Purification of LFG and Utilization of CO₂ by Extracting Calcium Hydroxide from MSWI Fly Ash

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INTRODUCTION

Landfill Gas (LFG) is a mixture generated by anaerobic digestion of organic waste, which contains mainly CO₂ (30-50 vol.%) and CH₄ (40-60 vol.%). The presence of CO₂ in LFG reduces its calorific value, so its remotion produces a high-quality methane gas. Carbon Capture Utilization and Storage (CCUS) is a technology that captures CO₂ from gas emissions for posterior storage and conversion into valuable materials¹. Monoethanolamine (MEA) is commonly used for CCUS due to its high absorption capacity and reaction rate in CO₂ reaction. Mineral carbonation can be applied to regenerate the absorbent and reduce the high energy required in the desorption process. The direct carbonation reaction requires high temperature and pressure, but using previously extracted metal ions allows the reaction to proceed in environments under 373 K at normal pressure. In this study, fly ash generated in Municipal Solid Waste Incineration (MSWI) was selected for the raw material of Indirect carbonation due to its high content of CaO. This research studies the CCUS process for LFG using MEA and posterior regeneration of loaded MEA using extracted Ca from MSWI fly ash.

MATERIALS AND METHODS

Materials

LFG was sampled from the Sudokwon Landfill Site Management Corporation in Seoul, Korea. containing 58 and 42 vol.% of CO₂ and methane respectively. MEA was used as CO₂ absorbent, hydrochloric acid and nitric acid was used to extract calcium from the MSWI fly ash, and Sodium Hydroxide was used to increase pH. MSWI Fly ash was sampled form the "N" Resource Recovery Facility in northern Seoul, Korea. Distilled water was used in all experiments.

Methods

The absorption experiment is conducted in a reactor at 298.15 K, and a gas analyzer to monitor CO_2 and CH_4 concentrations, while keeping the output line set to 268.15 K. The absorption experiment is conducted until the output concentration matches the inlet concentration of 42%. Calcium ions needed for MEA regeneration were extracted using a pH swing method. First, calcium ion is leached from MSWI fly ash using acid by lowering the pH to 3. Then, impurities are precipitated from the calcium-enriched solution by increasing the pH to 11. Lastly, the calcium ions are extracted from the leaching solution by raising the pH again to 12.7. The extracted calcium was used for regeneration of saturated MEA, mixing both for 36h and stirring at 500 rpm. The precipitate is recovered, dried and analyzed with XRD both before and after the absorbent regeneration.

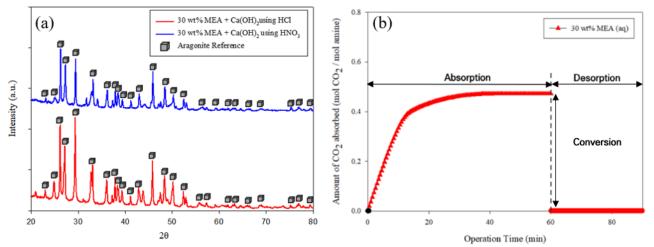
RESULTS AND DISCUSSION

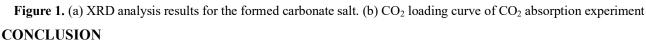
Results of overall calcium extraction process using acids and base from MSWI Fly Ash

Table 1 shows the main results of pH swing method for calcium extraction. The final amount of Calcium recovered was 502.83 g/Kg and 83.20 g/Kg for hydrochloric acid and nitric acid, respectively. The carbonate salts formed are identified as aragonite, as shown in Figure 1.(a). Figure 1.(b)shows the results of CO₂ absorption and desorption experiments. CO₂ in LFG was completely removed using MEA in the absorption experiment, while the calcium recovered from MSWI fly ash was used to regenerate saturated MEA. It is noticeable that the regeneration occurs almost instantly, unlike conventional thermal regeneration. **Table 1.** ICP-OES analysis results for relevant supernatants and precipitate of calcium extraction processes

Component -	Calcium Leaching Phase		Cation Impurity Removal Phase		Calcium Recovery Phase	
	(supernatant) [g/L]		(supernatant) [g/L]		(precipitate) [g/Kg]	
	Hydrochloric	Nitric	Hydrochloric	Nitric	Hydrochloric	Nitric
	Acid	Acid	Acid	Acid	Acid	Acid
Ca	10.94	11.94	0.99	8.90	502.83	83.20
Mg	1.51	1.25	<lod**< td=""><td><lod**< td=""><td>< LOD**</td><td><lod**< td=""></lod**<></td></lod**<></td></lod**<>	<lod**< td=""><td>< LOD**</td><td><lod**< td=""></lod**<></td></lod**<>	< LOD**	<lod**< td=""></lod**<>
Impurities*	44.22	43.93	54.69	43.97	4.31	3.77

*Impurities: Zn, Fe, Ti, Pb, Si, Na, Cu, K, Al, Sn, Cr, S ** LOD: Limit of Detection





This study aims to obtain high purity CH_4 by absorbing CO_2 in LFG and to extract calcium from MSWI fly ash for its regeneration. The results of this study show that calcium hydroxide was obtained in concentrations high enough to be used as feedstock in the regeneration of CO_2 loaded MEA. Adding an impurity removal step greatly improves the efficiency of this process. Using industrial byproducts to generate value stablishes this process as an eco-friendly way to close the carbon cycle.

ACKNOWLEDGEMENT

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