Hg content, to be environmentally more stable with respect to Hg release than the corresponding baseline ashes of lower Hg content.⁵⁴

SUMMARY

Over the past seven years, NETL has managed full-scale field tests of Hg control technologies at nearly 50 U.S. coal-fired power generation facilities. The flexible nature of this program allowed NETL to quickly incorporate insights and lessons learned from its network of partners into the development of advanced Hg control technologies tailored to specific areas of need. For instance, a determination that chlorine released during coal combustion promotes Hg⁰ oxidation in flue gas led to the development of technologies designed to provide a halogen “boost” for coals, such as subbituminous and lignite, that tend to contain low levels of chlorine and thus lower concentrations of the more reactive oxidized form(s) of Hg.

NETL has observed a step-change improvement in both the cost and performance of Hg control during full-scale field tests with chemically-treated (or brominated) ACI. The improved Hg capture efficiency of these advanced sorbent injection systems has given coal-fired power plant operators the confidence to begin deploying technology. As of April 2008, nearly 90 full-scale ACI systems have been ordered by U.S. coal-fired power generators. These contracts include both new and retrofit installations and represent over 44 GW of coal-based electric generating capacity. The ACI systems have the potential to remove more than 90% of the Hg in many applications based on results from NETL’s

field testing program, at a cost estimated to dip below \$10,000/lb Hg removed. However, while the results achieved during NETL's field tests met or exceeded program goals, only through experience gained during long-term continuous operation of these advanced technologies in a range of full-scale commercial applications will their actual costs and performance be determined.

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DISCLAIMER

References in this article to any specific commercial product or service are to facilitate understanding and do not imply endorsement by the U.S. Department of Energy.

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