Control of Trace Metal Emissions During Coal Combustion

Quarterly Report
January 1 - March 31, 1997

By
Thomas C. Ho

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P.O. Box 880
Morgantown, West Virginia 26507-0880

By
Lamar University
Beaumont, Texas 77710

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TECHNICAL PROGRESS REPORT

(January 1, 1997 through March 31, 1997)

Prepared

for the Project

CONTROL OF TRACE METAL EMISSIONS DURING COAL COMBUSTION

Thomas C. Ho
Department of Chemical Engineering
Lamar University
Beaumont, Texas

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Prepared by
LAMAR UNIVERSITY
Beaumont, Texas 77710
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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 eleventh quarterly technical progress report. Specifically, the following progress has been made during this performance period from January 1, 1997 through March 31, 1997:

1. **Metal Capture Experiments Continued** - Additional combustion experiments involving seven different coal samples were carried out to obtain more statistically representative results.

2. **Additional Results Obtained** - Additional metal capture results were obtained and were added into the existing database. The metals involved included lead, cadmium, chromium, arsenic, and selenium.

3. **Paper Presented** - A paper, entitled "Trace Metal Capture by Various Sorbents During Fluidized Bed Coal Combustion," was presented at the 22nd International Technical Conference on Coal Utilization & Fuel Systems held in Clearwater, Florida, March 16-19, 1997.

4. **Presentation Accepted** - An abstract, entitled “Simultaneous Sulfur and Metal Capture by Lime During Fluidized Bed Coal Combustion,” has been accepted for presentation at Fluidization IX to be held in Durango, Colorado, May 17-22, 1998.

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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 technology because metals are allowed to vaporize and condense before being collected. 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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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 project were to demonstrate the capture process, identify effective sorbents, and characterize the capture efficiency.

SCIENTIFIC DISCUSSION

Metal-Sorbent Reactions

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., Punjak et al., 1989; Uberol and Shadman, 1990; Ho et al., 1992, 1994). The following reactions between metals and sorbent constituents have been confirmed both theoretically and experimentally:

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

Note that the technology of metal capture by sorbents during fluidized bed coal combustion has never been evaluated. Due to the trace concentration and the nonuniformity nature of metals in coal, statistical approaches are essential in the evaluation.

Chemical Equilibrium

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's 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

Facilities and Procedure

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. A schematic diagram of the fluidized bed coal combustion system is shown in Figure 1. 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 samples is 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^oC are listed in Table 2.

In an experimental run, a bed of sorbent (normally 6 cm bed height) was preheated to the desired temperature under the designed operating conditions (normally 900^oC and 3 U_{mf}). A predetermined amount of coal (normally 60 g) 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, 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 (Gao and Silcox, 1993). Toxicity Characteristics Leaching Procedure (TCLP) tests were also performed to determine the leachability of the captured metals from sorbents.

Data Analysis

<u>Specific Capture Capacity (ϕ)</u> - This is defined to be the amount of metal captured by a unit mass of bed (or fly ash) sorbent. It was calculated as:

$$\phi_b = C_b - C_o \qquad (1)$$

or

$$\phi_f = C_f - C_o \qquad (2)$$

Percent Capture (ψ) - This is defined to be the percent of metal captured by fluidized bed sorbents (or fly ash sorbents) relative to the total amount of metal charged. It was calculated as:

$$\psi_b = \frac{\phi_b \text{ x } W_b}{C_c \text{ x } W_c} \text{ x } 100\% \tag{3}$$

or

$$\psi_f = \frac{\phi_f \text{ x } W_f}{C_c \text{ x } W_c} \text{ x } 100\% \tag{4}$$

All chemical symbols used in the equations are defined at the end of this report.

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 compositions 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^{o}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^{o}C, $PbSO_4$(s) is the thermodynamically preferred lead compound; however, between 950^{o}C and 1000^{o}C, $PbSiO_4$(s) is the preferred one; and, above 1000^{o}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^{o}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.

Detailed simulation results on Pb-S-SiO_2, Cd-S-SiO_2, and Cd-S-Al_2O_3 systems are also shown in Figures 2 through 4. They clearly indicate the effect of temperature on metal speciation during coal combustion with sorbents. For arsenic and selenium, efforts are currently ongoing to collect the necessary thermodynamic data for equilibrium simulations. The results will be reported in the near future.

Experimental Results

Typical experimental results indicating the effectiveness of metal capture by various sorbents are shown in Tables 6 through 10 for lead, cadmium, chromium, arsenic and selenium, respectively. Note that the results reported in these tables include only the amount captured by bed sorbents, i.e., the amount captured by fly ash sorbents is not included. It is worth pointing out at this point that the amount captured by fly ash sorbents can be significant especially for lime.

The results shown in Table 6 for lead capture indicate that all three sorbents tested are capable of in-bed capturing lead during fluidized bed coal combustion with the average capture efficiency ranging from 44% to 69%. Bauxite and zeolite appear to have better "bed sorbent" capture efficiencies than lime. Lime, however, has greater "fly ash sorbent" capture efficiency as compared to zeolite and bauxite which will be discussed later. 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 could be due to the formation of the same compound and/or alumino-silica compounds. The mechanism of lead capture by lime, however, is suspected to 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 "bed sorbent" capture efficiencies associated with the sorbents are similar to those of lead capture by bed sorbents. All three sorbents are seen to be relatively effective with an average capture efficiency being around 60%. The effectiveness of cadmium capture by bauxite and zeolite appears to suggest the formation of $CdAl_2O_4$(s) and $CdSiO_3$(s) as revealed from 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 31 and 30% which are much lower than those of lead and cadmium capture by the two sorbents. 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. For arsenic and selenium capture, the results shown in Tables 9 and 10 indicate that the average capture efficiencies of these two metals by sorbents are generally in the ranges of 13 to 34%, which are relatively low as compared to those of lead and cadmium capture.

One observation worth reporting is that the observed results strongly indicated that the amount of lead, cadmium and arsenic capture by a unit mass of sorbents was roughly proportional to the concentration of the metal in coal. Typical sets of results indicating this trend are shown in Figures 5 through 7. This trend, however, was not observed for chromium and selenium (see Figures 8 and 9). 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. A typical set of results is shown in Figure 10.

As mentioned previously, the results reported in the tables and figures include only the amount of capture by bed sorbents. It was observed that metal capture by "fly ash sorbent" can be very significant for lime and is insignificant for zeolite and bauxite. The results have indicated that, for lime, this "fly ash sorbent capture" can be as high as 200% as compared to the "bed sorbent capture" especially on arsenic and selenium. The complete results regarding this capture will be reported in the near future. It is also worth reporting that good fluidization is essential in the metal capture process. The metal capture efficiencies were observed to be much lower when the bed was operated under slugging regimes or near fixed bed conditions.

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 good fluidization operations are essential in the metal capture process. Metal capture by sorbents can be as high as 88% depending on the metal species and sorbent involved. All three sorbents tested, i.e., bauxite, zeolite and lime, were observed to be capable of effectively capturing lead and cadmium, and zeolite and lime were able to capture chromium. Arsenic and selenium capture by bed sorbents was found to be relatively ineffective. 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.

NOTATION

C_b	metal concentration in bed sorbent, ppm
C_c	metal concentration in coal, ppm
C_f	metal concentration in cyclone sorbent, ppm
C_o	metal concentration in original sorbent, ppm
d_p	particle diameter, mm
T	combustion temperature, ^oC
U	air superficial velocity, cm/s
U_{mf}	minimum fluidization velocity, cm/s
W_b	weight of collected bed sorbent, g
W_f	weight of collected fly ash sorbent, g
W_c	weight of coal, g
ϕ_b	specific capture capacity of bed sorbent, mg/Kg
ϕ_f	specific capture capacity of fly ash sorbent, mg/Kg
ψ_b	percent capture by bed sorbent, %
ψ_f	percent capture by fly ash sorbent, %

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

Coal	S	Cl	Cd	Cr	Pb	As	Se
IBC-101	4.4	0.1	1.1	31	8	2	1.5
IBC-102	3.3	0.0	0.8	7	149	32	1.3
IBC-106	3.8	0.0	0.2	10.4	6	4.1	2.0
IBC-109	1.2	0.4	<0.3	13	18	6.9	1.5
IBC-110	4.6	0.0	<0.4	11	10	4.7	2.5
IBC-111	2.0	0.0	<0.4	14	18	6.1	1.5
IBC-112	2.8	0.2	<0.3	14	27	3.3	1.6

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
As (ppm)	1.2	0.0	0.0
Cd (ppm)	2.0	3.0	3.6
Cr (ppm)	146	4.0	7.8
Pb (ppm)	43.2	60.4	72.4
Se (ppm)	1.0	0.8	0.9
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	0.15-0.3 g/min
Sorbent Size	0.4 - 0.6 mm
Sorbent Amount	22.5 - 30 g
Static Sorbent Height	3-9 cm
Air Flow Rate	1.2-5 U_{mf} of Sorbent
Combustor Temperature	900^oC
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	**$Pb_2SiO_4(s)$**	$<1000^oC$
			$PbO(g)$	$>1000^oC$
		With S	**$PbSO_4(s)$**	$< 950^oC$
			$Pb_2SiO_4(s)$	$<1000^oC$
			$PbO(g)$	$>1000^oC$
Al_2O_3	Pb	Without S	$PbO(s)$	$< 900^oC$
			$PbO(g)$	$> 900^oC$
		With S	**$PbSO_4(s)$**	$< 950^oC$
			$PbO(g)$	$> 950^oC$
CaO	Pb	Without S	$PbO(s)$	$< 900^oC$
			$PbO(g)$	$> 900^oC$
		With S	**$CaSO_4(s)$**	$> 500^oC$
			$PbSO_4(s)$	$< 950^oC$
			$PbO(g)$	$> 950^oC$

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 Bed Sorbents (ψ_b)

Coal\Sorbent	Bauxite	Zeolite	Lime
IBC-101	64	74	75
IBC-102	80	68	16
IBC-106	77	57	67
IBC-109	62	47	49
IBC-110	73	51	36
IBC-111	49	62	32
IBC-112	74	44	22
Average	**69**	**58**	**42**

Table 7. Percentage Cadmium Capture by Bed Sorbents (ψ_b)

Coal\Sorbent	Bauxite	Zeolite	Lime
IBC-101	54	52	56
IBC-102	50	58	58
IBC-106	76	72	70
IBC-109	71	88	50
IBC-110	47	58	61
IBC-111	56	30	73
IBC-112	55	49	50
Average	**58**	**58**	**60**

Table 8. Percentage Chromium Capture by Sorbents (ψ_b)

Coal\Sorbent	Bauxite	Zeolite	Lime
IBC-101	0	4	2
IBC-102	0	26	30
IBC-106	0	22	9
IBC-109	0	37	26
IBC-110	0	66	44
IBC-111	0	10	47
IBC-112	0	54	51
Average	**0**	**31**	**30**

Table 9. Percentage Arsenic Capture by Sorbents (ψ_b)

Coal\Sorbent	Bauxite	Zeolite	Lime
IBC-101	57	24	27
IBC-102	23	7	31
IBC-106	28	14	23
IBC-109	34	13	12
IBC-110	32	19	28
IBC-111	28	18	23
IBC-112	43	22	24
Average	**34**	**17**	**24**

Table 10. Percentage Selenium Capture by Sorbents (ψ_b)

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**

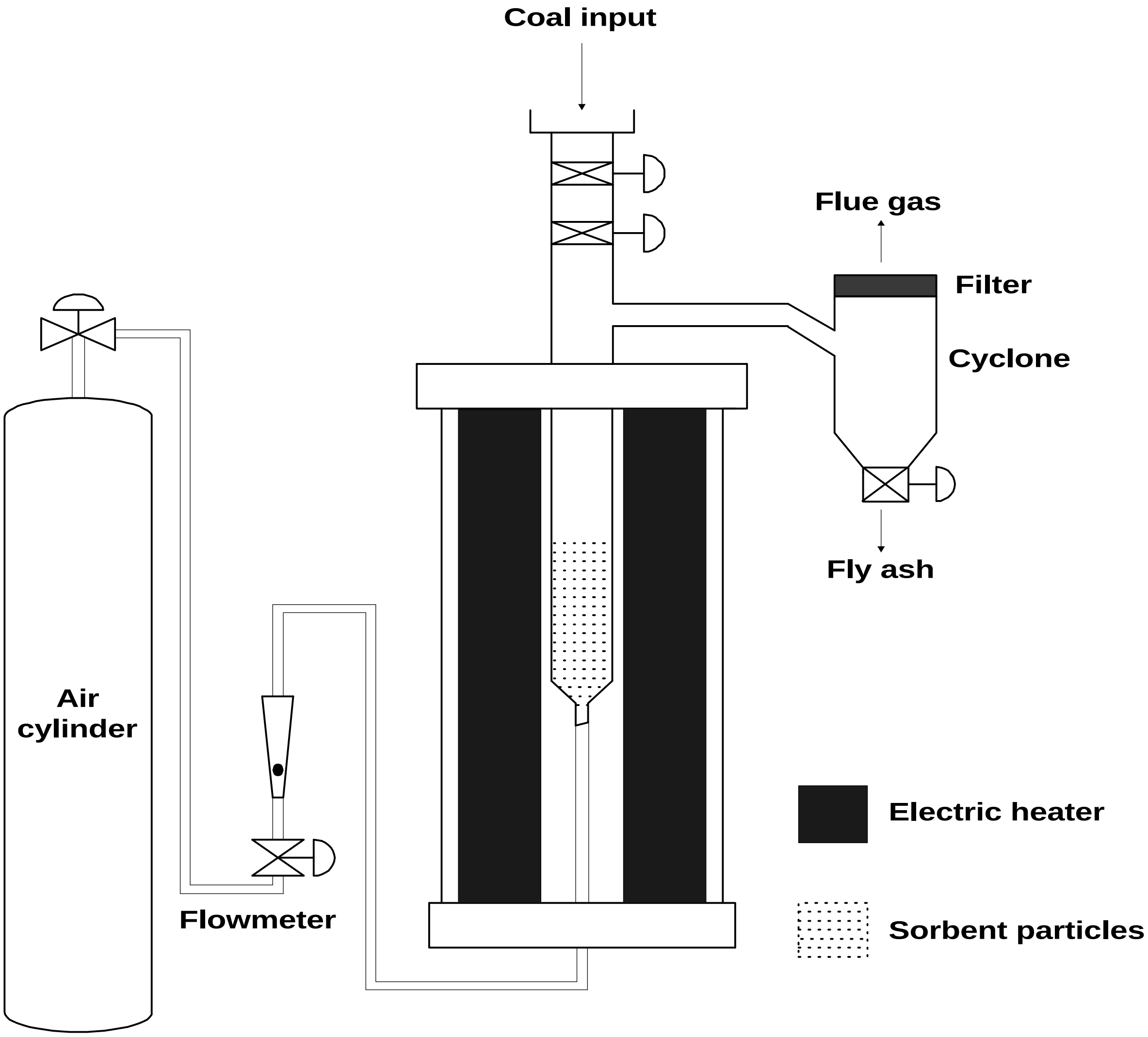

Fig. 1. Fluidized bed coal combustion system.

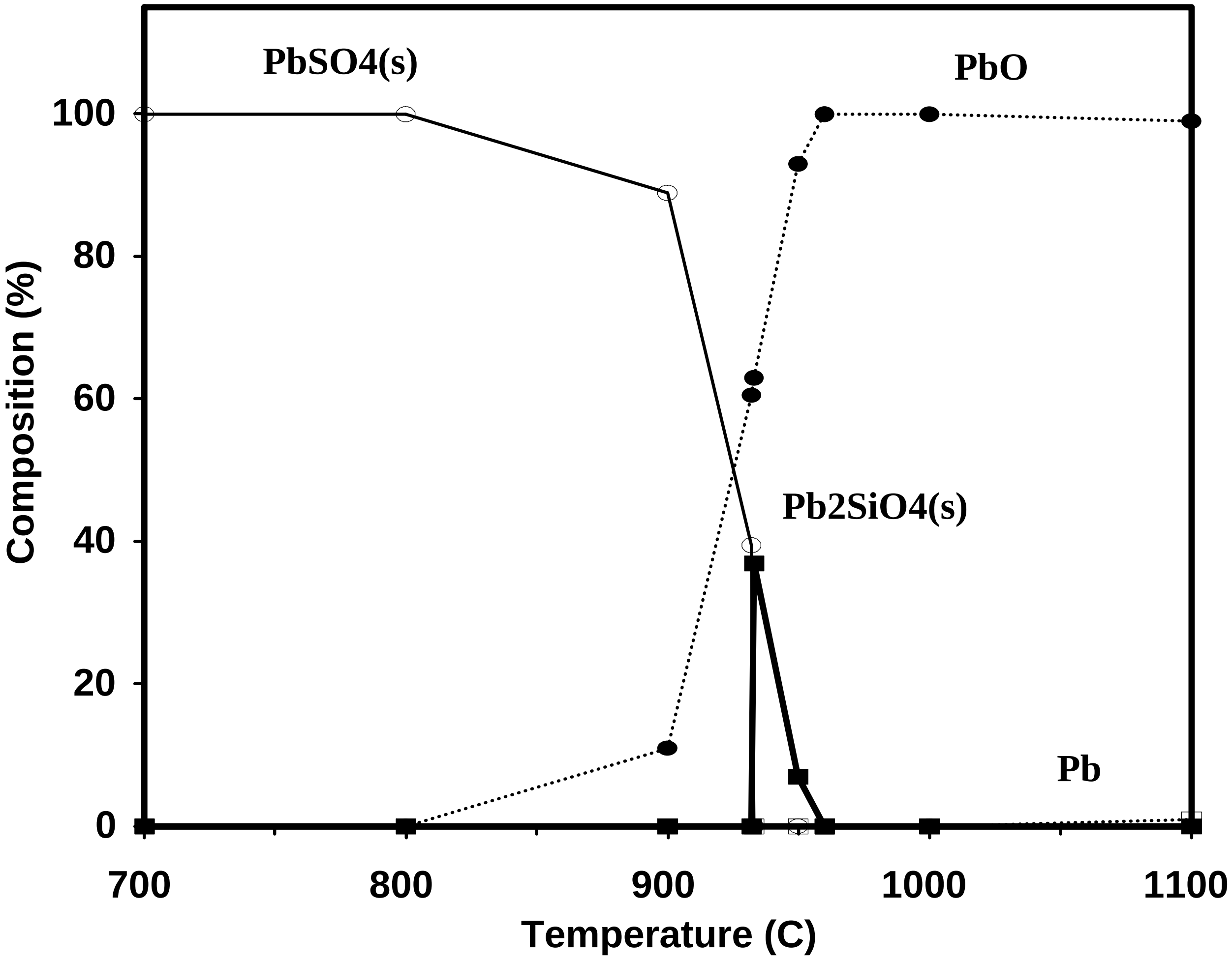

Fig. 2. Simulated lead speciation (System: Pb-S-S_iO_2).

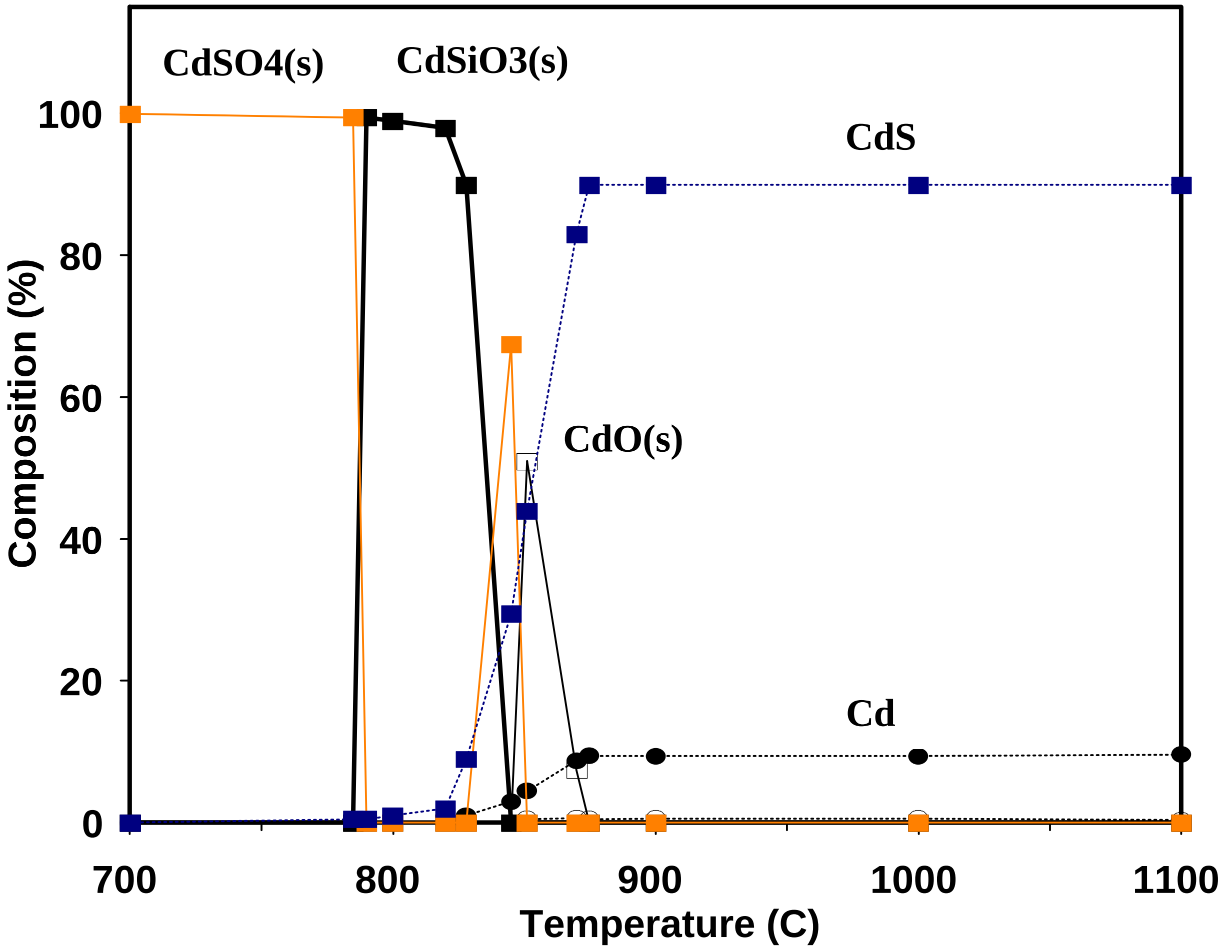

Fig. 3. Simulated cadmium speciation (System: Cd-S-S_iO_2).

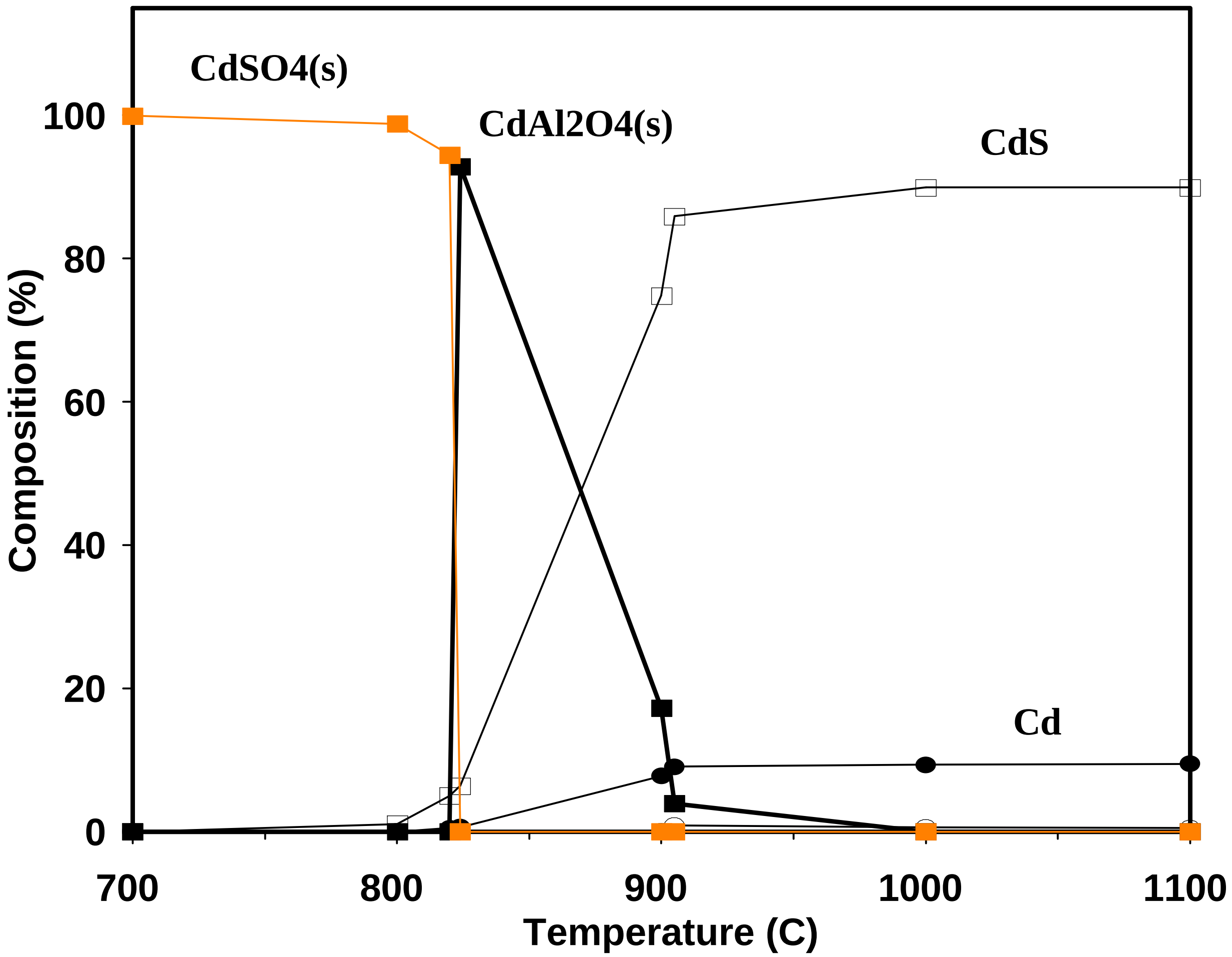

Fig. 4. Simulated cadmium speciation (System: Cd-S-Al_2O_3).

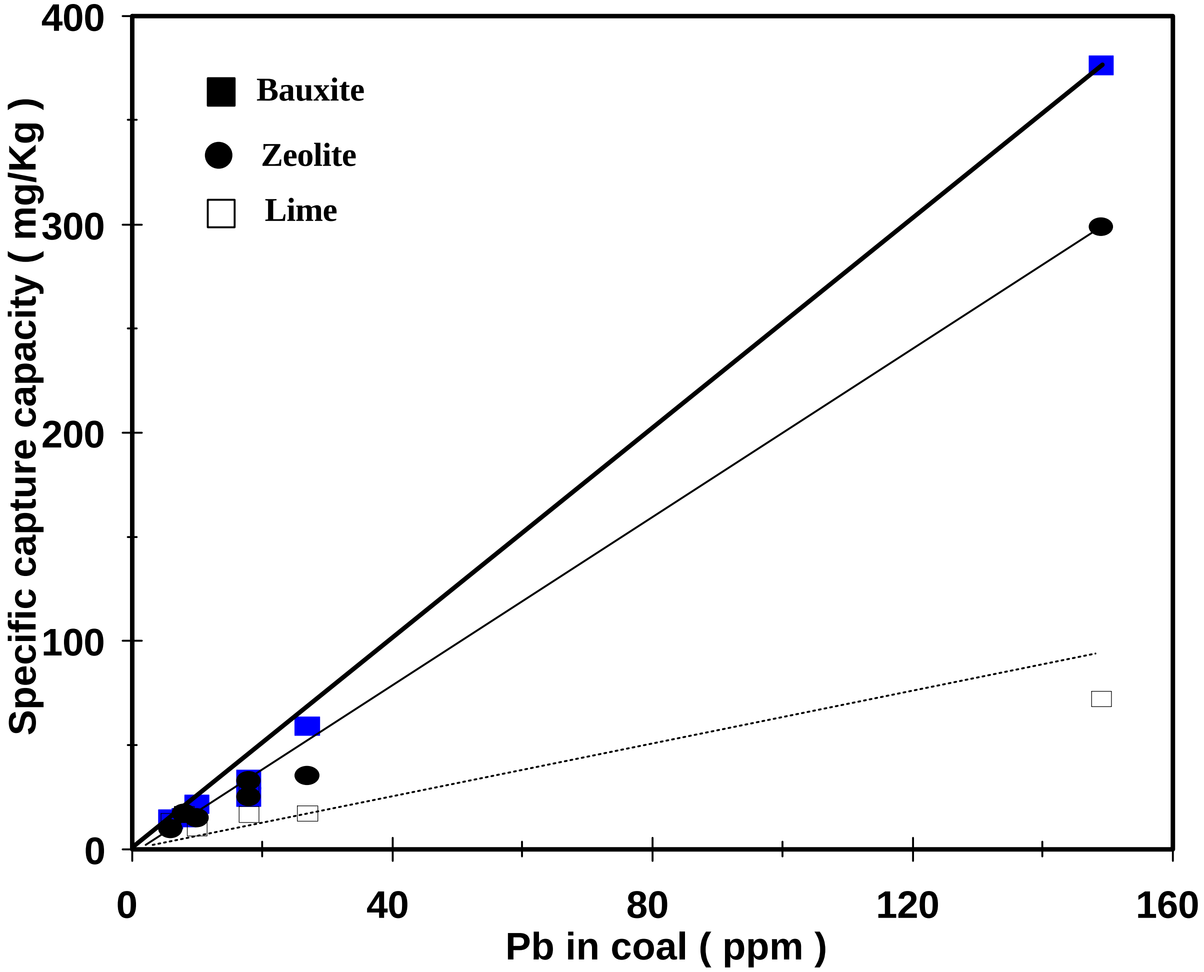

Fig. 5. Specific capture capacity of Pb by bed sorbents (ϕ_b).

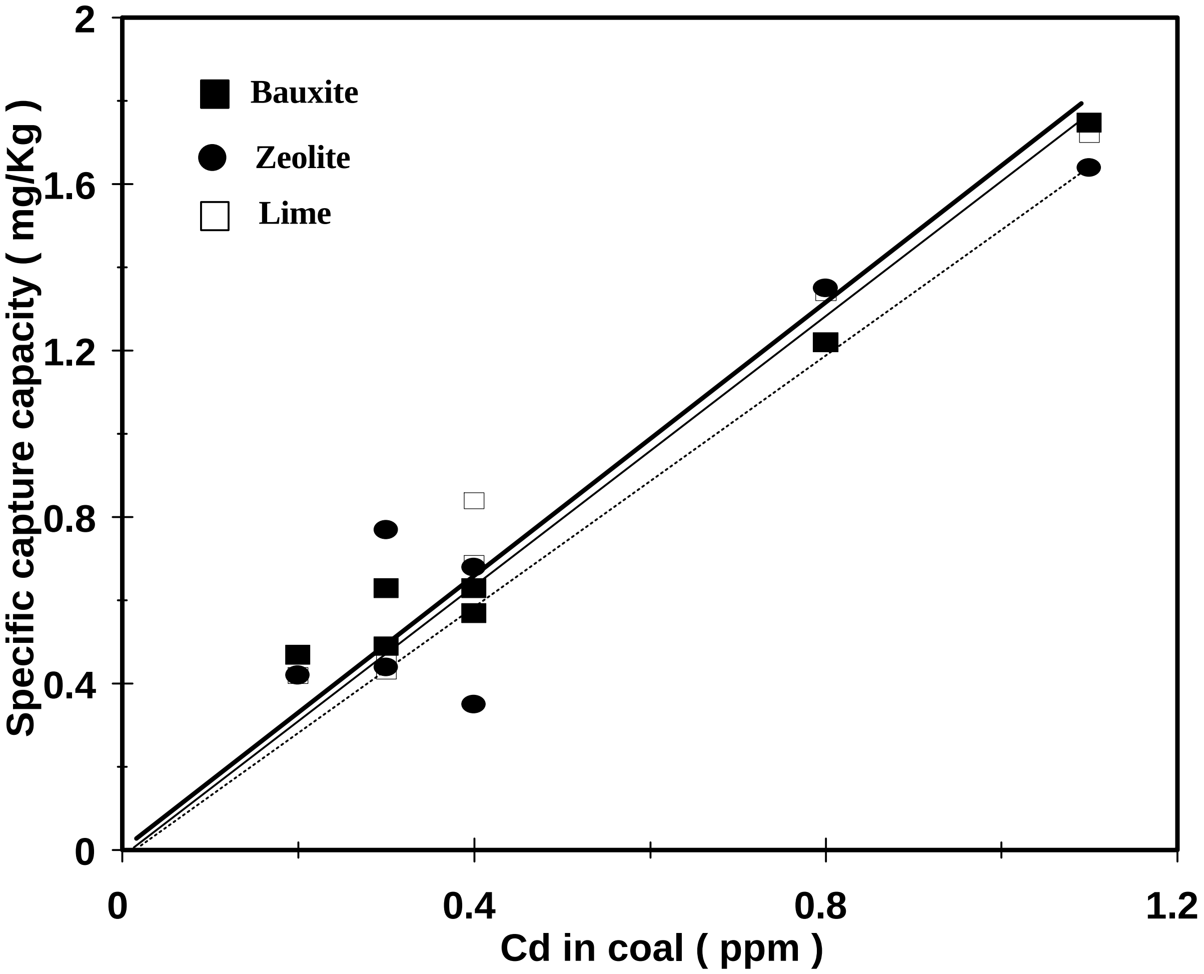

Fig. 6. Specific capture capacity of Cd by bed sorbents (ϕ_b).

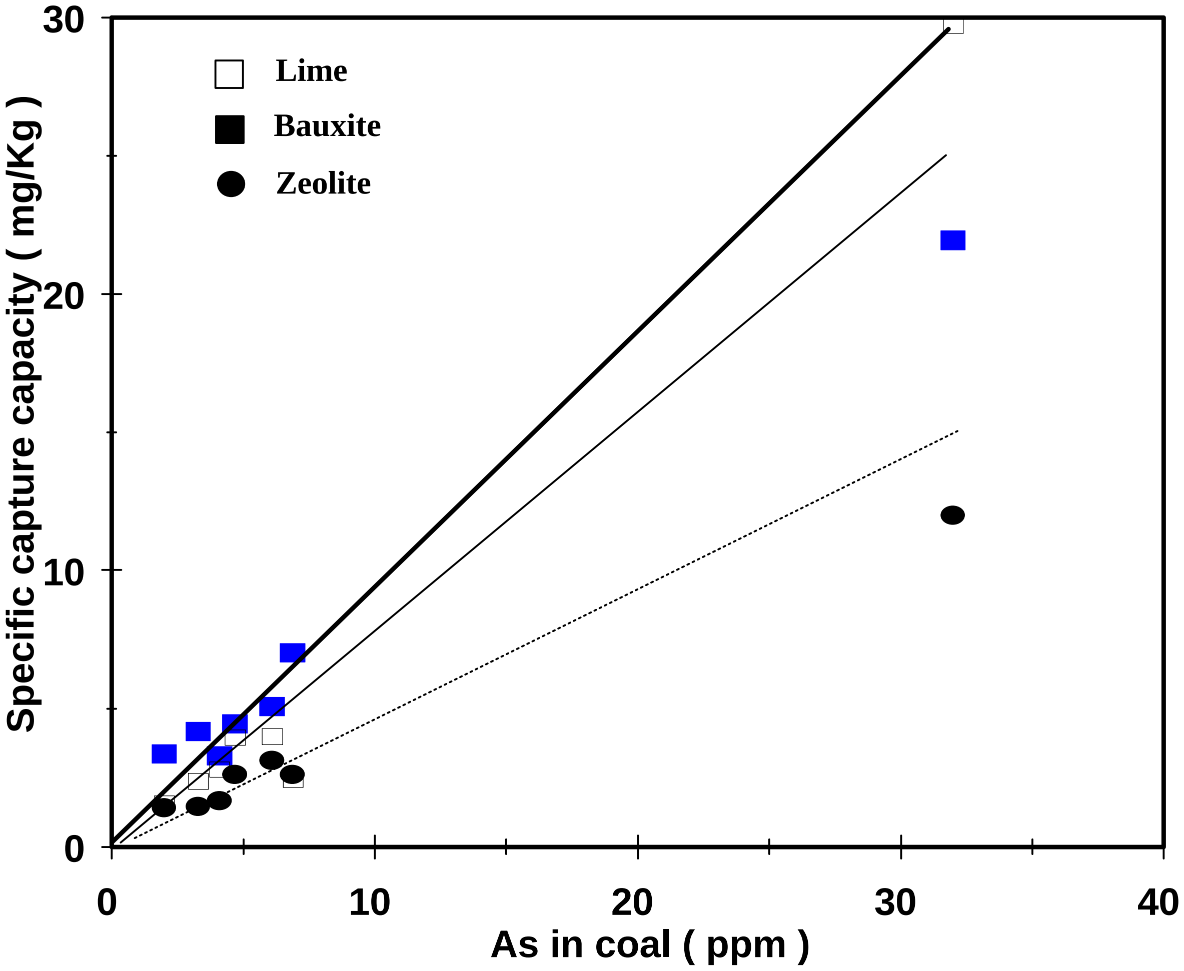

Fig. 7. Specific capture capacity of As by bed sorbents (ϕ_b).

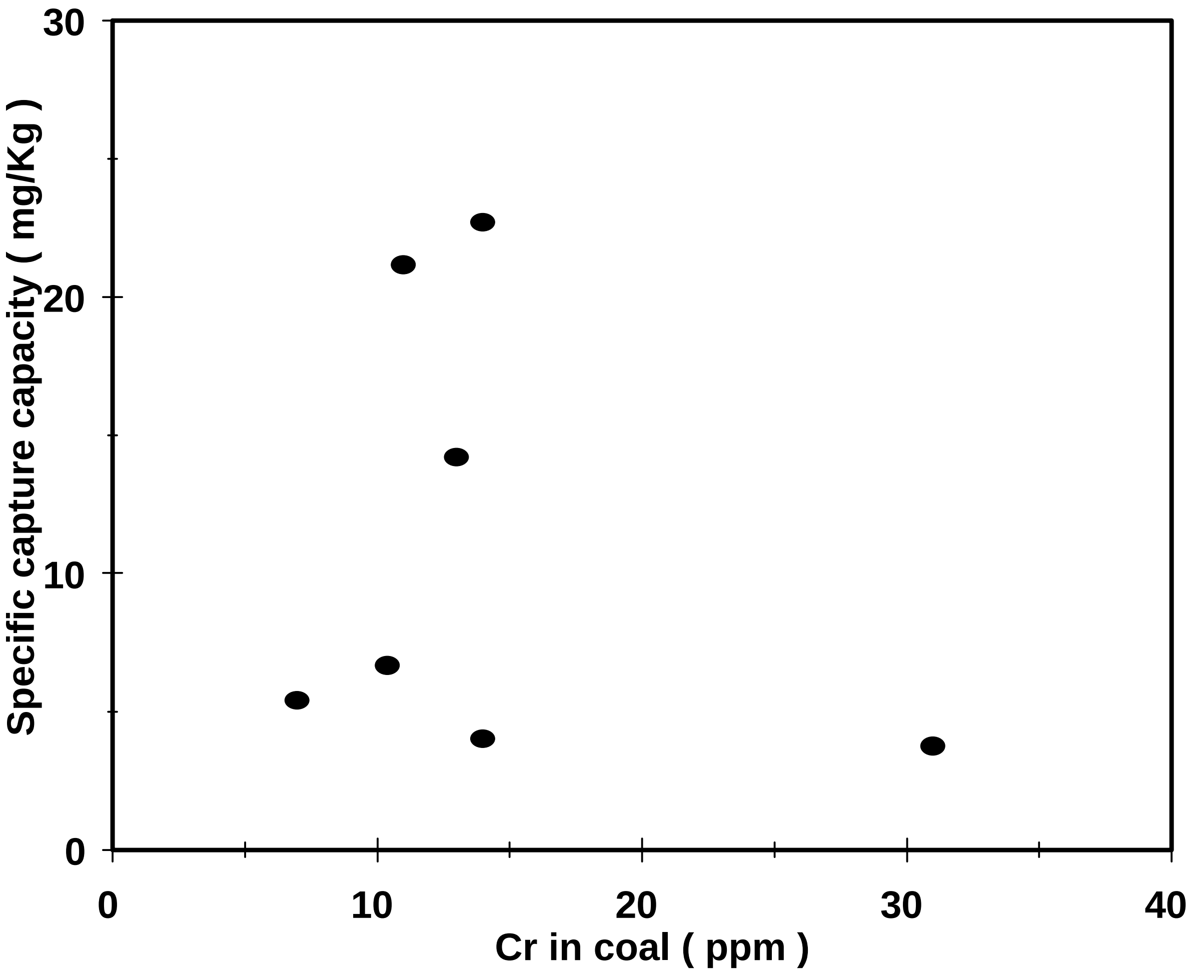

Fig. 8. Specific capture capacity of Cr by Zeolite (ϕ_b).

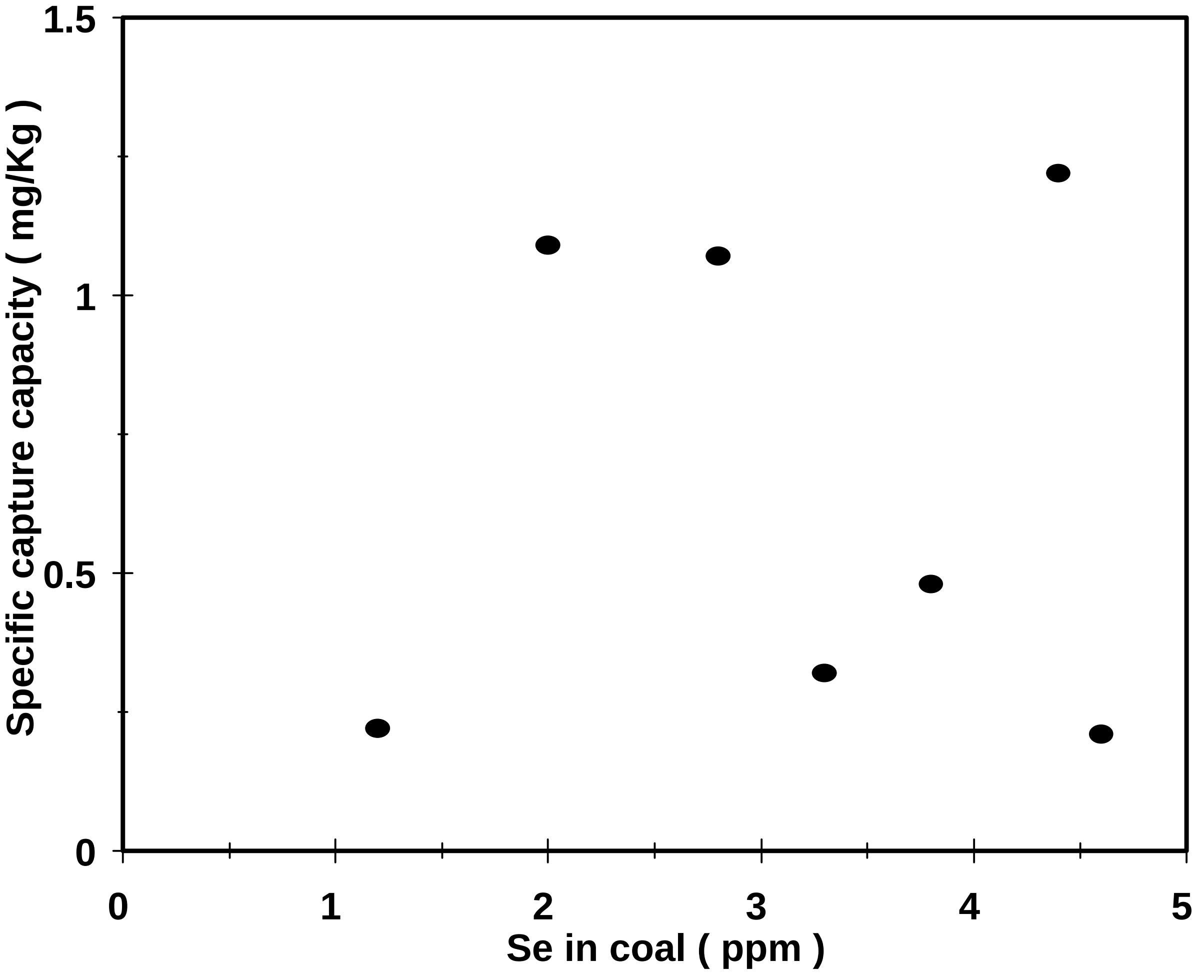

Fig. 9. Specific capture capacity of Se by Zeolite (ϕ_b).

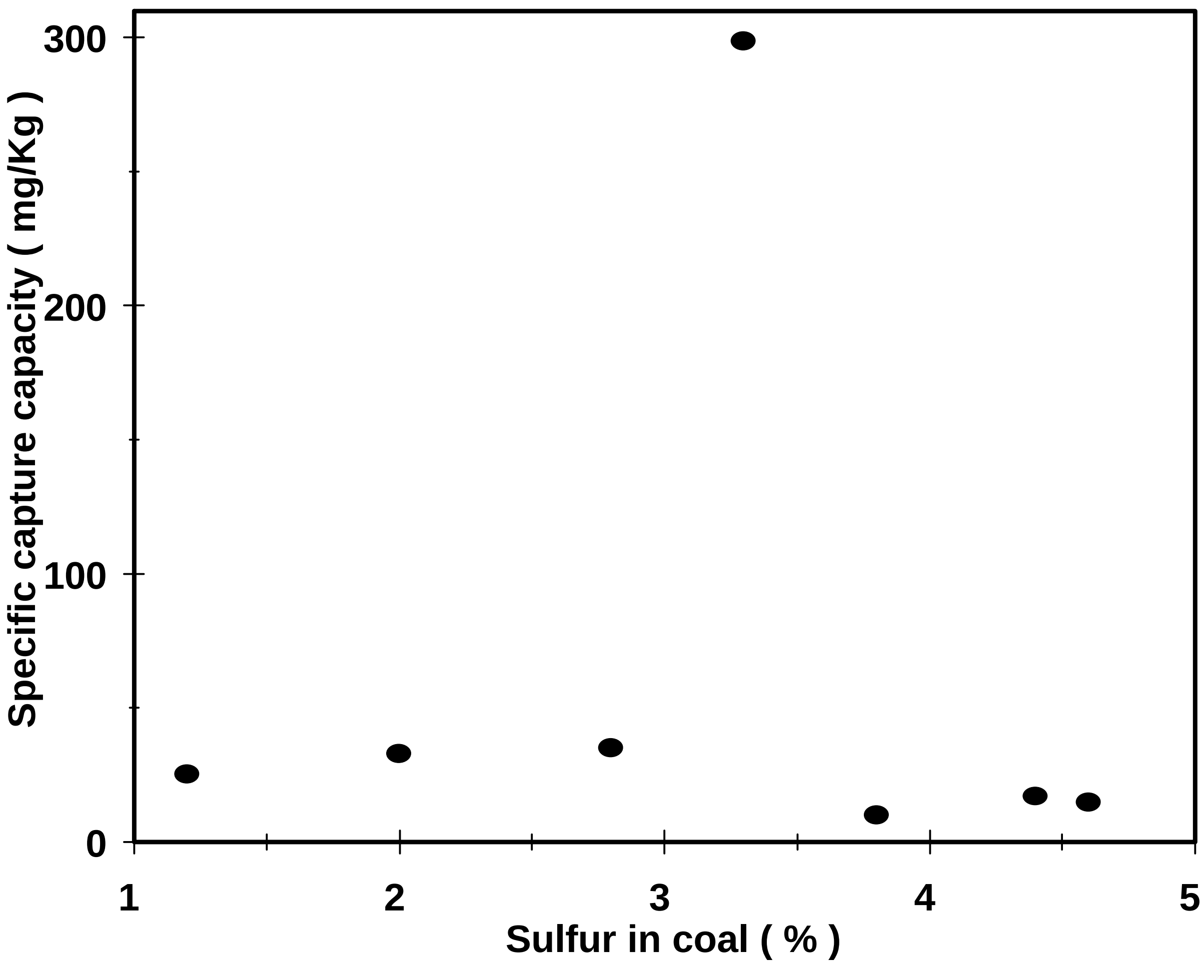

Fig. 10. Specific capture capacity of Pb by Zeolite (ϕ_b).