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# Self-sustaining carbon capture and mineralization via electrolytic carbonation of coal fly ash

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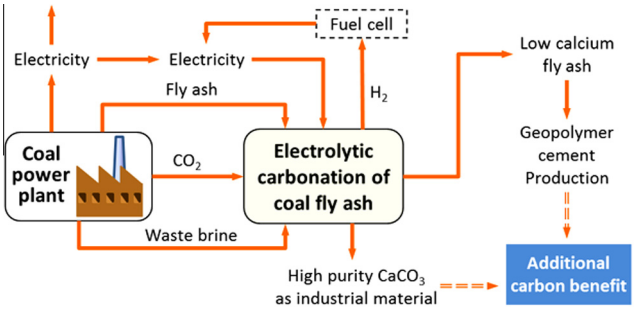
## HIGHLIGHTS

- Synergistic treatment of different waste streams in coal-fired power plant.
- Self-sustaining carbon sequestration by electrolytic carbonation of coal fly ash.
- High purity $CaCO_3$ production and promotion of fly ash as renewable material.
- Low energy, cost and environmental affect for carbon mitigation.

## GRAPHICAL ABSTRACT

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## ABSTRACT

This study presents a new electrolytic carbonation process to synergize the treatment of different waste streams generated by power plants, including fly ash, brine wastewater, and $CO_2$. The acidity generated by electrolysis of brine electrolyte directly liberates calcium from fly ash. The metal ions balance the $OH^-$ produced at the cathode to form hydroxide, which then fixes $CO_2$ into high purity carbonate precipitates. Results show that electrolysis increased fly ash dissolution by 32.4% compared to the control with spontaneous dissolution of fly ash, and the carbonation process captured 89% more $CO_2$ and increased capture capacity from 9.75 to 18.42 kg-$CO_2$/t-fly ash in the NaCl electrolyte. The energy expenditure was 19.4–29.3 kJ/mol-$CO_2$, lower than that required for sorbent or solvent based post-combustion carbon capture. The process takes advantage of all waste streams generated on site and can consolidate traditionally separated treatment processes to save costs, produce energy and value-added products, as well as generate carbon benefits.

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## 1. Introduction

Conventional coal-fired power plants generate multiple waste streams including solid waste, wastewater, and flue gas. In 2011, the coal plants in the United States generated 110 million tons of coal ash, emitted nearly 1.6 billion tons of $CO_2$, and consumed nearly 1 trillion gallons of water for processing [1–3]. To minimize waste discharge and the associated environmental impacts, separated treatment technologies for solid, liquid, and gaseous wastes have been applied, but treatment costs are high, especially when a new process such as $CO_2$ emission control is applied.

In an effort to synergize the treatment processes and reduce cost, mineral carbonation (sequestering $CO_2$ as a stable Ca/Mg carbonate) of alkaline residues such as coal fly ash has been used for post-combustion $CO_2$ capture and sequestration. This method carries the potential to reduce the cost for both fly ash reuse and on

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site carbon capture, and it avoids long distance $CO_2$ transportation, deep well injection, and risks of leakage and spills [4]. Direct $CO_2$ reaction with dry, semidry, or aqueous alkaline residues can't generate reusable products [5–7], rather the products are generally landfilled. In comparison, indirect extraction-carbonation is more beneficial as it extracts alkaline earth metals (mainly Ca and Mg) using chemical reagents to purify fly ash and carbonize the cations using injected $CO_2$ [8–10]. The resulting products are value-added high purity carbonates that can be used in various applications. However, the extracting media are usually ammonium salt [9,10] or weak acid [8], which are hazardous and non-sustainable. Water has also been used for metal extraction but with low efficiency, because only a small fraction of active ingredients can spontaneously dissolve in neutral water.

In this study, we present a new method called electrolytic carbonation to synergize and increase the performance of multi-stream waste management for fly ash carbonation, $CO_2$ capture and mineralization, as well as brine wastewater treatment. Because no electrochemical method for indirect carbonation of coal fly ash has been reported, we hypothesize that the acidity generated by electrolysis of brine wastewater could directly dissolve fly ash in the anode and liberate metal ions. The metal ions balance the $OH^-$ produced at the cathode to form metal hydroxide, which then fixes $CO_2$ into carbonate precipitates (Fig. 1). Recent studies support the theory and showed that electrolysis could extract metal ions from wollastonite [11,12], but no study investigated the feasibility of electrolytic dissolution of coal fly ash for $CO_2$ sequestration or the synergistic treatment of different waste streams in coal-fired power plant. In addition to minerals from fly ash, high concentrations of $Ca^{2+}$ and $Mg^{2+}$ already present in brine wastewater could also be removed through carbonation. The $H_2$ produced during electrolysis can offset energy consumption and potentially make the process energy neutral or positive. The advantage of this integrated process is that all the materials used such as sorbent (fly ash), electrolyte (brine wastewater), and $CO_2$, are waste materials generated on site that are required to be treated anyway. The resulting $CaCO_3$ from $CO_2$ is a common material for industries, and purified fly ash without calcium has been used to produce geopolymer cement to replace traditional Portland cement, which further reduces carbon emission associated in the cement industry [13] (Graphical abstract). In this study, we showed that electrolysis increased metal liberation from fly ash, which resulted in much improved $CO_2$ capture and mineralization compared to spontaneous dissolution of fly ash in water for $CO_2$ sequestration. The $CO_2$ mitigation and energy balance were quantified, showing great benefits for the power industry.

## 2. Materials and methods

### 2.1. Fly ash

Coal fly ash was obtained from Valmont Power Station in Boulder, Colorado, USA. The fly ash sample has a particle size 0.6–98.1 μm with a median size of 35.3 μm. The sample is composed of $SiO_2$ (46.76 wt%), $Al_2O_3$ (22.39 wt%), CaO (12.22 wt%), $SO_3$ (6.91 wt%), $Fe_2O_3$ (2.74 wt%), $K_2O$ (1.12 wt%), $Na_2O$ (1.10 wt%), MgO (1.13 wt%), Pb (0.005 wt%) and U (0.001 wt%).

### 2.2. Electrolytic cell construction

Electrolysis was carried out in a plastic electrolytic cell with 350 mL electrolyte (Fig. 1). Two graphite rods (1 cm diameter × 10 cm length) were placed 4 cm apart to serve as the anode and cathode, respectively. In some cases, the anode was coated with a layer Nafion film (5 mg/cm$^2$) to form oxygen selective electrode [14] for the NaCl electrolyte, while no coating was applied for the $Na_2SO_4$ electrolyte. The anode was vertically placed in the center of a porous tube container (50 mL plastic centrifuge tube) that contained fly ash slurry. The inside wall of the tube container was lined with a cellulose extraction thimble (2.5 cm diameter × 10 cm length, Whatman, UK), so fly ash slurry could be added into the tube without leaching to the main container. The electrolytic cell was covered to prevent air exposure. An external voltage of 3.5 V was applied between the electrodes across a 1 ohm resistor. The electrolyte was stirred by a magnetic stirrer at 120 rpm.

### 2.3. Experimental procedure

Two electrolytes were used (28 g/L NaCl and 35 g/L $Na_2SO_4$) to simulate the waste brine generated after boiler feed pretreatment in power plants. The fly ash slurry was firstly prepared

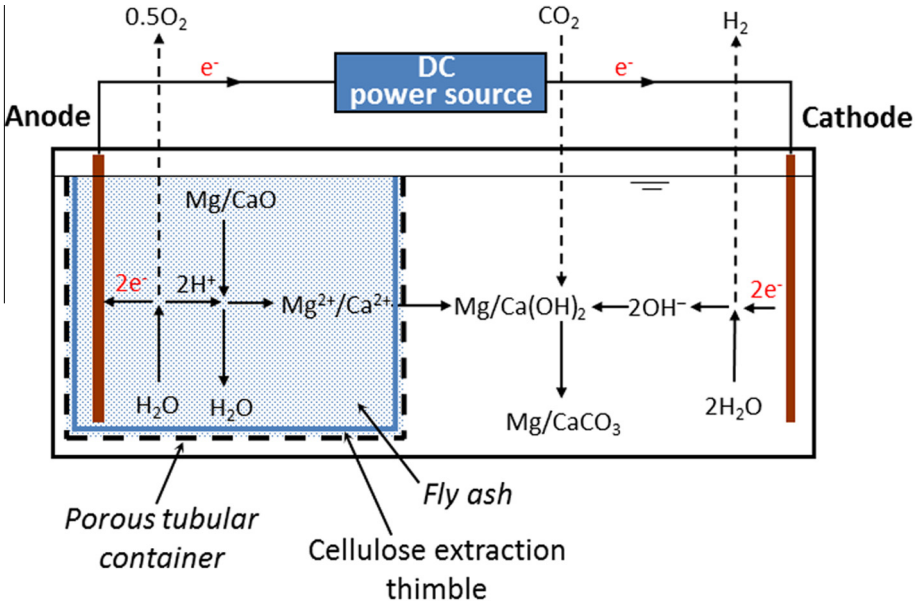


**Fig. 1.** Schematic of the electrolytic carbonation of coal fly ash for *in situ* carbon capture and mineral recovery.

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