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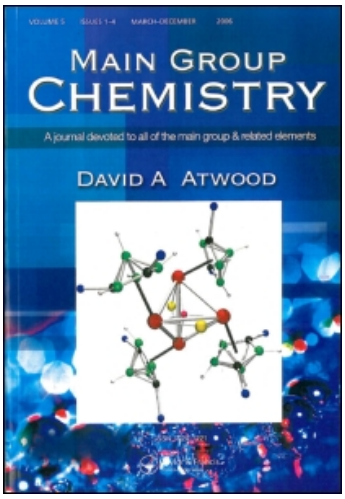

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Developing mercury control technology for coal-fired power plants – from concept to commercial reality

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The US Department of Energy's National Energy Technology Laboratory, under the Office of Fossil Energy's Innovations for Existing Plants (IEP) Program, has managed full-scale field tests of mercury (Hg) control technologies at nearly 50 US coal-fired power plants over the past 7 years. The high performance observed during many of these field tests, coupled with the reliability of Hg control system operations, has given coal-fired power plant operators the confidence to begin deploying technology. As of April 2008, nearly 90 full-scale activated carbon injection (ACI) systems, a signature technology of the IEP Program, have been ordered by US coal-fired power generators. These contracts represent over 44 gigawatts of coal-fired electric generating capacity. 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 per pound of Hg removed.

Keywords: mercury capture; activated carbon injection; chemically-treated; calcium bromide; oxidation; commercialization

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1. Introduction

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 mercury (Hg) because of its low concentration and reactivity during coal combustion. Although technical issues remain, the US Department of Energy's National Energy Technology Laboratory (NETL) has been successful, through public–private partnership, in significantly improving both the cost and performance of Hg control technology.

Under the Office of Fossil Energy's Innovations for Existing Plants (IEP) Program, NETL has carried out a comprehensive Hg research and development (R&D) program for coal-fired power generation facilities since the mid-1990s [1]. Working collaboratively with the US Environmental Protection Agency (EPA), the Electric Power Research Institute (EPRI), the University of North Dakota Energy and Environmental Research Center (UNDEERC), power plant operators, state and local agencies, and a host of research

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organizations and academic institutions, the IEP Programme has fostered the development of reliable measurement techniques for the different chemical forms of Hg. Through sampling and data analysis, the primary factors that affect Hg speciation and capture in coal combustion flue gas were identified, ultimately leading to the development of cost-effective Hg control technologies.

2. Experimental

The IEP Program initiated comprehensive Hg research in the 1990s to ensure that cost-effective and reliable pollution control technologies are available for the existing fleet of coal-fired power plants should regulations for controlling Hg be enacted. To comprehensively address the science of coal-fired Hg emissions, NETL has directed over $80 million in Federal funding over the last decade toward external and in-house research projects focusing on six inter-related research areas:

- Emissions characterization
- Development and testing of measurement devices
- Speciation research
- Development and field testing of control technologies
- Coal utilization by-products characterization; and
- Fate and transport of emissions.

The following focuses on the efforts directed toward understanding Hg speciation in coal combustion flue gas, and the development of Hg control technologies for coal-fired power plants.

2.1. Mercury speciation

Analysis of flue gas samples has 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 and unburned carbon, result in a portion of the Hg^0 being converted to gaseous oxidized forms of mercury (Hg^{2+}) and particulate-bound mercury (Hg_P) [2].

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 air pollution control device (APCD) configurations, which has been found to range from 0 to over 90% [3]. 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 higher percentage can be captured in wet flue gas de-sulfurization (FGD) systems, whereas the Hg^0 fraction is generally not captured by existing APCD. In addition, operation of a selective catalytic reduction (SCR) system for control of nitrogen oxides emissions has been shown to promote Hg^0 oxidation and enhance Hg capture across a downstream FGD [4].

Generally speaking, Hg speciation research spearheaded by NETL has revealed that: (1) several key factors influence Hg speciation in coal combustion flue gas; (2) Hg speciation impacts the level of Hg control achieved by existing APCD configurations; and (3) Hg capture across existing APCD configurations can be enhanced.

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2.2. *Mercury control technologies*

This knowledge was subsequently funnelled into the development of a suite of Hg control technologies for the diverse fleet of US coal-fired power plants. NETL initiated an R&D programme in the mid-1990s directed at two general approaches for controlling Hg (1) Hg^0 oxidation concepts that maximise co-benefit removal of Hg^{2+} in wet FGD systems and (2) Hg-specific control technology such as sorbent injection. In 2000, following laboratory through pilot-scale development of these approaches, NETL launched a three-phase field testing programme. This programme 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) focussed on untreated activated carbon injection (ACI) and improving the capture of Hg across wet FGD systems, whereas Phase II, which began in 2003, was expanded to include longer-term, full-scale field testing of chemically-treated ACI, sorbent enhancement additives, and sorbent-based technologies designed to preserve fly ash quality. Phase II also included evaluations of chemical additives and Hg^0 oxidation catalysts designed to enhance FGD Hg capture. 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 50–70% Hg capture at costs 25–50% less than the baseline (1999) estimate of about $60,000 per pound of Hg removed ($/lb Hg removed).

Although 30-day long-term tests were conducted in Phase II, the test period was not sufficient to answer all of the fundamental questions about long-term consistency of Hg removal and reliability of the system when integrated with plant processes. To assess potential balance-of-plant impacts associated with continuously operating an Hg-specific control technology for several months to years, NETL awarded nine new projects in 2006 to conduct longer duration Hg control tests of mature technologies at full-scale coal-fired units, as well as further laboratory and bench-scale development of novel Hg capture concepts. The Phase III projects support the IEP Programme's longer-term goal of developing advanced Hg control technologies (available for commercial demonstration by 2010) that could achieve at least 90% capture at costs 50–75% less than $60,000/lb Hg removed.[1]

3. Results and discussion

Over the past seven years, the IEP Program has managed full-scale field tests of Hg control technologies at nearly 50 US coal-fired power generation facilities. The flexibility of the IEP Program allowed NETL to quickly incorporate insights and lessons learned from its 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 Hg oxidation in flue gas led to field testing of technologies designed to provide a halogen "boost" for coals, such as subbituminous and lignite, that tend to contain low levels of chlorine. NETL has observed a step-change improvement in both the cost and performance of Hg control during full-scale field tests of coal treatment with an aqueous calcium bromide ($CaBr_2$) solution at plants equipped with a wet FGD system, and chemically-treated (or brominated) ACI upstream of a particulate control device.

3.1. *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 some coal-fired

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power plants. To optimize Hg capture across FGD systems, NETL has funded field tests of technologies, such as chemical additives and Hg^0 oxidation catalysts, which promote Hg^0 oxidation in coal combustion flue gas. The impact of combustion modifications, such as coal reburn, on flue gas Hg^0 oxidation has also been examined under the IEP Program [5]. In addition, NETL has evaluated FGD additives designed to suppress Hg^0 re-emissions across the scrubber.

3.1.1. Chemical additives

The ability of chemical additives, sprayed onto the coal as an aqueous salt solution, to promote flue gas Hg^0 oxidation and enhance FGD Hg capture was evaluated during a full-scale field test completed at Luminant Power's Monticello Station Unit 3 [6]. During a two-week trial conducted at Monticello Station, which burns a 50:50 blend of Powder River Basin (PRB) subbituminous and Texas lignite coals, total Hg capture across the ESP/FGD configuration averaged 86% with a $CaBr_2$ injection rate equivalent to 113 parts per million (ppm) Br in the dry coal. Greater than 90% total Hg capture was observed during a short-term test with a $CaBr_2$ injection rate equivalent to 330 ppm Br in the coal.

NETL has also conducted pilot- and full-scale field tests of wet FGD additives designed to limit Hg^0 re-emissions through the formation of insoluble salts with Hg^{2+}[7]. 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.

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 and 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 and 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 [8]. During long-term testing, total Hg removal averaged about 92% with the addition of Absorption Plus (Hg)^{TM}. Note that over 80% of total Hg removal was observed under baseline conditions.

3.1.2. Catalysts

The ability of fixed-bed catalysts to promote flue gas Hg^0 oxidation has been evaluated at pilot-scale, and a full-scale field test of a gold-based catalyst is scheduled to begin in 2008 at Lower Colorado River Authority's Fayette Unit 3 [9]. The catalysts are designed for installation 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.

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During pilot-scale testing at Great River Energy's North Dakota lignite-fired Coal Creek Station, about 67% Hg^0 oxidation was measured across a palladium-based (Pd#1) catalyst, after 20 months of operation. Following thermal regeneration, Hg^0 oxidation across the Pd#1 catalyst increased from 67 to 88% (near the 95% activity of the fresh catalyst). Meanwhile, nearly 80% total Hg capture was observed across the pilot-scale wet FGD, with 84% Hg^{2+} at the FGD inlet.

At Luminant Power's Monticello Station, severe fly ash build-up was observed on the catalyst surfaces, likely caused by frequent pilot unit outages during the test campaign. Following catalyst cleaning, Hg^0 oxidation was approximately 72% across the regenerated Pd#1 catalyst (transferred from Coal Creek) and 66% across a gold-based catalyst, after about 20 months of pilot-scale operation. Total Hg capture across a pilot-scale wet FGD ranged from 76 to 87%, compared with only 36% removal under baseline conditions. This equates to about 70% incremental Hg capture due to the catalysts.

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3.2. *Sorbent injection*

The cost-effectiveness of ACI has improved dramatically following technological advancements fuelled by the performance and cost goals set forth by the IEP Program. However, these improvements did not come easily. In 2001, concerns about the effectiveness of ACI in controlling Hg emissions were uncovered during field testing at We Energies' PRB-fired Pleasant Prairie Unit 2 where untreated ACI removed only about 65% of the total Hg [10]. It was determined that subbituminous and lignite coal combustion flue gases tend to contain high levels of Hg^0 – a form that is not readily captured by untreated ACI. This excess is caused by a lack of Cl, another coal constituent, which promotes Hg^0 oxidation.

In response to these findings, NETL initiated field tests of chemically-treated (or brominated) ACI that showed much promise at laboratory and bench scale in capturing both Hg^0 and Hg^{2+}. Chemically-treated sorbents tested by NETL were developed by several companies, including Alstom Power (Mer-Clean™), Norit Americas (DARCO® Hg-LH), and Sorbent Technologies Corporation (B-PAC™). Figure 1 provides a comparison of untreated and chemically-treated ACI performance at three of NETL's Phase II field testing sites: (1) Great River Energy's Stanton Station Unit 10 (Lignite/FF); (2) Basin Electric's Leland Olds Station Unit 1 (Lignite/ESP); and (3) Stanton Station Unit 1 (PRB/ESP). These parametric data curves illustrate the improved Hg capture efficiency of chemically-treated sorbents at power plants burning lower-rank coals as high levels of Hg capture are attainable at relatively low injection rates. In fact, the treated sorbents achieved at least 90% total Hg capture at an injection rate of 3 pounds per million actual cubic feet (lb/MMacf) of flue gas or less at these Phase II field testing sites.

An NETL economic analysis [11] released in May 2007 indicates that the high Hg capture efficiency of chemically-treated sorbents has drastically reduced the estimated cost of Hg control due to a reduction in the injection rate required to achieve a given level of control, which offsets the higher cost of these treated sorbents. As shown in Figure 2, the 20-year levelised incremental cost of 90% ACI Hg control ranges from about $30,000 to less than $10,000/lb Hg removed for seven of NETL's Phase II field testing sites where chemically-treated ACI was evaluated. These results point to the fact that NETL has surpassed the Hg control cost goal set forth by the IEP Program.

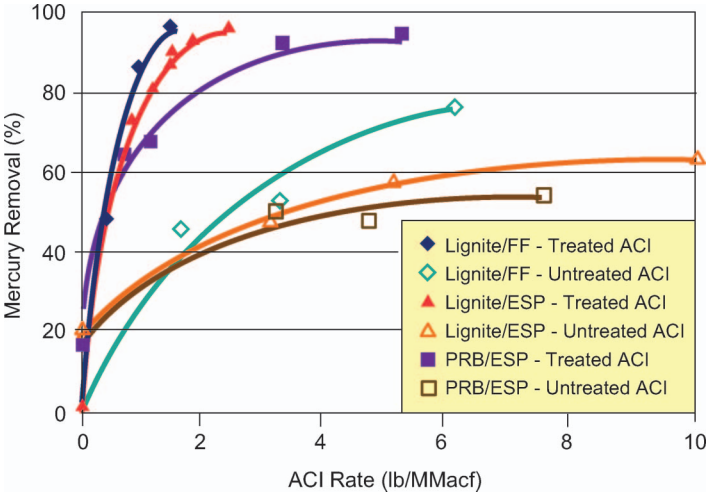

Figure 1. Comparison of untreated and chemically-treated ACI performance at facilities burning lower-rank coals.

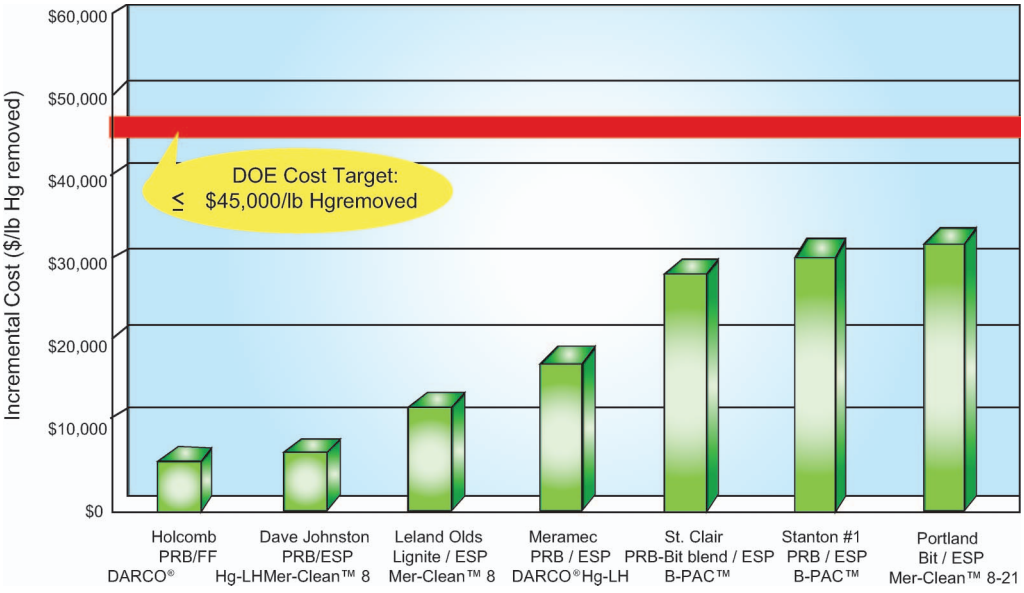

Figure 2. 20-year levelized incremental cost of 90% Hg control with chemically-treated ACI.

3.2.1. *Technical issues*

Although the advent of chemically-treated ACI has yielded improvements in Hg control cost and performance, technical uncertainties still remain. The following issues, if resolved, will further enhance the efficiency, economics, applicability and reliability of sorbent-based Hg control technologies.

3.2.1.1. Fly ash impacts. The typical ACI system is located upstream of a particulate control device to enable simultaneous capture of the spent sorbent and fly ash. This Hg

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control strategy leads to commingling of the sorbent and fly ash that can prohibit certain fly ash recycling efforts. One of the highest-value reuse applications for fly ash is as a substitute for Portland cement in concrete production [12]. 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. Accordingly, NETL's Hg control technology portfolio includes alternative sorbent injection technologies designed to minimize fly ash carbon contamination caused by ACI upstream of a particulate control device.

The toxic emissions control (TOXECON™) configuration, developed by EPRI, will not impact fly ash utilization because the ash is removed by an ESP upstream of the sorbent injection location, whereas the spent sorbent is captured by a downstream FF. TOXECON™ was selected for a first-of-a-kind commercial Hg control technology demonstration at We Energies' Presque Isle Power Plant in Marquette, Michigan, under DOE's Clean Coal Power Initiative. Operational since 2006, the TOXECON™ configuration maintained greater than 90% total Hg removal for 48 consecutive days. Sorbent injection rates of about 1.7 and 1.2 lb/MMacf are required to achieve at least 90% total Hg removal with untreated DARCO® Hg and brominated DARCO® Hg-LH, respectively [13].

EPRI's TOXECON II™ technology injects sorbents directly into the downstream collecting field(s) of an ESP. Because 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. During full-scale TOXECON II™ testing at Entergy's PRB-fired Independence Station Unit 1, DARCO® Hg-LH injection at 5.5 lb/MMacf achieved 90% total Hg removal [14]. A remaining concern with any Hg control strategy involving sorbent injection, particularly the TOXECON II™ configuration that limits ESP residence time, is the potential for increased particulate emissions that could trigger New Source Review requirements.

Activated carbon sorbents passivated during production could potentially allow coal-fired power generators to continue marketing fly ash commingled with the spent sorbent as a suitable replacement for Portland cement in concrete. Sorbent Technologies conducted a 30-day long-term evaluation of their brominated, "concrete-friendly" C-PAC™ sorbent at Midwest Generation's PRB-fired Crawford Station Unit 7 [15]. Total Hg removal averaged 81% with C-PAC™ injection upstream of the ESP at about 4.6 lb/MMacf.

More recently, a high-temperature version of C-PAC™ was tested at Midwest Generation's PRB-fired Will County Unit 3, which is equipped with a hot-side ESP [16]. During a six-day continuous test, Hg removal ranged from about 60 to 73% with C-PAC™ injection at 5 lb/MMacf. Most importantly, preliminary results indicate that fly ash collected during C-PAC™ injection at these sites remains suitable for reuse in concrete production.

During Phase III testing at Lower Colorado River Authority's PRB-fired Fayette Unit 3, Alstom evaluated three sorbents (eSorb™ 11, eSorb™ 13 and eSorb™ 18) designed by Envergex to preserve fly ash quality [17]. Results indicate that fly ash remains marketable with eSorb™ 13 at about 0.5 lb/MMacf (~85% ACI Hg capture).

3.2.1.2. Sulfur trioxide interference. Field testing has shown that sulfur trioxide (SO_3) in the flue gas, even at low concentrations, can impede the performance of ACI. It appears that SO_3 competes with Hg for adsorption sites on the sorbent surface, thereby limiting its performance [18].

During Phase II field testing at AEP's high-sulfur (3–4%) bituminous-fired Conesville Station Unit 6, total Hg removal was limited to approximately 30% with

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chemically-treated ACI at 12 lb/MMacf [19]. Consequently, a long-term field test was not conducted at this unit; instead, NETL funding was used to evaluate the impact of SO_3 flue gas conditioning (FGC) on ACI performance at AmerenUE's PRB-fired Labadie Station Unit 2 [20]. As shown in Figure 3, turning the SO_3 FGC system off at Labadie increased total Hg removal from about 50 to 80% with DARCO® Hg-LH injection at 8 lb/MMacf. Greater than 90% Hg removal was observed with no SO_3 injection and DARCO® Hg-LH injection upstream of the air preheater (APH) at about 5 lb/MMacf. The performance of brominated B-PAC™ was also impacted by SO_3 FGC at Progress Energy's Lee Station Unit 1 [21]. With B-PAC™ injection at 8 lb/MMacf, Hg capture increased from 32 to 82% when SO_3 FGC was idled.

One possible solution to the SO_3 issue is dual injection of Hg sorbents and alkaline materials. This approach was explored during 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 ($\sim$1% sulfur) and is equipped with a SCR system followed by two ESPs in series [22]. During parametric testing, several Hg sorbents were evaluated both with and without the injection of magnesium oxide (MgO) or sodium sesquicarbonate (trona) – two potential SO_3 mitigation additives. 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 SO_3 mitigation, Hg removal was limited to about 22% with brominated DARCO® Hg-LH injection between the two ESPs at 8 lb/MMacf. 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.

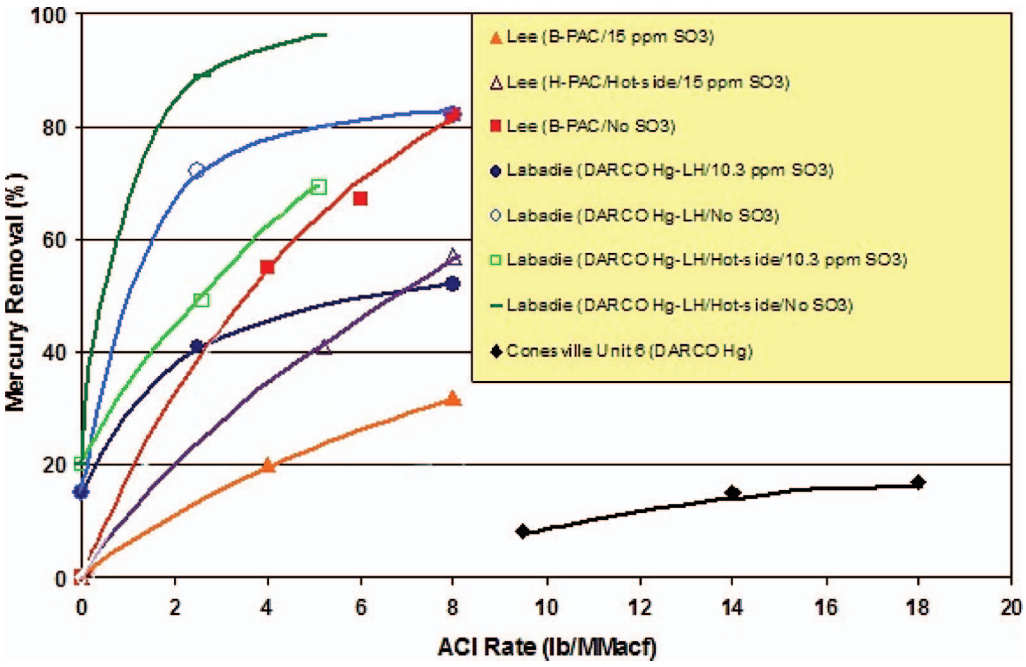

Figure 3. Impact of flue gas SO_3 on ACI performance.

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3.3. 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 method to produce sorbent *in situ* by extracting partially combusted coal from the furnace, which is subsequently injected into the flue gas as an alternative to conventional ACI. The cost for producing Thief carbon sorbents ranges 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 palladium (Pd) based sorbent that works on fuel gas at elevated temperatures. Unlike conventional sorbents such as activated carbon, which operate at lower 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.

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4. Summary

Insight into the factors that can influence Hg speciation and capture in coal combustion flue gas has allowed NETL to prioritize the search for reliable and cost-effective Hg control strategies. A determination that chlorine released during coal combustion promotes Hg^0 oxidation in flue gas led to field testing of technologies designed to provide a halogen "boost" for coals, such as subbituminous and lignite, that tend to contain low levels of chlorine. NETL has observed a step-change improvement in both the cost and performance of Hg control during full-scale field tests with chemically-treated ACI and $CaBr_2$ coal treatment. The improved Hg capture efficiency of these advanced control technologies has allowed NETL to satisfy the cost and performance goals set forth by the IEP Programme.

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 [23], NETL's field testing programme 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 Programme, have been ordered by US coal-fired power generators [24]. These contracts represent over 44 gigawatts (GW) of coal-fired electric generating capacity. This includes approximately 33 GW of existing capacity ($\sim$10% of total US 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 many applications based on results from NETL's field testing programme, at a cost estimated to dip below $10,000/lb Hg removed. However, although the results achieved during NETL's field tests met or exceeded the programme goals, only through experience gained during long-term continuous operation of these advanced technologies in a range of full-scale commercial applications, their actual costs and performance will be determined.

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Notes

1. In Fiscal Year 2008, the IEP Program's focus was redirected to the research, development, and deployment of advanced carbon dioxide capture and compression technologies for the existing fleet of coal-fired power plants.

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