



## An electrochemical membrane reactor for a recycled FGD process



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### HIGHLIGHTS

- NaOH sorbent was regenerated electrochemically from FGD residuals.
- Sulfur was recovered as H<sub>2</sub>SO<sub>4</sub> and H<sub>2</sub> was produced as a clean energy.
- The current efficiency could be as high as nearly 90% under the optimal conditions.
- The cost of the energy consumed to recover sulfur was estimated at 0.81 \$/kg S.

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### ABSTRACT

In the current process of removing sulfur dioxide, the high cost and treatment of fuel-gas desulfurization products are still the two bottlenecks that need to be resolved. This paper presents a novel method that can regenerate the adsorbent and obtain resources and energy recovery from FGD (Flue gas desulfurization) products by using an electrochemical membrane reactor. The effects of the operating parameters, such as the current, the initial concentration of the three chambers, the flow rate, and the power supply mode on the regeneration and current efficiency were analyzed, and the optimal operating condition of the lab-scale electrochemical membrane reactor was obtained. Under this optimal condition, the current efficiency of H<sub>2</sub>SO<sub>4</sub> production and NaOH regeneration were about 85% or above and as high as 90% correspondingly. In addition, the process cost from the consumption of energy is estimated to be 0.81 \$/kg sulfur; therefore, the profit from the electrolytic products is higher than the price of the electricity. Notably, the method is appealing both because it can expediently regenerate the adsorbent with no secondary pollution and because it is economically attractive.

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### 1. Introduction

Sulfur oxides, emitted from both natural and anthropogenic sources, are primarily formed during the combustion of sulfur-containing fuels, including coal, and the production of sulfuric acid and sulfates [1]. So far, coal continues to be the fuel of choice for power generation throughout much of the world. Then, in the process of thermal power generating, it is commonly known that SO<sub>2</sub> is emitted in the flue gas, which becomes a major problem of air pollution because of that SO<sub>2</sub> emitted has adverse effects on human health, and its atmospheric products can affect the environment on the local, regional, and global scales. Therefore, to achieve sustainable development, the control of the emission of sulfur dioxide at its sources is of growing importance.

Currently, there is a global focus on fuel gas decontamination with emphasis on diversification, integration, and greening; there-

fore, the control techniques that incorporate desulfurization or denitrification are being researched by governments and academic institutes worldwide [2,3]. The critical factors for desulfurization techniques are the desulfurization chemical used and the profitability of the byproducts. Different processes are used, including wet processes (WFGD) that use limestone–gypsum, ammonia liquor and other basic chemicals as the absorbent [4–6], semidry processes (SDFGD) such as spray dry scrubbing, dust sorbent injection, and circulating fluidized beds [7–9], dry processes (DFGD) such as furnace sorbent injection or the use of various absorbers [10,11]. Although WFGD have earned widespread use due to their high desulfurization efficiency and reliability, the byproducts, especially calcium sulfate, have low puerility and reuse ability, restricting the popularization of this techniques. SDFGD is confined by its low efficiency of desulfurization compared to WFGD and its poor operating stability. Some researchers working on investigating novel materials or processes for use in FGD found some absorbents with high removable efficiencies and presented new processes which were able to recycle the absorbent [12–14]. In

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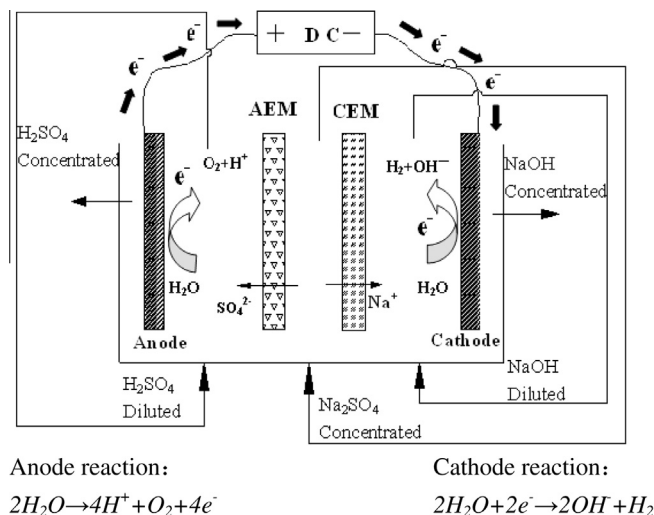


Fig. 1. The schematic of the membrane electrolysis process.

other studies, electrochemical methods have been praised highly for their few or no secondary pollution and the profitability of the FGD byproducts or residuals [15–17].

With respect to electrochemical processes, many scholars have researched various combinations of FDG technology with membrane electrolysis [14–23], which showed good prospects for the application. But most electrochemical processes presented are based on the improvement of wet flue gas desulfurization and some of them need relatively complicated configuration. In this study, a relatively simple process was presented, and the investment cost is low with less equipment required and the use of a traditional ionic membrane for recovery, which is adequate and stable. In addition, the shortcoming in desulfurization techniques, such as the disposal of huge amounts of FGD residuals and low efficiency in SDFGD, can be overcome by the combination of semidry FGD and membrane electrolysis. In the combined process, the  $SO_2$  is efficiently absorbed by the sodium alkali absorbent, which would be regenerated by membrane electrolysis. In an electrochemical membrane reactor, the desulfurization product which is mainly composed of  $Na_2SO_4$  can be highly diluted, and NaOH and  $H_2SO_4$  can be formed by utilizing the  $H^+$  and  $OH^-$  produced by water splitting. In this way, the process can not only regenerate the absorbent and maintain a steady and stable circulation but also produce hydrogen as a clean-energy, which can be used as a denitrification agent.

The primary purposes of this paper are to investigate the relevant parameters in the electrolyzing process and to seek the optimal operating conditions under which the absorbent NaOH is maximally recovered via membrane electrolysis from the desulfurization product. This paper explores how to increase current efficiency using the electrode process dynamics equation. This equation can be used to determine the preferable recovery rate and verify the efficiency, environmental benefits, and economic feasibility of this method by calculating its energy consumption during the process.

## 2. Materials and methods

### 2.1. Electrolysis equipment and procedure

A lab-scale electrolysis setup was used in a three-compartment configuration as shown in Fig. 1. A pair of membranes, a power supply (max. 30 V, 30 A), three peristaltic pumps (max. 24.84 L/h), and three solution tanks collecting either sulfuric acid, sodium hydroxide, or the electrolyte of sodium sulfate, were employed in the electrolysis unit, which was made of ceramic. The electrolysis unit was separated into the cathode, desalting and anode compartments by alternating the cation exchange membrane (JCM-II Wuxi Jianyi Co., China) and the anion exchange membrane (JAM-II Wuxi Jianyi Co., China). The cathode and anode compartments had the same volume (outer border:  $32 \times 20 \times 3$  cm; inside casing:  $24 \times 12 \times 1.9$  cm), whereas the middle compartment was relatively small (outer border:  $32 \times 20 \times 1.5$  cm; inside casing:  $24 \times 12 \times 1.5$  cm). Each compartment had a thickness of less than 0.1 cm gasket. The cathode, made of titanium, and the anode, made of nickel, were 1.5 cm apart and were connected to a direct current power supply through conducting wires. In this way, the overall geometrical surface area of the membrane was  $338 \text{ cm}^2$ . The properties of the membranes used in the membrane electrolysis cell were listed in Table 1.

All reagents were purchased from the Shanghai Chemical Reagent Co., (China) and were analytical grade. The practical desulfurization residuals were obtained from a Shandong Zaozhuang power plant using NaOH as the scrubbing agent in the process of semidry FGD. The analysis of the residuals shows that it contained 82%  $Na_2SO_4$  and 11%  $Na_2SO_3$  by weight. The other components of the residuals were composed by the oxides of Si, Fe, and Ca. Before the FGD residuals were introduced into the electro-reactor as aqueous solution, most of calcium and magnesium including some soluble impurities could be removed by chemical precipitation with NaOH in the primary brine treatment system. In this study, for simplicity, the simple test solution of  $Na_2SO_4$  was used as artificial solution of FGD residuals in the lab-scale experiments. Different weight percent of acid ( $H_2SO_4$ : 4–8%), base (NaOH: 4–8%) and electrolyte (5–20%) were pumped into the anode chamber, cathode chamber, and desalting compartment, respectively, under constant temperature ( $T = 25 \text{ }^\circ\text{C}$ ). Before the three operation units could be carried out simultaneously, some solutions were prepared in preliminary experiments to initiate the process cycle. The independent solutions were circulated for 30 min, and all the visible gas bubbles in every compartment were eliminated [24]. The membrane electrolysis process consisted in applying a constant current (1.5 A–3.0 A). Samples of the solutions (10 ml) were collected every 10 min or 30 min from 0 to 120 min or 210 min in the three compartments for mineral analysis.

### 2.2. Limiting current density measurement

The test solution was pumped into the electrolyzer while the concentration of the added solutions and the flow range were maintained to be the same as the electrolysis operation mentioned above. When the flow rate and the inlet and outlet pressures were stable, the power supply was turned on and the data were re-

Table 1  
The properties of the membranes used in the ED cell.

Membrane	Thickness ( $\mu\text{m}$ )	IEC (mg/g)	Area resistance ( $\Omega \text{ cm}^2$ )	Efficiency (%)	Water content (%)
JCM-II	0.20	1.8–2.0	3–6	95–99	33–38
JAM-II	0.20	1.6–1.8	6–10	90–95	22–24

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