



Mineral carbonation of Victorian brown coal fly ash using regenerative ammonium chloride – Process simulation and techno-economic analysis



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HIGHLIGHTS

- A detailed techno-economic analysis of four conceptual processes was implemented.
- Mineral carbonation could convert a zero-value waste into value-added products.
- The leaching waste from mineral carbonation could be served as additive to cement.
- The production cost for different scenarios range ~\$61–333 per tonne of products.
- NPV, IRR and payback period for the best scenario could be \$49 M, 53.4% and 2.3 years.

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ABSTRACT

This report examined the technical and economic feasibility of four process scenarios for the mineral carbonation of Victorian brown coal fly ash. The first two design scenarios aimed to compare the performance of two leaching agents, namely, ammonium chloride (NH_4Cl) and a mixture of ammonium chloride and hydrochloric acid ($\text{NH}_4\text{Cl} + \text{HCl}$), on product yields and cost, whereas the other two scenarios were designed to recycle the leaching residue via single or multi-stage leaching steps to co-produce a carbonate precipitate and cement additive-grade by-product. Detailed designs were developed in Aspen Plus to determine the technical and economic potential of the selected process configurations and identify the concept with the lowest overall costs relative to the product yields. As has been confirmed, the overall production costs and carbon dioxide (CO_2) capture cost of the evaluated process scenarios range from ~\$61 to 333 per tonne of product and from \$135 to 1091 per tonne of CO_2 , respectively. The process scenario that used $\text{NH}_4\text{Cl} + \text{HCl}$ as the leaching reagent had a significantly larger cost and a higher carbonation conversion compared to the other scenarios. The process configuration that recycled the leaching residue resulted in the lowest cost per tonne of fly ash and the lowest CO_2 capture cost among the four proposed scenarios. The largest net present value (NPV) and the internal rate of return (IRR) as well as the shortest payback period for this scenario further confirmed its highest profitability. The NPV, IRR and payback period of \$49 million, 53.4% and 2.3 years, respectively, could be achieved using Victorian brown coal fly ash in this scenario. A sensitivity analysis suggests that the change in the ammonium chloride price exerts the largest effect on the production cost. A 50% increase in the ammonium chloride cost could result in the production cost increasing by 29.5%. Additionally, the selling price of the carbon precipitate product and the production cost strongly affect the financial indices. However, the production of the cement-additive by-product exerts a marginal role on the process profit. The extra income created from the cement-additive by-product is counteracted by the larger cost related to the purchase and consumption of hydrochloric acid used in the final leaching stage.

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1. Introduction

Fossil fuel combustion was responsible for approximately 31 Gt of CO_2 emissions into the atmosphere in 2011, and this amount is expected to increase to 57 Gt in 2050 [1]. Due to the significant concerns for global warming, carbon dioxide capture and storage

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(CCS) is considered to be one of the potential options for addressing the increase of CO₂ concentration in the atmosphere and climate change mitigation [2–5]. Although CO₂ geological storage is the only option that has been practically used in the industry [2], storing CO₂ as a mineral carbonate has received increased attention due to its numerous advantages, particularly its small environmental load in terms of product disposal [6,7]. Furthermore, the resultant carbonate can be used in a value-added way. The growing interest in the development of mineral carbonation methods still requires intensive investigation to reduce its cost and demonstrate potential applications for these technologies in existing power plants [8].

In mineral carbonation, carbon dioxide is fixed with divalent metal oxides, such as magnesium and calcium, from natural minerals or industrial wastes to produce stable carbonate as an end product [2,3]. Despite the intrinsic advantages of mineral carbonation, it still suffers from two major drawbacks: prohibitive energy consumption and poor economic aspects [9]. Efforts to accelerate the kinetic reaction and reduce the required activation energies have motivated the development of an indirect mineral carbonation process using two separate steps for mineral leaching and CO₂ capture [9,10]. Indirect mineral carbonation involves the extraction of the reactive minerals prior to carbonation [11]. This method is appealing due to the possibility of occurring under mild operating conditions (*i.e.*, temperature <100 °C and pressure up to 5 bar) and producing high-purity carbonate with a high market value [12]. To ensure that this process is economically and environmentally sustainable, the chemicals used in the process need to be regenerated within the process itself [13]. The leaching media for the dissolution of calcium and magnesium in the minerals are either strong or weak acidic reagents, ammonium salts, or alkaline solutions [14–16]. Among them, the ammonium extraction process appears to be a favorable technique, given its potential for reagent recovery and the relatively large selectivity of magnesium and calcium over other elements [16,17].

Coal-fired power plants, which provide ~40% of the world's electricity and annually generate 12,000 Mt of CO₂ and 600 Mt of fly ash (FA) [18], could benefit from this option by using its own byproducts, *i.e.*, fly ash for CO₂ sequestration. Generally, approximately 30% of the fly ash produced in coal power plants, which has a high content of calcium (Ca), aluminum (Al) and silicate (Si), is used for construction materials [3,18]. The maximum CO₂ sequestration potential of bituminous coal fly ash is relatively low (~9%); however, it could reach as high as 43–49% for Ca-rich lignite or low-rank coal fly ash [18,19]. The high-Ca fly ash (>20%) produced from lignite or sub-bituminous coals is comprised of Ca–Al–Si glass and a wide variety of crystalline phases. These crystalline phases can react with water and harden due to the formation of cementitious hydration products, which causes the fly ash to react more rapidly than low-Ca ash and renders the fly ash both pozzolanic and hydraulic [20]. However, excessive amounts of magnesia (MgO) or free lime (CaO) in the cementitious materials may cause undesirable volume changes when these materials are used in concrete [20]. It was determined that an MgO content of up to 5% in cement is useful for modifying the shrinkage behavior of concrete [21]. A prior removal of free MgO and CaO out of the fly ash in a mineral carbonation process can selectively remove and convert free MgO and CaO into carbonates. In addition to the above-mentioned benefits for a typical mineral carbonation process, this process is also beneficial for the production of cement additive-grade by-products. The development of an appropriate mineral carbonation technology using fly ash as feedstock may help enhance the economic, technological and environmental benefits of fly ash [22].

Currently, only a few commercial technologies are available for the carbonation of industrial wastes [23,24]. Thus far, the most

comprehensively studied process for fly ash carbonation is the direct route, which involves high temperature and CO₂ pressure in the range of 100–500 °C and 10–20 bar, respectively [3,19,25–28]. Montes-Hernandez et al. achieved a carbonate conversion of 82% and a carbon sequestration capacity of 26 kg CO₂/t ash at 30 °C and 10 bar initial CO₂ pressure over 18 h in water [19]. Uliasz-Bochenczyk et al. achieved a maximum carbon sequestration capacity of 7.85 g CO₂/100 g ash at ambient temperatures and 10 bar CO₂ initial pressure over 24 h in water [25]. There is a noticeable lack of published techno-economic feasibility studies in this area that could potentially support the feasibility of these processes [18,29]. It is envisioned that mineral carbonation demonstration plants of up to 1Mt CO₂ per annum may be operational as early as 2020 [30].

We previously tested the possibility of the application of Victorian brown coal fly ash in the indirect mineral carbonation process [12]. This process involved the use of ammonium chloride (NH₄Cl), which initially extracted the free MgO and CaO in the fly ash into an aqueous form *via* the substitution reaction of NH₄⁺ + Ca/MgO = NH_{3(g)} + Ca²⁺/Mg²⁺. The resulting Ca²⁺ and Mg²⁺ cations were subsequently carbonated *via* a continuous bubbling of CO₂-containing flue gas and ammonia (NH₃) for pH control. These ammonium salt solvents were found to be recyclable, although minor losses of ammonia were noted [12]. The objectives of the research presented here is to evaluate whether the proposed mineral carbonation method could be scaled up to a relative large scale for both the CO₂ capture and the production of high-purity carbonate products to be sold to compensate for the high cost of the process. Process flow-sheeting was initially schemed using Aspen Plus[®] based on our earlier experimental studies. Subsequently, four different scenarios were established to optimize the process integration to minimize the process energy consumption. Then, an economic analysis was conducted using the US-based economic analysis package as well as a modified version to assess the cost for different contexts in the world. Lastly, a sensitivity analysis was performed to examine the variation in the production cost with different operating and economical parameters, and a cash flow analysis was performed to clarify the profitability of the process by determining its net present value (NPV), internal rate of return (IRR) and payback period.

2. Methodology

2.1. Process description

The block diagram of the proposed mineral carbonation process using regenerative ammonia chloride is presented in Fig. 1, including the system boundaries of this study. The mineral carbonation plant was assumed to be located next to the Victorian brown coal-fired power plant, which produces both fly ash and flue gas in Latrobe Valley, Australia. The overall process consists of two major areas: fly ash leaching and carbonation. Fly ash was assumed to be supplied free, as slurry, at the battery limits. In this study, the fly ash produced by the International Power Hazelwood power plant was used as a feedstock. The typical fresh and weathered fly ash compositions used for simulation as well as the variation range of individual species are presented in Table 1 [31]. Depending on the ageing extent and the heterogeneity of the coal, the composition of the fly ash is considerably variable. Due to the limitations of the model to consider all of these changes, an average composition of a typical fresh and weathered fly ash was used as the input data. Despite the potential uncertainties in the production yield when changing the fly ash composition, a sensitivity analysis can predict a few of these changes and provide more flexibilities to link the model to actual cases.

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