CO₂ sequestration and heavy metal stabilization by carbonation process in bottom ash samples from coal power plant

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Abstract

Coal-fired power plants supply roughly 50 percent of the nation's electricity but produce a disproportionate share of electric utility-related air pollution. Coal combustion technology can facilitate volume reduction of up to 90%, with the inorganic contaminants being captured in furnace bottom ash and fly ash residues. These disposal coal ash residues are however governed by the potential release of constituent contaminants into the environment. Accelerated carbonation process has been shown to have a potential for improving the chemical stability and leaching behavior of bottom ash residues. The aim of this work was to quantify the volume of CO_2 that could be sequestrated with a view to reducing greenhouse gas emissions and stabilize the contaminated heavy metals from bottom ash samples. In this study, we used PC boiler bottom ash, Kanvera reactor (KR) slag and calcined waste lime for measuring chemical analysis and heavy metals leaching tests were performed and also the formation of calcite resulting from accelerated carbonation process was investigated by thermo gravimetric and differential thermal analysis (TG/DTA).

Key words : Heavy metals, Carbonation, Coal power plant bottom ash

1. INTRODUCTION

Major environmental problems around the world are usually due to the disposal of waste materials such as industrial waste, construction waste, household waste and etc. However, industrial waste, also known as production by-products, has been investigated extensively as sustainable alternatives to Portland cement in concrete [1,2]. Recently, waste accumulation has become a major problem to the environment as well as human beings [3]. Heavy metals also contamination in air, soil, and water is a global problem that is a growing threat to human beings. There are hundreds of sources of heavy metal pollution, including coal combustion in thermal power plants [4]. Bottom ash and fly ash produced during the combustion of coal contains several toxic heavy metals like arsenic (As), lead (Pb), nickel (Ni), zinc (Zn), nickel (Ni), etc. It is now a global concern to find environmentally friendly solutions for the safe disposal of industrial waste to sustain a cleaner and greener environment.

Coal ash is one of the biggest sources of industrial waste that is produced from power plant stations. Cheriaf et al. [5] stated that 1.2 million metric tons

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of coal ash have been produced during the combustion of 2.9 million metric tons of coal. Coal is widely used in the production of electricity, steel and cement manufacturing and is, therefore, an important source of energy. However, based on Bajare et al. [6], several types of coal ash waste have been identified such as bottom ash, fly ash, boiler slag, flue gas, desulfurized material and etc. Coal bottom ash and boiler slag are coarse, granular, incombustible materials that are collected from the bottom of coal burning furnaces. The majority of coal bottom ash and boiler slag are produced at coal-fired electric utility generation stations, with some coming from coal-fired boilers or independent coal-burning electric generation facilities. The type of bottom ash or boiler slag produced depends on the type of coal-burning furnace.

Bottom ash is one of the well-known industrial waste that has been produced as a result of burning coal in a dry bottom pulverized coal boiler. Unburned material from a dry bottom boiler consists of about 20 percent bottom ash. Bottom ash is a porous, glassy, dark gray material with a grain size similar to that of sand or gravelly sand [7]. Although similar to natural fine aggregate, bottom ash is lighter and more brittle and has a greater resemblance to cement clinker [8]. Boiler slag is the molten inorganic material that is collected at the bottom of the boilers and discharged into a water-filled pit where it is quenched and removed as glassy particles resembling sand. When pulverized coal is burned in wet-bottom boilers (slag-tap boiler and the cyclone boiler), as much as 50 percent of the ash is retained in the slag tap furnace as boiler slag. In a cyclone furnace, 70 to 80 percent of the ash is retained as boiler slag, with only 20 to 30 percent leaving the furnace in the form of fly ash [7,8].

Huge amounts of limestone are quarried and used worldwide for construction and landscaping purposes. A large amount of waste is generated during cutting and sieving process of limestone which includes fragments, fine powder, and slurry. 60% to 70% of the stone is believed to be wasted in this process, of which around 30% is believed to be fine powder [9]. A large amount of limestone waste generated from the processing has no useful utilization and is disposed of as waste, occasionally used for landfill purposes.

The products of the coal combustion process are flue gas and solid byproducts (e.g., fly ash particles, bottom ash, and boiler slag). Flue gas CO₂ emissions from coal-fired power plants totaled 1718 Mt in the USA in 2011 [10]. Addressing the continuous rise of atmospheric carbon dioxide levels has become a focus of global efforts. Research in carbon capture and storage (CCS) has increased substantially in the last decade [11,12]. Current carbon storage research has been primarily concentrated on sequestering CO₂ in underground geologic formations such as saline aquifers, depleted oil and gas fields, and unmiserable coal seams. These methods of geologic sequestration have the advantage of being the relatively low cost when separated from CO₂ capture, separation, and transportation. However, potential issues associated with sequestration in geologic formations include permanence, long-term monitoring, and verification, with many unknown effects and potential risks still to be determined [13,14]. An alternative to conventional geologic sequestration is carbon mineralization, where CO₂ is reacted with metal cations such as magnesium, calcium, and iron to form carbonate minerals.

The literature to date indicates that in the coming decades mineral carbonation can play an important role in rebalancing the global carbon cycle and providing a long-term carbon storage solution [15,16] and the carbonation process can reduced heavy metal from the carbonated materials. Heavy metal extraction should be considered seriously for usage of carbonated materials since carbonation seriously effects on precipitation of minerals and contaminant leaching through neutralization. To reduce the heavy metal leaching on the carbonation process is the pH neutralization and formation of new materials from the raw minerals. Lowing pH to 8~9 minimizes dissolution of metal oxides [17] and physical changes of the waste material structures and the extraction characteristics and mechanical properties. The carbon dioxide source can be either pure CO_2 or any other CO_2 -rich gas such as certain industrial emissions. Consequently, using bottom ash CO_2 sequestration capacity to treat industrial emissions could be another interesting possibility of ash utilization. This process could both help reducing the greenhouse effect and reducing bottom ash storage duration by accelerating weathering reactions [18-21].

The focus of the research in this paper is aimed to CO_2 sequestration characteristics and capture potentials of those target waste materials such as coalfired power plants bottom ash, Kanvera Reactor (KR) slag and lime waste particles and stabilization of toxic heavy metals from bottom ash sample were investigated.

2. EXPERIMENTAL PROCEDURE:

2.1. Materials and sample preparation

In this study, we chose pulverized coal (PC) power plant bottom ash sample, kanvera reactor (KR) slag and waste lime stone was used. After receiving the samples we dried the samples at room temperature for the removal of moisture from that samples. After drying, samples were grinded by using a comminution equipment and collected the fine powder size less than 75µm for reproducible experiments. Accelerated carbonation process was conducted through bench scale reactor for stabilizing the high content of hazardous heavy metals in coal bottom ash sample, and CO₂ sequestrated in PC boiler bottom ash using with KR slag and waste lime stone as a calcium source. These experiments were conducted using a various condition with a solid-liquid ratio, temperature, and CO₂ concentrations.

In the accelerated carbonation process, gaseous CO_2 was injected into a hydrated bottom ash sample mixed with KR slag or calcined limestone as a calcium source as shown in Fig. 1. CO_2 sequestration process by carbonation method. In the carbonation

process, we used bottom ash mixed with KR slag (8:2 ratio) sample and bottom ash mixed with calcined lime (9:1 ratio) samples. These two samples were used for carbonation process to stabilized heavy metals and CO2 sequestration process. The detailed experiment conditions were conducted with a liquid-solid ratio as 10-30 dm³/kg, conducted with temperature as 20° C to 40° C and CO₂ concentration as 10%, 30%, and 100%. Other conditions of the experiments used here were injection at a rate of 1L/min. The experiment of carbonation was terminated when the measured pH was below 7 and did not change any further. Two phases are possible in bench scale carbonation process (a) liquid phase as a calcium ion (Ca^{2+}) source and (b) gas phase as carbonate ion (CO_3^{2-}) source. In a short time, the gas and the liquid mix together and form a turbulent flow for optimum mass transfer; finally, this process leads to the formation of calcite crystals.

2.2. Carbonation Process:

In this carbonation process, the CaO was hydrated to form $Ca(OH)_2$ slurry and is shifted to the carbonation reactor chamber, the gaseous CO_2 was injected into the $Ca(OH)_2$ slurry. The general mechanism involved in the carbonation process can be expressed as follows:



Fig. 1. CO_2 sequestration process by carbonation method.

$$CaO + H_2O \rightarrow Ca(OH)_2 \tag{1}$$

$$\operatorname{Ca(OH)}_{2} \xrightarrow{\text{Water}} \operatorname{Ca}^{2+} + 2\operatorname{OH}^{-}$$
 (2)

The dissociation of aqueous carbon dioxide,

$$CO_{2(aq)} + H_2O \to CO_3^{2^-} + 2H^+$$
 (3)

These processes produce a fast supersaturating (S_1) of solution with respect to calcite,

$$S_{1} = \frac{(Ca^{2+})(CO_{3}^{2-})}{K_{sp}} > 1$$
(4)

Where (Ca^{2+}) and (CO_3^{2-}) are the activities of calcium and carbonate ions in the solution, respectively, and K_{sp} is the thermodynamic solubility product of calcite.

$$\operatorname{Ca}^{2+} + \operatorname{CO}_{3}^{2-} \to \operatorname{CaCO}_{3}(\operatorname{nuclei})$$
 (5)

Finally, the crystal growth occurs spontaneously until the equilibrium calcite and the solution is reached

$$CaCO_3(nuclei) \rightarrow CaCO_3(calcite)$$
 (6)

The carbonation reaction starts from the hydration of carbon dioxide and the ionization of calcium hydroxide or calcium oxide, as shown in Eqs. (1), (2), (3), and (4). The aqueous carbonation of calcium hydroxide in contact with compressed CO_2 at moderate temperature allows the synthesis of fine particles of calcite. Carbonation is a strongly exothermic reaction. The reaction mechanism of calcite precipitation via aqueous carbonation of $Ca(OH)_2$ was then described by the global reaction.

3. RESULTS AND DISCUSSION:

3.1. Characteristics of PC power plant bottom ash, KR slag, and calcined limestone powder:

X-ray fluorescence (XRF) and X-ray diffraction (XRD) were used in the analyses of the chemical compositions and crystalline structures of PC bottom ash, KR slag and calcined lime waste samples. Table 1 shows the chemical compositions PC power plant bottom ash sample. The major components of bottom ash are 57.75% SiO₂, 21.29% Al₂O₃, 8.74% Fe₂O₃, 1.34% MgO and 4.31% of CaO. These XRF results indicated that the calcium source (4.31%) was very less amount present in the bottom ash sample.

The chemical compositions of KR slag results as shown in Table. 2. The major components of the KR slag are 61.84% of CaO, 6.94% of SiO₂, 2.25% of Al₂O₃, 6.81% Fe₂O₃ and 1.81% MgO are present remaining elements such as K₂O, Na₂O, TiO₂, MnO, and P₂O₅ are less than 1% (<1%) present in the sample. The results indicate that KR slag has the major calcium source, it more than 60% of CaO present in the sample.

Table 1. PC type power plant Bottom ash XRF Chemical analysis result (Unit: wt %)

Sample	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	K ₂ O	Na ₂ O	TiO ₂	MnO	P ₂ O ₅
PC Bottom ash	57.75	21.29	8.74	4.31	1.34	1.07	0.72	0.13	0.09	0.31

Table 2. KR (Kanvera Reactor) Slag XRF chemical analysis result (unit: wt %)

Sample	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	K ₂ O	Na ₂ O	TiO ₂	MnO	P ₂ O ₅
KR slag	6.94	2.25	6.81	61.84	1.81	0.11	0.02	0.19	0.85	0.21

Sample	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	K ₂ O	Na ₂ O	TiO ₂	MnO	P ₂ O ₅
Waste lime	4.67	2.64	0.44	81.10	1.99	0.26	0.12	0.10	0.30	0.02

Table 3. Calcined lime waste XRF chemical analysis (unit: wt %)



Fig. 2. XRD analysis of PC power plant bottom ash, KR slag, and calcined waste limestone.



Fig. 3. The carbonation process for different solid-liquid ratio (10-30 dm³/kg) with different temperature (20-40oC) at 100% CO₂ concentration.

The wasted calcined limestone chemical compositions as shown in Table. 3. The major components of the calcined limestone are 81.10% of CaO, 4.67% of SiO₂, 2.64% of Al₂O₃, and 1.99% of MgO are present remaining elements such as Fe₂O₃, K₂O, Na₂O, TiO₂, MnO, and P₂O₅ are less than 1% (<1%) are presented in waste limestone sample.

In Fig. 2 shows XRD results of PC power plant bottom ash, KR slag, and calcined waste limestone. The results indicated that high amount of quartz (SiO₂) and less amount of lime (CaO) are present in PC power plant bottom ash sample. KR slag and calcined lime waste having a high amount of lime (CaO) and less amount of quartz (SiO₂) are present.

3.2. CO₂ sequestration of PC power plant bottom ash along with KR slag.

In the CO₂ sequestration by accelerated carbonation process, we chose mixed pulverized coal (PC) power plant bottom ash with kanvera reactor (KR) slag (8:2 ratio) with fine particles (<75 μ m) and we can estimate the CO₂ sequestration potential of solid residues of before and after carbonation. The experiment conditions were conducted with a liquid-solid ratio as 10-30 dm³/kg, temperature as 20 °C to 40 °C and CO₂ concentration as 10%, 30%, and 100%. The experiment of carbonation was terminated when the measured pH was below 7 and did not change any further.

3.2.1. The effect of the liquid-solid ratio of coal bottom ash sample by carbonation:

The mixed PC power plant bottom ash sample was used for the carbonation process with different liquid-solid-ratio (10-30 dm³/kg) at different temperatures (20-40 °C). In Fig. 3, show that when the liquid-solid-ratio of bottom ash was increased, the carbonation reaction time also increased. These results indicated that the reaction was terminated quickly due to the low content of alkaline components that can react with CO₂ in the bottom ash sample.

3.2.2. Temperature effect (20-40℃) on the carbonation process with the different liquid-solid ratio (10-30 dm³/kg) of coal bottom ash sample.

The carbonation process at different temperatures with different liquid-solid ratios as shown in Fig. 4, these results clearly indicated that when the liquid-solid ratio was increased along with temperature the carbonation time also increased due to the low solubility of a CO₂ gas at high temperatures. The carbonation process refers to carbon dioxide dissolved in a liquid, and the rate at which carbon dioxide dissolves or is soluble depends on temperature. When the temperature is raised, the rate of dissolution in liquid is decreased, and when the temperature is lowered the rate of dissolution in liquid is increased, because at high temperatures the molecules within that substance gain energy and move more rapidly and the dissolved carbon dioxide molecules move faster and escape the liquid more rapidly, simultaneously the carbonation time also increased at high temperatures.

3.2.3. The effect of CO₂ concentrations (10, 30 and 100%) with different liquid-solid ratio (10-30 dm³/kg) for carbona-tion process at 20 °C temperature.

To evaluate the effect of CO₂ concentrations on the bottom ash carbonation process, several experiments were performed at 10% to 100% CO2 concentrations. Fig. 5 shows the CO₂ concentrations effect of carbonation process with the different liquid-solid ratio at 20°C. From these figures was obvious that CO₂ concentrations greatly influences the reaction kinetics. When the 10% concentration of carbon dioxide was used for the carbonation with 10-30 dm³/kg liquid-solid (LS) ratio, the carbonation process was increased from 70min to 175min reaction time, these CO_2 concentrations were increased up to 30% CO_2 concentration along with 10-30 dm³/kg liquid-solid (LS) ratio, the carbonation process was increased from 15min to 75min reaction time and the pure 100% CO_2 concentrations were used with 10-30 dm³/kg liquid-solid (LS) ratio, the carbonation time was increased from 5min to 20min reaction time due to the less source of carbonate (CO₃²⁻) ions and increasing Ca²⁺ ion source in the solutions are present along with increasing liquid-solid (LS) ratio from 10 to 30 dm³/kg in the solution for carbonation process.

These results also clearly indicated that the CO_2 concentrations were increased from 10% to 100%, the carbonation reaction time was drastically de-



Fig. 4. Temperature effect on the carbonation process with the different liquid-to-solid ratio at 100% CO2 concentration.



Fig. 5. CO₂ concentration effect of carbonation process with the different liquid-solid ratio at 20oC temperature.



Fig. 6. TG / DTA results before and after carbonation of PC Bottom ash-KR Slag and PC Bottom ash-Calcined lime waste samples.

creased from 70min to 5min in 10 dm³/kg liquid-solid (LS) ratio solution, 150min to 10min in 20 dm³/kg liquid-solid (LS) ratio solution and 175min to 20min in 30 dm³/kg liquid-solid (LS) ratio solution by carbonation process, because the high amount of carbonate (CO_3^{2-}) ions available in the carbonated solution.

3.3. Thermal analysis:

Samples were analyzed by simultaneous TG/DTA before and after a complete carbonation as shown in Fig.6. The TGA curves are plotted in wt% (in percentage) on the first axis and the second axis shows DTA (μ V) value. The PC power plant bottom ash with KR slag (8:2) sample mass decreased by 0.152wt% before carbonation, and after carbonation, it decreased by 4.382wt%. As a result, CO2 solid-

ification rate was 4.23% by carbonation, and 42.3g of CO2 could be stored in 1kg of KR slag contained bottom ash sample. But in PC power plant bottom ash mixed with waste calcined lime (9:1 ratio) sample mass decreased by 0.275wt% before carbonation, and after carbonation, it decreased by 5.921wt%. As a result, CO2 solidification rate was 5.646% by carbonation, and 56.46g of CO2 could be stored in 1kg of calcined lime waste containing bottom ash sample, these results as shown in Table. 4, it indicates the specific CO₂ sequestration capacity was high in calcined lime waste containing bottom ash sample (9:1 ratio) was estimated by carbonation process.

3.4. Leaching concentrations of heavy metals and stabilized with carbonation.

Coal is a carbon-rich combustible material con-

taining organically bound mineral matter. This organic material is released during coal combustion to form an ash residue [22]. Various factors that control the concentrations of trace elements in the coal and ashes include element sources, modes of element occurrence, combustion conditions, volatilization-condensation mechanism, and particle size of the ash [23]. The PC power plant coal bottom ash has different concentration of heavy metals was observed as As, Pb, Cu, Cr, Cd, Ni, and Zn etc. In order to evaluate the environmental stability of PC coal bottom ash heavy metal content and leaching test were conducted before and after carbonation process. In the carbonation process, CO₂ gas was injected at a rate of 1L/min and 400rpm stirring speed at 20°C temperature was maintained and the carbonation process was terminated when the measured pH was below 7 and sample was filtered, the leachate solution was measured by ICP-MS for heavy metal analysis.

We investigated the leaching concentrations of heavy metals in PC power plant coal bottom ash sample. The heavy metals leaching characteristics are reported in mg/kg as shown in the Table. 5, for PC power plant coal bottom ash sample. The results indicated that Ni has been eluted in the leaching solution, after carbonation process these Ni heavy metal are not eluted in the leaching process, it indicates Ni has been stabilized effectively by carbonation process.

4.CONCLUSIONS

The effects of accelerated carbonation process have the major advantage of chemically binding CO₂ and at the same lowering the hazardous nature of bottom ash. This accelerated carbonation process can chemically block the leaching of Ni and other heavy metals. The CO₂ volume that can solidify in the material has been quantified and is on average 40-60g of CO₂ / kg of PC coal bottom ash along with KR slag or waste calcined limestone. Tests performed to improve the efficiency of the process showed that the CO₂ partial pressure can drastically modify the kinetics of the reaction: the higher the CO₂ partial pressure, the faster the reaction. However, it does not increase the solidified volume of CO₂. Our research results show the carbonation studies using PC power plant bottom ash and CO2 in order to increase the recycling percentage of the ash and reduce the concentration of atmospheric CO₂. This process is an eco-friendly for the removal of heavy metals from PC power plant bottom ash samples.

Mixed samples with ratio	Before carbonation wt %	After carbonation wt%	CO ₂ solidification rate (%)		
PC Bottom ash : KR slag (8 : 2)	0.152	4.382	4.230		
PC Bottom ash : Waste lime (9 : 1)	0.275	5.921	5.646		

Table 4. PC Bottom ash-KR Slag-complex treatment of waste limestone CO₂ before and after carbonation

Table 5. The analysis result of heavy metal leaching of PC boiler bottom ash sample before and after carbonation process.(Unit: mg/L) (N.d. = Not detected).

Heavy metals		As	Pb	Cu	Cr	Cd	Ni	Zn
PC Bottom ash	Before carbonation	N.d.	N.d.	N.d.	N.d.	N.d.	0.01	N.d.
	After carbonation	N.d.						

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