Control of Trace Metal Emissions During Coal Combustion

Topical Report
July 1 - September 30, 1996

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TECHNICAL PROGRESS REPORT
July 1, 1996 through September 30, 1996

Project Title: **CONTROL OF TRACE METAL EMISSIONS DURING COAL COMBUSTION**

DOE Grant Number: DE-FG22-94PC94221
Principal Investigator: Thomas C. Ho, Lamar University
DOE Project Officer: Mike Baird, PETC

ABSTRACT

Emissions of toxic trace metals in the form of metal fumes or submicron particulates from a coal-fired combustion source have received greater environmental and regulatory concern over the past years. Current practice of controlling these emissions is to collect them at the cold-end of the process by air-pollution control devices (APCDs) such as electrostatic precipitators and baghouses. However, trace metal fumes may not always be effectively collected by these devices because the formed fumes are extremely small.

The proposed research is to explore the opportunities for improved control of toxic trace metal emissions, alternatively, at the hot-end of the coal combustion process, i.e., in the combustion chamber. The technology proposed is to prevent the metal fumes from forming during the process, which would effectively eliminate the metal emission problems. Specifically, the technology is to employ suitable sorbents to (1) reduce the amount of metal volatilization during combustion and (2) capture volatilized metal vapors. The objectives of the project are to demonstrate the technology and to characterize the metal capture process during coal combustion in a fluidized bed combustor.

The project was started on July 1, 1994 and this is the ninth quarterly technical progress report. Specifically, the following progress has been made during this performance period from July 1, 1996 through September 30, 1996:

1. **Metal Capture Experiments Continued** - Additional combustion experiments involving seven different coal samples were carried out in a quartz fluidized bed combustor.

2. **Arsenic and Selenium Results Observed** - Arsenic and selenium capture efficiencies were determined using a Buck Scientific Model 210VGP Atomic Absorption Spectrophotometer equipped with a continuous flow hydride generator.

3. **Paper Presented** - A paper, entitled "Trace Metal Capture by Various Sorbents during Fluidized Bed Coal Combustion," was presented at the 212th American Chemical Society National Meeting held in Orlando, Florida, August 25-29, 1996.

4. **Presentation Invited** - An invited paper related to metal capture during coal combustion was prepared and accepted for presentation at the Fifth Asian Conference on Fluidization and Three-Phase Reactors to be held in Hsitou, Taiwan, December 16-20, 1996.

EXECUTIVE SUMMARY

Toxic (or potentially toxic) trace metallic elements such as barium, beryllium, boron, cadmium, chromium, lead, mercury, nickel, selenium, strontium, vanadium, zinc and zirconium are usually contained in coal in various forms. These metals will either stay in the ash or be vaporized during high temperature combustion. Portions of the vaporized metals may eventually be emitted from a combustion system. Most of the emitted metals will be in the form of metal fumes or particulates with diameters less than 1 micron and are potentially hazardous to the environment. The U.S. EPA has reported that metals account for almost all of the identified risks from waste incineration systems.

Concern over toxic trace metal emissions from coal-fired combustion sources is growing, especially as the result of the passage of the 1990 Clean Air Act Amendments (CAAA). To address the concern, the U.S. DOE has recently co-sponsored a workshop jointly with the Electric Power Research Institute (EPRI) and the Energy and Environmental Research Center (EERC) on Trace Elements Transformations in Coal-Fired Power Plants. The objective of the workshop was to evaluate the current level of understanding on metal behavior during coal combustion and to identify potential technologies for improved metal emission control.

Current practice of controlling trace metal emissions during coal combustion employs conventional air pollution control devices (APCDs), e.g., venturi scrubbers, electrostatic precipitators, baghouses etc., to collect fly ash and metal fumes. This type of control is essentially a cold-end control because metals are allowed to vaporize and condense before being controlled. The control may not always be effective on metal fumes due to their extremely fine sizes.

An alternative technology for metal emission control is to minimize the formation of metal fumes at the hot-end of the coal combustion process, i.e., in the combustion chamber. The technology proposed is to prevent the metal fumes from forming during the process, which would effectively eliminate the metal emission problems. Specifically, the technology is to employ suitable sorbents to (1) reduce the amount of metal volatilization during combustion and (2) capture volatilized metal vapors. The objectives of the project are to demonstrate the technology and to characterize the metal capture process during coal combustion in a fluidized bed combustor.

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4. **Presentation Invited** - An invited paper related to metal capture during coal combustion was prepared and accepted for presentation at the Fifth Asian Conference on Fluidization and Three-Phase Reactors to be held in Hsitou, Taiwan, December 16-20, 1996.

DETAILED PROGRESS REPORT

1. Metal Capture Experiments Continued

Additional combustion experiments involving different coal samples were conducted in the constructed quartz fluidized bed combustor during this performance period. In an experiment, a coal sample was burned in the bed with a sorbent under a specific set of combustion conditions and the amount of metal capture by the sorbent was determined. Seven different coal samples from the Illinois Basin Coal Sample Bank were tested in the study. The metals involved were lead, cadmium, chromium, arsenic and selenium, and the sorbents tested included bauxite, zeolite and lime. Typical current experimental results are summarized in Tables 1 through 5 in ATTACHMENT 1, 2, 3, 4 and 5, respectively.

The observed experimental results shown in Tables 1 through 3 for lead, cadmium and chromium indicate that metal capture by sorbent can be as high as 91% depending on the metal species and sorbent involved. All three sorbents are observed to be capable of capturing lead and cadmium in various degree, and zeolite and lime are able to capture chromium. Bauxite, however, is not capable of capturing chromium. Note that the description of the corresponding experimental conditions is given in ATTACHMENT 6.

2. Arsenic and Selenium Results Observed

Arsenic and selenium capture efficiencies were determined using a Buck Scientific Model 210VGP Atomic Absorption Spectrophotometer equipped with a continuous flow hydride generator. The equipped hydride generator provides much needed detectability in the measuring of trace metal concentrations, especially for arsenic and selenium. A description of the Buck Scientific Continuous Flow Hydride/Cold Vapor System for parts-per-trillion level detectability was included in the fifth TECHNICAL PROGRESS REPORT.

The results shown in Tables 4 and 5 for arsenic and selenium indicate that capture of these two metals by sorbents during fluidized bed coal combustion is possible, however, the capture efficiency is, in general, lower than that for lead and cadmium. As indicated in these two tables, the capture efficiency for arsenic and selenium is seen to be around 20%. Additional experiments and measurements are currently being carried out to obtain more statistically representative results.

3. Paper Presented at ACS Meeting

A paper, entitled "Trace Metal Capture by Various Sorbents during Fluidized Bed Coal Combustion," was presented at the 212th American Chemical Society National Meeting held in Orlando, Florida, August 25-29, 1996. A copy of the paper was included in ATTACHMENT 4 in the last technical progress report.

4. Presentation Invited by Asian Conference

A paper related to metal capture during coal combustion was invited for presentation at the Fifth Asian Conference on Fluidization and Three-Phase Reactors to be held in Hsitou, Taiwan, December 16-20, 1996. The paper was entitled "Trace Metal Capture by Various Sorbents during Fluidized Bed Coal Combustion." A copy of the paper is included in ATTACHMENT 6.

FUTURE WORK PLANNED

The work planned for the next quarter will be to continue metal capture experiments in the quartz fluidized bed combustor to provide statistically representative results. All five targeted metals, i.e., lead, cadmium, chromium, arsenic and selenium, will be involved in the experiments. The newly tested Buck Scientific Model 210VGP Atomic Absorption Spectrophotometer will mainly be used to measure arsenic and selenium concentrations. The experiments will continue to use the seven coal samples received from the Illinois Basin Coal Sample Bank, i.e., IBC-101, IBC-102, IBC-106, IBC-109, IBC-110, IBC-111, and IBC-112.

ATTACHMENT

1. Table 1. Percentage Lead Capture by Sorbents (%)

2. Table 2. Percentage Cadmium Capture by Sorbents (%)

3. Table 3. Percentage Chromium Capture by Sorbents (%)

4. Table 4. Percentage Arsenic Capture by Sorbents (%)

5. Table 5. Percentage Selenium Capture by Sorbents (%)

6. "Trace Metal Capture by Various Sorbents during Fluidized Bed Coal Combustion," paper invited for presentation at the Fifth Asian Conference on Fluidization and Three-Phase Reactors to be held in Hsitou, Taiwan, December 16-20, 1996.

ATTACHMENT 1

Table 1. Percentage Lead Capture by Sorbents (%)

Coal\Sorbent	Bauxite	Zeolite	Lime
IBC-101	45	63	86
IBC-102	86	83	17
IBC-106	73	52	67
IBC-109	59	64	68
IBC-110	68	60	28
IBC-111	58	91	32
IBC-112	66	51	21
Average	**65**	**66**	**46**

ATTACHMENT 2

Table 2. Percentage Cadmium Capture by Sorbents (%)

Coal\Sorbent	Bauxite	Zeolite	Lime
IBC-101	46	47	32
IBC-102	38	53	26
IBC-106	76	86	81
IBC-109	54	86	46
IBC-110	66	64	42
IBC-111	74	22	57
IBC-112	72	85	21
Average	**61**	**63**	**44**

ATTACHMENT 3

Table 3. Percentage Chromium Capture by Sorbents (%)

Coal\Sorbent	Bauxite	Zeolite	Lime
IBC-101	0	7	4
IBC-102	0	51	37
IBC-106	0	26	13
IBC-109	0	25	22
IBC-110	0	57	7
IBC-111	0	19	4
IBC-112	0	17	13
Average	**0**	**29**	**14**

ATTACHMENT 4

Table 4. Percentage Arsenic Capture by Sorbents (%)

Coal\Sorbent	Bauxite	Zeolite	Lime
IBC-101	57	24	10
IBC-102	19	7	30
IBC-106	28	14	15
IBC-109	34	13	7
IBC-110	32	19	21
IBC-111	28	18	17
IBC-112	43	22	14
Average	**34**	**17**	**16**

ATTACHMENT 5

Table 5. Percentage Selenium Capture by Sorbents (%)

Coal\Sorbent	Bauxite	Zeolite	Lime
IBC-101	29	28	24
IBC-102	18	9	16
IBC-106	9	8	10
IBC-109	43	11	12
IBC-110	12	3	12
IBC-111	18	24	22
IBC-112	8	10	12
Average	**20**	**13**	**15**

ATTACHMENT 6

TRACE METAL CAPTURE BY VARIOUS SORBENTS DURING FLUIDIZED BED COAL COMBUSTION*

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ABSTRACT

This study investigated the potential of employing suitable sorbents to capture toxic trace metallic substances during fluidized bed coal combustion. Metal capture experiments were carried out in a 25.4 mm (1") quartz fluidized bed combustor enclosed in an electric furnace. The metals involved were cadmium, lead and chromium, and the sorbents tested included bauxite, zeolite and lime. In addition to the experimental investigations, potential metal-sorbent reactions were also identified through chemical equilibrium calculations based on the minimization of system free energy. The observed experimental results indicated that metal capture by sorbents can be as high as 91% depending on the metal species and sorbent involved. Results from thermodynamic equilibrium simulations suggested the formation of metal-sorbent compounds such as Pb_2SiO_4(s), $CdAl_2O_4$(s) and $CdSiO_3$(s) under the combustion conditions.

INTRODUCTION

Toxic trace metallic elements such as arsenic, cadmium, chromium, lead, mercury, and selenium are usually contained in coal in various forms and trace amounts. Portions of these metals may eventually be emitted from a combustion system in the form of metal fumes or particulates with diameters less than 1 micron, which are potentially hazardous to the environment (Davidson et al., 1974). Current practice of controlling trace metal emissions during coal combustion employs conventional air pollution control devices (APCDs), such as electrostatic precipitators and baghouses, to collect fly ash and metal fumes. The control may not always be effective on metal fumes due to their extremely fine sizes (Oppelt, 1987).

Concern over toxic trace metal emissions from coal-fired combustion sources is growing, especially as the result of the passage of the 1990 Clean Air Act Amendments (CAAA) where eleven metallic elements, i.e., antimony, arsenic, beryllium, cadmium, chromium, cobalt, lead, manganese, mercury, nickel, and selenium are listed as potential hazardous air pollutants. This study is to explore the opportunities for improved control of toxic trace metal emissions from coal-fired combustion systems. Specifically, the technology proposed is to employ suitable sorbents to (1) reduce the amount of metal volatilization and (2) capture volatilized metal vapors during fluidized bed coal combustion. The objectives of the study were to demonstrate the capture process, identify effective sorbents, and characterize the capture efficiency.

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* For presentation at the Fifth Asian Conference on Fluidization and Three-Phase Reactors to be held in Hsitou, Taiwan, December 16-20, 1996.

SCIENTIFIC DISCUSSION

Chemical absorption reactions between metal vapors and a variety of sorbents at high temperatures have been observed both in a packed bed and in a fluidized bed (see, e.g., Uberol and Shadman, 1990; Ho et al., 1992, 1994). The following reactions between metals and sorbent constituents have been confirmed both theoretically and experimentally:

$$2\ PbO + SiO_2 \text{ ----> } Pb_2SiO_4(s) \qquad (A)$$
$$CdO + SiO_2 \text{ ----> } CdSiO_3(s) \qquad (B)$$
$$CdO + Al_2O_3 \text{ ----> } CdAl_2O_4(s) \qquad (C)$$
$$PbCl_2 + Al_2O_3{:}2SiO_2 + H_2O \text{ ----> } PbO{:}Al_2O_3{:}2SiO_2(s) + 2\ HCl(g) \qquad (D)$$
$$CdCl_2 + Al_2O_3 + H_2O \text{ ----> } CdAl_2O_4(s) + 2\ HCl(g) \qquad (E)$$

The technology of metal capture by sorbents, however, has never been evaluated during fluidized bed coal combustion.

EQUILIBRIUM CALCULATION

Equilibrium composition represents the most stable chemical composition within a system under a specific state. Thermodynamically, this composition is corresponding to the one where the system free energy is minimized. The calculated equilibrium composition would reveal the preferred chemical speciation under a specific state, which in turn, suggest potential chemical reactions which may occur within the system. In this study, combustion equilibrium was calculated using a PC-based computer software package (Ho, 1996) especially developed for predicting equilibrium compositions during fuel or waste combustion.

EXPERIMENTAL

Metal capture experiments were carried out semi-batchwise in a 25.4 mm (1") OD quartz fluidized bed coal combustor enclosed in an electric furnace. Seven coal samples from the Illinois Basin Coal Sample Bank (IBCSB) were tested in the experiments. The concentration of sulfur, chlorine, and the target metals in these sample was summarized in Table 1. The sorbents tested included bauxite, zeolite and lime. Their chemical composition and the corresponding minimum fluidization velocity (U_{mf}) at 900°C are listed in Table 2.

In an experimental run, a bed of sorbent was preheated to the desired temperature under the designed operating conditions. A predetermined amount of coal was then charged in the bed at a constant feed rate for combustion. After the combustion was completed, the bed residue including sorbent and ash was discharged and separated for analysis of metal concentration. The experimental parameters and operating conditions are summarized in Table 3.

Metal concentration in coal, wood pellets, original sorbent, and combustor residue was determined by an atomic absorption spectrophotometer. An HF modified EPA Method 3050 was used to digest metals from the sorbent, which involves the use of HNO_3, HCl and HF acids. Toxicity Characteristics Leaching Procedure (TCLP) tests were performed to determine the leachability of the captured metals from sorbents.

RESULTS AND DISCUSSION

Simulation Results

Two typical sets of simulation results indicating potential metal-sorbent reactions and the effect of sulfur on metal capture by sorbents are shown in Tables 4 and 5, for lead and cadmium, respectively. The corresponding elemental composition and combustion conditions used in the simulations were: carbon - 71.3 wt%, hydrogen - 5.2 wt%, nitrogen - 1.4 wt%, oxygen - 12.4 to 7.8 wt%, sulfur - 0 to 4.6 wt%, metal concentration - 50 ppm, ash - 9.3 wt%, combustion temperature - 900°C, and percent excess air - 50%.

The results shown in Table 4 indicate that lead will react with both sulfur and silica during combustion. At a temperature below 950°C, $PbSO_4$(s) is the thermodynamically preferred lead compound; however, between 950°C and 1000°C, $PbSiO_4$(s) is the preferred one; and, above 1000°C, PbO(g) is the dominating species. These results suggest that silica is thermodynamically capable of capturing lead. However, the existence of sulfur will affect the capture process especially at a temperature below 950°C. Note that lead does not seem to react with Al_2O_3 according to the equilibrium simulation. It should also be pointed out that lead does not show any reactions with CaO because there are no thermodynamic data available in the literature.

The results shown in Table 5 for cadmium indicate that cadmium will react with Al_2O_3 and SiO_2 to form $CdAl_2O_4$(s) and $CdSiO_3$(s), respectively. It, however, will not react with CaO. The existence of sulfur does not seem to interfere with the reactions according to the equilibrium results shown in the table. These simulation results, again, suggest that silica and alumina have potential to capture cadmium under the combustion conditions. Note that, although not shown, the simulation results for chromium have indicated that the thermodynamically preferred chromium compound under the combustion conditions is exclusively Cr_2O_3 (s) and no chromium-sorbent compounds are observed. The results suggest that, thermodynamically, the tested sorbents are not expected to chemically absorb chromium during combustion.

Experimental Results

Typical experimental results indicating the effectiveness of metal capture by various sorbents are shown in Tables 6, 7 and 8 for lead, cadmium, and chromium, respectively. The results shown in Table 6 for lead indicate that all three sorbents tested are capable of capturing lead during fluidized bed combustion with the average capture efficiency ranging from 44% to 66%. Zeolite and bauxite are seen to be more effective than lime. As suggested by equilibrium simulations, the mechanism of lead capture by zeolite appears to be due to the formation of Pb_2SiO_4(s) and the mechanism of lead capture by bauxite may be due to the formation of the same compound or an alumino-silica compound. The mechanism of lead capture by lime, however, may be due to the "melt capture" as suggested by Linak and Wendt (1993).

For cadmium capture, the results shown in Table 7 indicate that the average capture efficiency associated with a sorbent is very similar to that of lead capture by the sorbent. Zeolite and bauxite again are seen to be more effective than lime, suggesting the formation of $CdAl_2O_4$(s) and $CdSiO_3$(s) based on equilibrium simulations. The formation of these compounds, however, could not be analytically confirmed due to their low concentrations in the sorbents.

The chromium capture results shown in Table 8 indicate that zeolite and lime are both capable of capturing the metal. The average capture efficiencies are seen to be from 14% to 29% which are much lower than those of lead and cadmium capture. The mechanisms of chromium capture by these sorbents, however, are not clear at this time. Efforts are currently devoted to analytically identify the chromium state in the sorbents. Note that bauxite was not observed to capture any chromium because the original bauxite contained a high concentration of chromium which continued to vaporize during combustion. The net result was that, in contrast to chromium capture, bauxite gave away chromium during the process.

Although not shown, it was observed that the amount of lead and cadmium capture by a unit mass of sorbents was found to be roughly proportional to the concentration of the metal in coal. This, however, was not observed for chromium capture by sorbents. Note that, the observed results have not clearly indicated the effects of sulfur and other coal properties on capture efficiency of the metal capture process.

CONCLUSIONS

This study investigated the potential of employing suitable sorbents to capture toxic trace metals during fluidized bed coal combustion. The observed experimental results indicated that metal capture by sorbents can be as high as 91% depending on the metal species and sorbent involved. All three sorbents tested, i.e., bauxite, zeolite and lime, were observed to be capable of capturing lead and cadmium in a various degree, and zeolite and lime were able to capture chromium. Results from thermodynamic equilibrium simulations suggested the formation of metal-sorbent compounds such as Pb_2SiO_4(s), $CdAl_2O_4$(s) and $CdSiO_3$(s) under the combustion conditions.

ACKNOWLEDGEMENTS

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Table 1. Sulfur, Chlorine and Target Metals in Coal Samples
(Units: % for S and Cl, ppm for metals)

Coal\Element	S	Cl	Cd	Cr	Pb
IBC-101	4.4	0.1	1.1	31	8
IBC-102	3.3	0.0	0.8	7	149
IBC-106	3.8	0.0	0.2	10.4	6
IBC-109	1.2	0.4	<0.3	13	18
IBC-110	4.6	0.0	<0.4	11	10
IBC-111	2.0	0.0	<0.4	14	18
IBC-112	2.8	0.2	<0.3	14	27

Table 2. Major Composition, Trace Metal Concentration and Fluidization Properties of the Three Tested Sorbents

Composition or Property	Bauxite	Zeolite	Lime
SiO_2 (%)	9.0	66.7	0.7
Al_2O_3 (%)	78.0	12.1	0.3
CaO (%)	0.0	3.1	97.2
Cd (ppm)	2.0	3.0	3.6
Cr (ppm)	146	4.0	7.8
Pb (ppm)	43.2	60.4	72.4
d_p (mm)	0.5	0.5	0.5
U_{mf} (cm/s)	3.8	3.5	3.8

Table 3. Experimental Parameters and Operating Conditions

Parameter	Range
Coal Size	2.0 - 2.8 mm
Coal Amount	60 g
Coal Feed Rate	0.22 g/min
Sorbent Size	0.4 - 0.6 mm
Sorbent Amount	22.5 - 30 g
Static Sorbent Height	6 cm
Air Flow Rate	3 U_{mf} of Sorbent
Combustor Temperature	900°C
Combustion Duration	4.5 hrs

Table 4. Equilibrium Simulation Results for Lead with or without Sulfur

Sorbent Constituent	Metal	With or Without Sulfur	Sulfur-Metal-Sorbent Compound	
SiO_2	Pb	Without S	**$Pb_2SiO_4(s)$**	< 1000°C
			$PbO(g)$	> 1000°C
		With S	**$PbSO_4(s)$**	< 950°C
			$Pb_2SiO_4(s)$	< 1000°C
			$PbO(g)$	> 1000°C
Al_2O_3	Pb	Without S	$PbO(s)$	< 900°C
			$PbO(g)$	> 900°C
		With S	**$PbSO_4(s)$**	< 950°C
			$PbO(g)$	> 950°C
CaO	Pb	Without S	$PbO(s)$	< 900°C
			$PbO(g)$	> 900°C
		With S	**$CaSO_4(s)$**	> 500°C
			$PbSO_4(s)$	< 950°C
			$PbO(g)$	> 950°C

Table 5. Equilibrium Simulation Results for Cadmium with or without Sulfur

Sorbent Constituent	Metal	With or Without Sulfur	Sulfur-Metal-Sorbent Compound	
SiO_2	Cd	Without S	**$CdSiO_3(s)$**	< 850°C
			CdO(s)	<1000°C
			Cd(g)	>1000°C
		With S	**$CdSO_4(s)$**	< 800°C
			CdO(s)	< 900°C
			CdS(g)	> 900°C
Al_2O_3	Cd	Without S	**$CdAl_2O_4(s)$**	< 950°C
			CdO(s)	<1000°C
			Cd(g)	>1000°C
		With S	**$CdAl_2O_4(s)$**	< 950°C
			CdS(g)	> 950°C
CaO	Cd	Without S	CdO(s)	<1000°C
			Cd(g)	>1000°C
		With S	**$CaSO_4(s)$**	> 500°C
			CdO(s)	< 900°C
			CdS(g)	> 900°C

Table 6. Percentage Lead Capture by Sorbents (%)

Coal\Sorbent	Bauxite	Zeolite	Lime
IBC-101	45	63	86
IBC-102	86	83	17
IBC-106	73	52	67
IBC-109	59	64	68
IBC-110	68	60	28
IBC-111	58	91	32
IBC-112	66	51	21
Average	**65**	**66**	**46**

Table 7. Percentage Cadmium Capture by Sorbents (%)

Coal\Sorbent	Bauxite	Zeolite	Lime
IBC-101	46	47	32
IBC-102	38	53	26
IBC-106	76	86	81
IBC-109	54	86	46
IBC-110	66	64	42
IBC-111	74	22	57
IBC-112	72	85	21
Average	**61**	**63**	**44**

Table 8. Percentage Chromium Capture by Sorbents (%)

Coal\Sorbent	Bauxite	Zeolite	Lime
IBC-101	0	7	4
IBC-102	0	51	37
IBC-106	0	26	13
IBC-109	0	25	22
IBC-110	0	57	7
IBC-111	0	19	4
IBC-112	0	17	13
Average	**0**	**29**	**14**