

# Carbon Dioxide Capture with Accelerated Carbonation of Industrial Combustion Waste

Jong Soo Cho, Soon Mi Kim, Hee Dong Chun, Gun Woo Han, Chang Hoon Lee

**Abstract—** One of the options of carbon dioxide capture and storage is the accelerated carbonation of industrial waste. The chemical process of carbonation of those materials and extraction characteristics were studied. Several target materials were screened based upon their physical and chemical characteristics for carbonation, amount of domestic production, and transportation distance between the waste production facility and carbon dioxide generation source. Selected candidate materials were coal combustion ash and municipal solid waste incinerator (MSWI) ash. Carbon dioxide capture potentials were estimated from the chemical compositions of alkaline materials such as CaO, MgO, Na<sub>2</sub>O, and K<sub>2</sub>O and acid neutralization capacities. From the carbon dioxide capture potentials and annual production of waste, the total amount of CO<sub>2</sub> removal by carbonation of industrial combustion waste in Korea could be estimated.

**Index Terms —** carbonation, CO<sub>2</sub>, capture potential, industrial combustion waste

## I. INTRODUCTION

Motivated by concerns about climate change, technical solutions are searched to minimize these harmful consequences. The main actions include: (1) increase of the efficiency of energy conversion, (2) reduction of energy demand and (3) use of carbon free energy sources. The total amount of the greenhouse gas (GHG) emission from Korea reported to be the ninth in the world (590 million ton CO<sub>2</sub> equivalent in 2004) and ranked first in the rate of production increment among OECD countries. Along with many other governments in the world working very hard to solve this difficult problem, the Korean government declared that it would reduce the global warming gases. It announced that it would cut its greenhouse gas (GHG) by 30% by 2020. The

target was roughly equivalent to cutting GHG emissions 4% by 2020 compared with 2005. In doing so, the government aimed to cut the 2020 “business as usual” projection from 813 million tons of emissions to approximately 569 million tons [1]. The Korean government’s target-setting was a unilateral action separate from the Copenhagen summit’s outcome and carbon emission cuts of other countries. To achieve this goal of reduction, it is required to develop economical processes to reduce CO<sub>2</sub> emission. The energy sector produces 85% of the total GHG emission in Korea. Using the well-developed amine process, CO<sub>2</sub> capture with \$70 per ton would cost about \$21 billion and could damage the industrial productivity seriously. Geological storage in this small country may not be feasible unless the safety of the underground or deep-ocean storage can be assured. In addition to other options, we are trying to develop CO<sub>2</sub> capture and storage processes with natural minerals and industrial wastes. CO<sub>2</sub> capture processes should be economical, not require additional storage space, and produce valuable materials after CO<sub>2</sub> treatment. Availability of natural minerals such as olivine and serpentine in Korea is very limited that CO<sub>2</sub> capture and storage with industrial wastes such as slag from steel making byproduct, coal combustion byproduct, and construction and demolition waste is one of the options suitable to meet the goal of CO<sub>2</sub> reduction in Korea.

We investigated the possibilities of utilizing the industrial wastes which are capable of capturing CO<sub>2</sub> through chemical and physical processes. We estimated the capacity potential from the total amount of domestic production. CO<sub>2</sub> capture potentials were estimated from the carbonation potential per unit mass and the annual domestic production of the target waste. Carbonation processes and sequestration characteristics were investigated and CO<sub>2</sub> capture potentials of those target waste materials were estimated. We followed the procedure reported by Huijgen and Comans [2] in Energy Research Center of Netherland, ECN. The candidate materials were screened based on following characteristics; 1) solid (liquids are possible, but solid may be advantageous in storage, 2) inorganics, 3) alkali material (pH > 8) to react with acidic CO<sub>2</sub> solution, 4) uncarbonated Ca and Mg contents (preferred to alkali metals and Fe, Ba, and Sr), 5) domestic byproduct. Based upon these screening criteria, the following materials were selected; municipal solid waste incinerator (MSWI) ash including bottom ash and fly ash, acidic pollution control residue, coal combustion byproducts including bottom ash and fly ash, slag from steel manufacturing processes including steel slag and blast furnace slag, and construction wastes including waste

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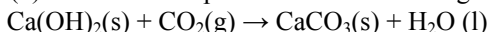
cement, concrete, and asbestos containing materials. An important factor considered in selection was the distance between waste producing facilities and CO<sub>2</sub> sources. Final points were the marketability of the carbonation products and the environmental impacts of carbonation, i.e. reduced heavy metal extraction potential from the carbonated materials. Heavy metal extraction should be considered seriously for usage of carbonated materials since carbonation seriously affects 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. It is reported that lead and zinc in MSWI bottom ash was trapped in the newly formed carbonated salts [3]. Lowering pH to 8~9 minimizes dissolution of metal oxides [4]. Physical changes of the waste material structures change the extraction characteristics and mechanical properties. Since many waste materials are already recycled and reused, the carbonation products should maintain some added value after treatment such as more stable physical properties and valuable byproducts such as precipitated carbonated calcium or magnesium.

## II. CARBONATION REACTION

The increase in atmospheric concentration of CO<sub>2</sub> is forcing the international community to develop technologies to capture this greenhouse gas [5]. To compensate the cost, new attempts have been made to use alkali wastes from electricity generation, concrete, cement from construction waste as raw materials for carbonation. Those wastes are frequently produced from CO<sub>2</sub> sources and chemically unstable compared to those from geological resources. Their chemically unstable characteristics make the pre-treatment process simple and require less energy [6] [7]. This process was proposed initially by Seifritz [8] and systematically studied by Lackner et al., [9]. Since then, quite a large number of theoretical and experimental works have been reported. In addition, metal stabilization in alkaline waste through fast carbonation provides the chance of reducing the hazardous metal extraction that carbonation enables engineering application and safe disposal of waste.

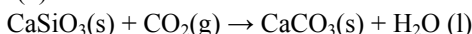
Major carbonation reactions including major industrial wastes and their applications are:

(1) Reaction with portlandite in case of high pH



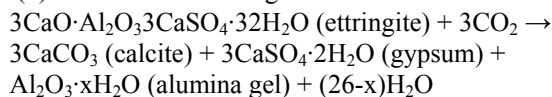
This reaction includes the carbonations of cement and MSWI bottom ash [10].

(2) Ca-silicate reaction



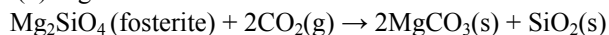
This reaction was reported for carbonation of coal combustion by-product, fly ash [11].

(3) Ca-mineral in ettringite



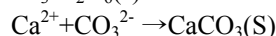
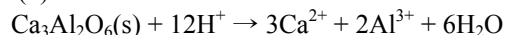
Oil residue weathering was reported with this reaction [12].

(4) Mg-silicate reaction



Oil shale residue was reported to include the above reaction [12].

(5) Tricalciumaluminate Reaction



This reaction was typical for bauxite residue (red mud) neutralization with CO<sub>2</sub> [13].

In this paper, analysis was focused on three target materials. The first one is the coal combustion ash, the second one is the municipal solid waste incinerator ash and the third one is the oil shale combustion residue. The coal combustion ash is a major product of electricity generation plants which are also major contributors of CO<sub>2</sub> emission [11], [14], [15]. About 40% of total electrical energy in the world is generated with coal combustion. In 2003, 5 billion tons of coal was consumed and its consumption is expected to be increased. A power plant generating 500MW produces about 3 million tons of CO<sub>2</sub> every year. About 10% of coal combusted becomes ash and its production reaches about 600 million tons per year in the world. In Korea alone, the amount of ash production is about 6 million tons per year. The coal combustion ash is distinguished as bottom ash and fly ash. About 90% of ash is collected as the fly ash in the electrostatic precipitator and filters of the bag-house and the rest of ash is from the bottom of the furnace. A large part of fly ash produced is being used as a cement alternative in concrete, but the production of fly ash absolutely overwhelms the consumption as the cement alternative in the world. Only about 30% of total production is being consumed as the construction material. Other applications of coal fly ash include production of zeolite and heavy metal adsorbent for contaminated soil and acid mine drainage treatment. Other applications are continuously being sought.

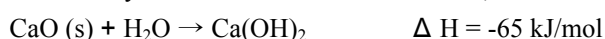
The fast carbonation process has been developed for various ash products [16], [17], [18], [19], [20], [21], [22]. Rho et al. [23] reported the biogeochemical carbonation process with coal fly ash containing a large amount of metals. Recently, Soong et al. [15] suggested wet mineral carbonation using coal fly and brine solution for CO<sub>2</sub> capture. This process was found to generate a high purity CaCO<sub>3</sub> product. The use of CaO-rich fly-ash as reactant to sequester CO<sub>2</sub> via aqueous mineral sequestration could be an very attractive option for CO<sub>2</sub> mitigation in punctual sources at the industrial scale. Uliasz-Bochenczyk and Mokrzycki [14] investigated the optimum conditions for CO<sub>2</sub> capture with various kinds of coal fly ash. They investigated the fly ash producing conditions, coal combustion processes, original resources of coal (hard coal and brown coal), and chemical compositions of the original coal resources.

The other combustion waste is the ash from the municipal solid waste incinerators which produce the bottom ash, fly ash and the air pollution control residue [24], [25], [26], [27]. These ashes are alkaline in nature, with pH values ranging from 9.5 to 11.5, and contain alkaline and alkaline earth elements and metal oxides, which would form hydroxides during hydrolysis. The elemental composition of MSWI bottom ash depends primarily on the composition of the feed stock and on the type, capacity and operational temperature of the incinerator. The carbonation of MSWI bottom ash has

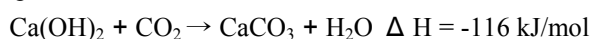
been investigated to produce mineralogical changes similar to those occurring as a result of natural weathering and to induce a related reduction in trace metal mobility [24], [28], [29]. Recent studies also focused on the principal carbonation mechanisms and controlling operating parameters, such as temperature, CO<sub>2</sub> partial pressure, water content, residence time as well as particle size. The exclusively adopted carbonation route was the direct aqueous process, however different water contents were used, with values of the water-to-solids ratio either in the low range (typically 0.05–0.6 w/w), or in the range corresponding to slurry-phase conditions (typically 5–20 w/w). Other operating parameters were the reaction pressure from atmospheric to 17 bars, the reaction temperature between ambient and 50°C, CO<sub>2</sub> percentage in gas phase from 0.03 to 100% and the residence time from a few hours to a few weeks. The mechanisms involved in the carbonation of MSWI bottom ash are not yet fully known, due to the material's complex chemistry and mineralogy. However, it is likely that more than one alkaline oxide-containing mineral (including Ca and Mg silicates) reacts with CO<sub>2</sub> in addition to calcium hydroxide. Most of the studies hitherto reported have considered a simplified approach for the carbonation of Ca(OH)<sub>2</sub> via the aqueous route [30], [31]. The rate limiting steps of the carbonation reaction involve the dissolution of calcium from the solid matrix into the liquid phase and the diffusion of CO<sub>2</sub> into the pore system. The kinetics of CO<sub>2</sub> uptake is generally characterized by two reaction steps: (1) a first steep increase in the rate of CO<sub>2</sub> uptake with time, followed by (2) a decrease in the rate until an approximately constant value of CO<sub>2</sub> uptake is attained.

The third one is the ash of oil shale combustion. Research on the carbonation of oil shale ash is being conducted in Estonia [32], [33]. Oil shale ash is not produced in Korea at all so it cannot be the candidate material for carbonation in Korea, but approaches and test methods conducted on the oil shale ash could be referred.

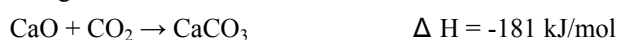
A simple reaction mechanism for CO<sub>2</sub> mineral sequestration by fly-ash in two successive steps: the irreversible hydration of calcium oxide or lime,



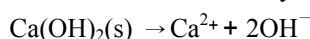
and the spontaneous carbonation of calcium hydroxide suspension.



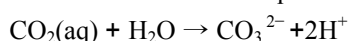
The global reaction becomes



It was assumed that this global reaction takes place at high pH because the dispersion of the fly-ash in pure water at atmospheric conditions for liquid to solid rates (10 to 20) increase rapidly the solution pH up to about 12.5. The aqueous carbonation of Ca(OH)<sub>2</sub> is an exothermic process that concerns simultaneously the dissolution of Ca(OH)<sub>2</sub>,



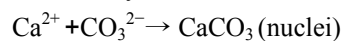
and the dissociation of aqueous CO<sub>2</sub>.



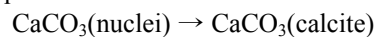
These processes produce a fast super-saturation of solution with respect to calcite,

$$S_1 = (\text{Ca}^{2+})(\text{CO}_3^{2-})/K_{sp} > 1$$

where (Ca<sup>2+</sup>) and (CO<sub>3</sub><sup>2-</sup>) are the activities of calcium and carbonate ions in the solution, respectively, and K<sub>sp</sub> is the thermodynamic solubility product of calcite. Then, the nucleation stage (formation of nuclei or critical cluster) takes place in the system,

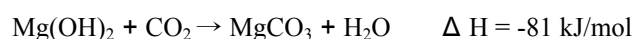
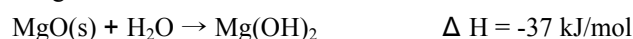


Finally, the crystal growth occurs spontaneously until the equilibrium calcite and the solution is reached,



Theoretical amount of CO<sub>2</sub> (44 g/mole) capture with lime (CaO, 56.08 g/mole) can be calculated based upon the assumption that one mole of CaO reacts with one mole of CO<sub>2</sub> (0.785 g CO<sub>2</sub>: 1g CaO). Montes-Hernandez et al. [18] demonstrated in the experiment that 1 ton of fly-ash, an industrial waste that contains about 4.1 wt% of lime (CaO) with the theoretical amount of CO<sub>2</sub> capture of 32.2 g, could sequester up to 26 kg of CO<sub>2</sub>. In other words, it needs 38.18 ton of fly-ash for one ton of CO<sub>2</sub> sequestered. This result was obtained at a low temperature (30°C) and decent mild CO<sub>2</sub> pressure (10–40 bar). It was about 81% of the CO<sub>2</sub> capture efficiency even after the 18 hours of reaction. They showed that the carbonation efficiency was not significantly affected by reaction temperature and fly ash dose.

In case of MgO (MW 40.3 g/mol), 1 g of MgO reacts with 1.091 g CO<sub>2</sub>.



The overall reaction becomes



Na<sub>2</sub>O and K<sub>2</sub>O will form Na<sub>2</sub>CO<sub>3</sub> and K<sub>2</sub>CO<sub>3</sub>.

### III. ESTIMATION OF CAPTURE CAPACITY AND POTENTIAL

In this paper, the specific CO<sub>2</sub> sequestration capacities per kg fly ash were estimated with two methods. CO<sub>2</sub> sequestration capacity could be related to elemental composition using the Steinour [16] [34] formula in the following equation, which calculates the maximum theoretical amount of reaction based upon the amount of calcium, potassium, sodium and sulfur present in the material.

$$\text{CO}_2(\%) = 0.785\text{CaO} + 1.091 \text{MgO} + 1.41\text{Na}_2\text{O} + 0.935\text{K}_2\text{O} - 0.55\text{SO}_3$$

where CO<sub>2</sub>, CaO, MgO, Na<sub>2</sub>O, K<sub>2</sub>O, SO<sub>3</sub> are in weight % in the ash. From the Steinour's formula and composition data in Table 1, the CO<sub>2</sub> capture capacity could be estimated in Table 2.

The second estimation method was based on the acid neutralization capacity (ANC). Based on the required amount of acid to reach the specific pH value, the amount of CO<sub>2</sub> consumption could be calculated. However, the number of protons released as CO<sub>2</sub> dissolves differs over the pH range. Furthermore, no carbonate minerals are actually formed and precipitated during acid neutralization capacity measurement. The values calculated based on the acid

neutralization capacity was used to derive the minimum value of the capacity. The acid neutralization capacity values were obtained from titration. At high pH values, 1 mol eq H<sup>+</sup> corresponds with 0.5 mol CO<sub>2</sub>, because 2H<sup>+</sup> are created as CO<sub>3</sub><sup>2-</sup> is formed. At medium pH values, HCO<sub>3</sub><sup>-</sup> is formed and only 1 mol H<sup>+</sup> is created for every CO<sub>2</sub> dissolved. As a consequence, it is difficult to translate the acid neutralizing capacity given in mol eq H<sup>+</sup> to mol CO<sub>2</sub>. As a conservative starting point, CO<sub>2</sub> capture capacities were conservatively estimated that 2 mol eq H<sup>+</sup> = 1 mol CO<sub>2</sub>. The capture potential values estimated from the acid neutralization capacity values are listed in Table 3.

#### IV. CO<sub>2</sub> CAPTURE CAPACITY IN KOREA

The amount of domestic production of coal ash was 5.8 million tons/year which was estimated from the hard coal consumption for electricity generation (ash production=10% of hard coal consumption, [35]) and MSWI ash production was estimated to be about 400 thousands ton/year [36]. Even though the total amount of CO<sub>2</sub> captured (285,000 ~ 653,000 tons/year in Table 4) with coal ash or MSWI ash is small compared to the total emission, it is worth to considering the usage of combustion by-product to capture the CO<sub>2</sub>. Since the ash production points and CO<sub>2</sub> emission points are at the same location, the transportation cost is nothing, the carbonation products are more stable than the original waste materials in the environment, and those product have more economical values. Further research on the process in lab-and pilot-scale, derivation of design factors and the economic evaluation can provide competitive technologies in this field.

#### V. COMMERCIALIZATION AND COST

At the present time, note of the processes have been designed in the commercial scale, yet. For instance, Fujii, et al. [37] proposed commercialization of artificial carbonation of concrete with addition of acetic acid as CO<sub>2</sub> capture process which Kakizawa, et al. [38] proposed. With the assumption of complete recycling of acetic acid, cost estimated that the electricity portion of CO<sub>2</sub> capture in the 100 MW oil power

TABLE 1. COMPOSITIONS OF COAL AND MSWI ASH (WT%)

Component	Coal Ash	MSWI Ash
SiO <sub>2</sub>	62.9	49.3
Al <sub>2</sub> O <sub>3</sub>	16.9	7.5
Fe <sub>2</sub> O <sub>3</sub>	5.71	7.6
CaO	2.97	16.3
MgO	1.62	2.6
Na <sub>2</sub> O	0.23	6.0
K <sub>2</sub> O	0.20	1.1
MnO	0.15	0.1
Others	8.64	9.5

TABLE 2. CO<sub>2</sub> CAPTURE POTENTIALS FROM THE MODIFIED STEINOUR EQUATION

	Potential (gCO <sub>2</sub> /kg)
Coal Ash	45.6
MSWI Ash	251

TABLE 3. CO<sub>2</sub> CAPTURE POTENTIALS WITH ANC VALUES

	ANC (mol eq H <sup>+</sup> / kg)	Potential (gCO <sub>2</sub> /kg)
Coal Ash	4.4	96.6
MSWI Ash	2.5	54.9

TABLE 4. TOTAL PRODUCTION AND CO<sub>2</sub> CAPTURE CAPACITIES OF ASHES IN KOREA

	Production (ton/year)	Minimum CO <sub>2</sub> Capture Capacity (ton/year)	Maximum CO <sub>2</sub> Capture Capacity (ton/year)
Coal ash	5,800,000	264,480	560,280
MSWI ash	400,000	21,960	100,400
Total	6,200,000	286,440	663,680

plant with 40% efficiency would be 13.9 MW. Considering CO<sub>2</sub> capture, the efficiency becomes 34.4%. In this case, CO<sub>2</sub> capture would cost 35 €/ton-CO<sub>2</sub> which is lower than that of wollastonite (57 €/ton-CO<sub>2</sub>). In case of using water in place of acetic acid to cement in the process, energy cost of 100 MW plant would be 25.9 MW and monetary wise 22.6 €/ton-CO<sub>2</sub> [39]. Low cost alternatives will be low temperature and low pressure processes. In this case, cost would be 8 €/ton-CO<sub>2</sub> [21], but CO<sub>2</sub> capture efficiency will be decreased significantly.

The final products of carbonation of mine waste or industrial waste are calcium carbonate or magnesium carbonate. The analysis of the added value to the final product is very important. The carbonated material can be disposed, used for mine reclamation, or re-used as raw materials for other industrial purposes. A simple analysis of calcium carbonate in Korean market is conducted as an example.

In Korea, the size of calcium carbonate market is about 100,000,000 tons per year and about 10,000,000 tons of high purity hard calcium carbonate is needed in Korea. Theoretically, since one ton of CO<sub>2</sub> is fixed into 2.3 tons of calcium carbonate, a minimum of 655,000 to 1,500,000 tons of calcium carbonate can be produced. Assuming that the price of calcium carbonate is about 100 dollars, the production of calcium carbonate through fly ash carbonation can be worth between 65.5 million and 150 million dollars. When CO<sub>2</sub> sequestration cost can be estimated to about 44 dollar for a ton of calcium carbonate and raw material cost of fly ash is very low, we can achieve CO<sub>2</sub> sequestration cost about 50 dollars per ton CO<sub>2</sub> which is very comparable to other CO<sub>2</sub> sequestration processes. In addition to the profit from calcium carbonate production, we can obtain other environmental benefits such as industrial waste treatment. Carbonation process can be applied to solidify or stabilize solid combustion byproducts and to manufacture precipitated calcium carbonate and/or magnesium carbonate. Carbonated products can be utilized as aggregates in the concrete industries and as alkaline fillers in the paper making industry.

#### VI. CONCLUSION

The carbonation of pure compounds, such as alkaline and

alkaline earth oxides and hydroxides, is a well known process and the reaction mechanisms and variables involved have all been investigated. However, this paper has shown that heterogeneous materials such as coal and MSWI ash can also be carbonated using both aqueous and gas–solid carbonation processes. The final products of industrial waste carbonation are carbonated minerals such as calcium carbonate ( $\text{CaCO}_3$ ) or magnesium carbonate ( $\text{MgCO}_3$ ). These products have economical value of final products from utilization of carbonated minerals. These materials can be reused as substitute of construction materials and filler of abandoned mine or land-reclamation. Development of carbonation process with these heterogeneous waste materials in economical and industrial scale requires more work since materials involved in the carbonation processes have more complicated structures and diverse chemical compositions.

A preliminary estimation of  $\text{CO}_2$  capture potentials of these waste materials showed that a reduction in carbon dioxide emissions to the atmosphere could also be achieved if the solid wastes from the emission sources are used for the capture of  $\text{CO}_2$  from the stack. The annual amount of  $\text{CO}_2$  capture with these industrial wastes in Korea could be significant. As an environment-friendly and economical method, it can be very feasible to use those industrial wastes as a  $\text{CO}_2$  capturing material. The industrial carbonation technology can contribute to both reduction of the greenhouse gas and conversion of industrial wastes into useful products.

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