CO₂ sequestration of real flue gases from landfill municipal solid waste incineration (MSWI) -Pilot scale demonstration

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Abstract: Municipal solid waste is being produced in large quantities, in keeping with a dramatic population increase and the growth of industrialization. Most of this waste is sent to landfills in South Korea. However, current treatment methods, including incineration and recycling is limited owing to insufficient landfill space and the generation of leachates and soil contamination. This study seeks to determine the degree of influence of several main factors, such as the optimum temperature, solid-towater ratio and CO₂ concentration, on the process of accelerated carbonation of the landfill flue gases (7% concentration) mixed with KR slag. This paper presents the experimental results obtained from a pilot scale demonstration of landfill real flue gases mixed with KR slag. This demonstration was performed with the capacity of 20 tons/hour.

Index Terms: Utilization, municipal solid waste, flue gases,

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I. INTRODUCTION

In general, solid waste arises from lot of human activities such as domestic, agricultural, industrial, commercial, waste water treatment, construction, and mining activities etc. If the waste is not properly handled and treated, it will have a negative impact on the hygienic conditions in urban areas and pollute the air with greenhouse gases (GHG) and surface and ground water, as well as the soil and crops. Currently, wide variety of smart technologies available for MSWI management in developed countries. Recycling is the other major alternative process for MSWI landfill issues.

In 2004, 94% of greenhouse gas emissions come from energy consumption and industrial processes in Korea. To achieve the 5% reduction that was mandated to Korea in 2004, 30 million tons of CO₂ gas emissions must be reduced, and such reduction will cost domestic industry approximately 4 trillion won every year. With the recent highlights of CO₂ reduction and resource recycling, various policies have been delivered to achieve greenhouse gas reduction and improved competitiveness of the national resource industry. According to the results of the UNFCCC, the attempts of each country to secure certified emission reductions and greenhouse gas reduction technology have attracted a great deal of attention due to the enormous economic profit from CDM business. In the case of South Korea as a whole, the waste disposal rate on a per capita basis dropped by 40% from 1995 with the remaining waste treated by incinerators and then landfilled. ^{1,2} The capital city Seoul, with a population of more than 10 million, has built four incinerators for treating MSW with a daily capacity of 2,850 tonnes in total. ³

Current domestic greenhouse gas emissions constitute 1.8% of global emissions and it is highly probable that Korea will be classified as a greenhouse gas reduction mandatory subject for the 2nd commitment period (2013-2017). Therefore, corresponding preparations are urgently required. Developed countries are nurturing green environment industries based on the traditional green technology of 21st century, and they are promoting open innovations, grafting the recycling industry of waste byproducts of the past with novel business paradigms. Regarding technology in response to climate change, the world has set national agendas that take resource recycling as the basis of not only every industry but the social economic system as a whole. Lead the development of an important future industry with high global potential and provide a resource recycling action plan as a model fused with energy.

The International Energy Agency (IEA) has predicted that approximately 22% of carbon dioxide emitted in 2050 can be reduced with CCS technology. Industries with high energy consumption occupy

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the majority in Korea. Therefore, it is predicted that approximately 40% or more of carbon dioxide emissions can be reduced with CCS technology. A demonstration plant that will stably conduct a solid-solution treatment on independently condensed CO₂ from landfill gas, with core technology completed from the research mentioned above, and recycle the resource is required.

Therefore, the construction of a CO₂-utilizing resource treatment and CO₂ fixation demonstration plant by establishing a treatment technology that recycles inorganic wastes through the fixation of CO₂ and CO₂ carbonation for CO₂ greenhouse gas reduction and recycling of inorganic wastes is required. The present objective of our study is to characterize the municipal solid waste incineration bottom ash and utilization of MSWI landfill real flue gases mixed with KR-slag for CO₂ sequestration at pilot scale demonstration

II. MSWI bottom ash (Practical Approach, Pilot plant 20 tons/hour)

The South Korean Government has been promoting the 3R (Reduce, Reuse and Recycle) policy in recent years.⁴(Fig.1).



Figure 1. Waste treatment process in South Korea

- i) By-product that is produced during the combustion of municipal solid waste in solid waste combustor facilities (Fig.1 (a) bottom ash and KR Slag (Fig.1 (c)). (Table-1).
- ii) Category : Bottom ash(Grate ash, siftings), fly ash(boiler ash, precipitator or bag house ash)
- iii) Bottom ash

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- Approximately 75 to 80 percent of the total combined ash stream
- Similar in appearance to a grayish, silty sand with gravel
- Consists primarily of glass, ceramics, ferrous and nonferrous metals, and minerals.



Figure 1(b). Municipal Solid Waste Incineration Ash



Figure 1(c). KR slag

MSWI bottom ash waste treatment process

State-of-the-art plants produce approximately 200 kg great or bottom ashes per 1000 kg of burnt waste. In semi-dry or dry systems the amount of residues is significantly increased because of not reacted additives. Figure 2 shows the flow diagram of bottom ash treatment process.



Figure 2. MSWI bottom ash treatment process flow sheet in KIGAM

Pretreatment Process

In general, the mixture of MSWI bottom ash was stabilized as the following pretreatment processes: water washing, phosphate stabilization and carbonation.⁵ After the optimum processing condition for each pretreatment was determined, the performance of each pretreatment was examined using pH stat leaching tests (pH 6,9, 12) and availability tests (Figure 3). For the performance tests, fly ash alone was used, instead of a mixture of MSWI residues, to observe leaching differences among the pretreatments more distinctly.

In addition, these pretreatment techniques were usually used for the MSW of high calorific value. However, the relatively high water content of waste (74%), a common characteristic of the refuse produced in Asian countries, will lead to low calorific values.⁶



Figure 3. Pretreatment process of MSWI bottom ash

Accelerated carbonation of MSWI bottom ash

Accelerated carbonation has been developed and it has been commercialized CO_2 is artificially injected into the target material, the waste, to accelerate the carbonation reaction and the treatment is not limited by space. Therefore, time and space are saved compared to natural carbonation⁷⁻¹¹. In addition, the main culprit of the greenhouse effect, CO_2 , is produced in many areas. Using CO_2 to accelerated carbonation is a significantly effective method from the perspective of CCS.

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Major factors in accelerated carbonation include temperature, water content, CO₂ density, and pressure, and these factors are closely related to reaction rate. The reaction rate increases with increasing temperature. However, high temperature has a negative effect on high CO₂ content. The actual CO₂ employment obtained from the process demonstration in pilot plant scale using the municipal waste was 26.0kg-CO₂/ton-waste, and 35.0kg-CO₂/ton-waste less than 4.75mm. Considering the domestic municipal solid waste incineration bottom ash production, CO₂ emission reduction of 16,500ton/year is expected.

The highest carbonation rate was observed in 4.75mm or smaller bottom ash at 20° C, solidliquid ratio of 0.3, and 30% of CO₂, proving the efficiency of the utilization of incinerator exhaust gas. The phase-boundary control reaction was dominant in the initial rate-determining step of dry carbonation. However, it was later revealed that the product layer diffusion control reaction becomes the rate-determining step as the reaction proceeds. The result also showed effective reduction of Pb and Cu, which exceed the elution standard, among the heavy metals of domestic municipal solid waste incineration bottom ash. The heavy metal stabilization of municipal solid waste incineration bottom ash seems to be comprehensively influenced by the generation of stable compounds, such as carbonate, due to the decrease in pH, physical blocking with capsulation, and the generation of absorbents from the decomposition of hydrates.

MSWI bottom ash landfill flue gases with KR slag

After carbonation, the employment of CO 31.38 wt% of KR slag was obtained. Treatment of a ton of KR slag is predicted to employ approximately 457.3kg of carbon dioxide. Construct and operate a recycle treatment demonstration plant capable of treating 50,000ton/year of inorganic wastes, such as municipal solid waste incineration bottom ash, and capture 3,000 ton/year of CO₂(Figure 4). This research aims to develop and demonstrate an environmentally friendly waste treatment process by integrating CCS and resource recycling technology. The technology fixes greenhouse gas (CO₂) in various inorganic wastes, such as incineration ash and others, which are traditionally buried, to manufacture safe cyclic aggregates by preventing the elution of harmful heavy metals. The current demonstration plant mainly treats municipal solid waste incineration bottom ash and KR slag produced during the steel-making process.

The current demonstration plant mainly treats municipal solid waste incineration bottom ash and KR slag produced during the steel-making process. The imported wastes go through impurities removal, primary size sorting, iron separation, and other pretreatment processes. The wastes are

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finally categorized into cyclic aggregates for manufacturing processes and CCS processes through secondary size sorting. The fine powder wastes under a 100mesh (0.15mm) are made into a slurry of an appropriate solid-liquid ratio in a mix-tank, and combined with exhaust gas (CO_2) in a loop-reactor to collect CO₂ in large amounts, and then discharged after the dehydration process.

Particles larger than a 100mesh (0.15mm) go through dry carbonation, dechlorination, and other stabilization processes that generate carbonate layers on the particle surfaces in an extractor. The particoles are then recycled and stored as cyclic aggregates. Such pretreatment, cyclic aggregates production, CCS, and other processes of the demonstration plant are composed as an automated central system controlled by a small number of people. The system operation was stabilized and the plant processed 15 ~ 20 tons of waste every hour. In CO₂ capturing, bottom ash powders, KR slag, and combination of the bottom ash and KR slag in a 10:1 ratio yielded stable carbonation rates of approximately 5%, 31% and 19% respectively in CO₂ capture.



Figure 4. Carbonation process of MSWI bottom ash

	Al ₂ O ₃	CaO	Fe ₂ O ₃	K₂O	MgO	MnO	Na₂O	P ₂ O ₅	SiO ₂	SO₃	L.O.I
Ash	15.66	39.36	0.88	0.20	1.72	0.14	0.68	1.86	6.28	1.19	29.8
(Wt%)											

Table 1. XRF analysis of MSWI bottom ash samples

Effect of CO₂ pressure before and after carbonation

 CO_2 pressure change $(\Delta P - \Delta P_{blank})$ in the case of using CO_2 in 10 and 20 bar ranged from 0.296 to 0.300 bar. Interestingly, the pressure drop was not shown in the case of using CO_2 in 30 and 40 bar. In the case of using 10 and 20 bar of initial CO_2 pressure, CO_2 pressure drop $(\Delta P - \Delta P_{blank})$ was 0.296 and 0.300 bars. However, in the case of using 30 and 40 bar of CO_2 , the pressure rather increased after the carbonation, indicating no carbonation effect in those conditions.

Sequestrated CO_2 as mol was calculated by the ideal gas equation, and then the amount of sequestrated CO_2 as g/kg was max. 25.85 g/kg. Carbonation efficiency (CE) was also calculated from the ratio of real and theoretical CO_2 sequestration amount, and then the value was max. 7.88% (Figure.5).



Figure 5. Effect of CO₂ pressure before and after carbonation

Conclusions and Outlook

The accelerated carbonation of solid wastes containing alkaline minerals such as Ca and Mg before their landfill treatment is effective for decreasing the mobility of heavy metals by adjusting pH to below 9.5 at which their solubility is lowest. In the worldwide trend towards encouraging carbonation treatment of MSWI bottom ash as described above, our research results shows the carbonation studies using MSWI bottom ash and CO_2 in order to increase the recycling percentage of the ash and reduce the concentration of atmospheric CO_2 in Korea.

We established a pilot plant successfully for multi-processing of MSWI ash in South Korea. The capacity of this pilot plant is 200 kg/hour. This pilot plant was the first one in the waste recycling field in Korea and even throughout the Asian countries, and includes multi-processes of particle separation, heavy metal stabilization, chloride removal, and green aggregate/concrete manufacture. The higher capacity 20 tonns / hour plant (Fig.3), for all inorganic waste treatment by accelerated carbonation and utilized direct flue gases from landfill sites, mixed with KR slag for CCS model in South Korea.

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