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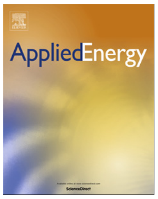

Pilot-scale experimental work on carbon dioxide sequestration using steelmaking slag

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HIGHLIGHTS

- The world's first pilot plant that converts steel slag and CO_2 into PCC.
- The pilot plant can handle up to 20 kg steel slag and 190 L of liquid solvent.
- Calcium extraction efficiency is about 80%.
- Solvent can be regenerated and reused.

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ABSTRACT

The production of precipitated calcium carbonate (PCC) from steel slag has been proposed as a potential method of simultaneously reducing the CO_2 emissions from the steelmaking process and turning its waste stream into a valuable product. On average the production of one ton of steel results in two tons of CO_2 emissions and 600 kg of slag. Globally, more than 400 Mt of steel slag are produced annually. If all the slag were used for the production of PCC, 64 Mt CO_2 could be utilized and 145 Mt of calcium carbonate would be produced. In 2014 the research group Energy Engineering and Environmental Protection at Aalto University in Finland has designed, constructed and tested the world's first mineral carbonation pilot plant test facility that converts steel slag and CO_2 into PCC. In batch mode the pilot plant can handle up to 20 kg of solid steel slag and 190 L of liquid solvent, and it can produce about 10 kg of calcium carbonate. The solvent can be regenerated and reused in the calcium extraction stage, which makes the process economically more feasible. Almost 80% of the calcium in the slag was extracted, while more than 70% of the CO_2 was utilized and converted into PCC. In high temperature carbonation tests, ammonia gas was detected from the flue gases. At 60 °C more than 2 vol.% of NH_3 was detected in the flue gas, and at 50 °C it was 0.65 vol.%, while at 45 °C the NH_3 concentration in the flue gas was only 0.11 vol.%. To avoid ammonia evaporation, aragonite PCC can be produced at 45 °C by optimizing the CO_2 flow rate.

The paper presents the process design as well as the early results achieved from the pilot plant. The paper also presents technical challenges that occurred during the scale-up work and experiments.

1. Introduction

The mineralization of CO_2 has been considered to be a potential technology for the sequestration of CO_2. It provides permanent storage for CO_2 without the requirement of capture, a storage site, transport pipelines, or monitoring for leakage problems. In other words, the mineralization of CO_2 offers permanent storage for CO_2 without leakage and thus provides greater public acceptance [1]. In addition to that, the final product, precipitate calcium carbonate (PCC), will be valuable if it meets certain product qualities like purity and particle size distribution. On the other hand, the mineral carbonation technology has some barriers and challenges that must be addressed: scalability, large-scale material consumption, and energy intensiveness.

To meet these challenges, researchers have made several studies over the last few years, and reported a number of advanced process routes [2–18]. However, despite these promising achievements, so far no mineral carbonation case has found a large-scale application or even the pilot or demonstration stage [19].

In 2014 the research group Energy Engineering and Environmental Protection at Aalto University in Finland has designed, constructed and tested the world's first mineral carbonation pilot

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plant test facility that converts steel slag and CO_2 into PCC the so-called Slag2PCC concept [20]. Also researchers at Åbo Akademi University have built 28 L reactors aimed at process optimization and crystal morphology control for the Slag2PCC process [19]. In the transition from laboratory scale to commercial scale, it is extremely important to have an intermediate scale, the pilot scale, to validate the techno-economic feasibility of the technology. Therefore, the scale-up of the Slag2PCC process is a major step forward for the research and development of CO_2 sequestration by minerals and one step closer to the commercialization of the technology.

The production of PCC from steel slag has been proposed as a potential method of simultaneously reducing CO_2 emissions from the steelmaking process and turning its waste stream into a valuable product [14–15,19–24]. On average the production of one ton of steel results in two tons of CO_2 emissions and 400 kg of blast and basic oxygen furnace slag, and 200 kg of electric arc furnace slag. Globally, more than 400 Mt of steel slag is produced annually (Fig. 1) [25].

In the Slag2PCC process, calcium is selectively extracted from the steel slag by an ammonium salt solvent (e.g., NH_4Cl, NH_4NO_3, or CH_3COONH_4). After filtration the filtrate (a Ca-rich solution) reacts with CO_2 gas to precipitate PCC [14–15,19–23]. A fundamental investigation of the process was conducted on a laboratory scale over a number of years and promising results were achieved. These lab-scale experiments have now been scaled-up 400 times—from a 0.5 L reactor to a 200 L reactor—prior to a pre-commercial stage. Some laboratory-scale results indicated an eventual loss of NH_3 during the carbonation process [26]. Thus, it is critical to quantify the level of NH_3 loss at different carbonation temperatures and offer solutions to avoid the problem.

The pilot plant can handle at batch mode up to 20 kg of solid steel slag and 190 L of liquid solvent, and it can produce about 10 kg of PCC. After carbonation, the chemical solvent can be recovered and reused in the calcium extraction stage, which makes the process economically more feasible. Steel slag is not a homogenous material, comprising a variety of other elements that could be dissolved together with calcium by ammonium salt solutions, resulting in a lot of impurities in the process. The pilot plant aimed to demonstrate the true potential of the Slag2PCC technology as an example of green technology that enables the conversion of CO_2 emissions and industrial solid waste into a very valuable product, such as PCC. Therefore this study reveals techno-economic assessments of CO_2 sequestration from steelmaking slag to produce high-quality PCC.

This paper will present the process design as well as the early results achieved from the Slag2PCC pilot plant test facility. The paper will also present technical challenges that occurred during the scale-up work and experimental time. The possibility of extracting additional calcium from the residual slag by a second extraction stage was also investigated. In order for the Slag2PCC method to be ecumenically feasible, solvent recyclability is extremely important as the process otherwise becomes too expensive. Thus, the paper will present experiment results on the possibility of recycling the used NH_4Cl solvent and its implications for Ca extraction efficiency.

2. The Slag2PCC process's scale-up

Fig. 2 shows the design of the Slag2PCC pilot plant and Table 1 summarizes the key components of the pilot plant. It consists of three reactors, each having a maximum volume of 200 L: an extraction reactor (Reactor 1) and two carbonation reactors (Reactors 2 and 3). In Reactor 1, calcium is extracted from steel slag, after which this Ca-rich solution is filtered through extraction filters (Extraction Filters 1 and 2). These filters are equipped with 1 μm nylon filter bags to remove the bulk of the slag. To guarantee the purity of the Ca-rich solution prior the carbonation stage, a secondary filtration stage (qualitative filtration) was introduced for removing particles up to 0.45 μm. After filtration, the Ca-rich solution can be stored in a reserve tank (Tank 1) for intermediate storage or carbonated directly in a carbonation reactor, Reactor 2. After carbonation, the PCC is collected using an identical filtration system in the extraction stage (PCC Filters 1 and 2, followed by qualitative filtration) and the filtrate (the recovered solvent) can be stored in a solvent tank (ST1), or it can be directly pumped into the secondary carbonation reactor (Reactor 2). The aim of the secondary carbonation reactor is to purify the recovered solvent from the small amount of unreacted calcium in the Ca-rich solution prior to its recycling in the extraction reactor.

Each reactor (Reactors 1, 2, and 3) is equipped with temperature and pH indicators as well as an agitator with a maximum speed of 202 rpm for Reactor 1 and 170 rpm for Reactors 2 and 3. The primary carbonation reactor has a gas sparger with 470 holes that have a diameter of 1 mm each. The gas sparger is below the agitator and 5 cm above the bottom of the reactor, it points downwards to avoid any rapid blocking of the sparger holes. The pilot plant also includes two plate heat exchangers (HX-1 and HX-2) that

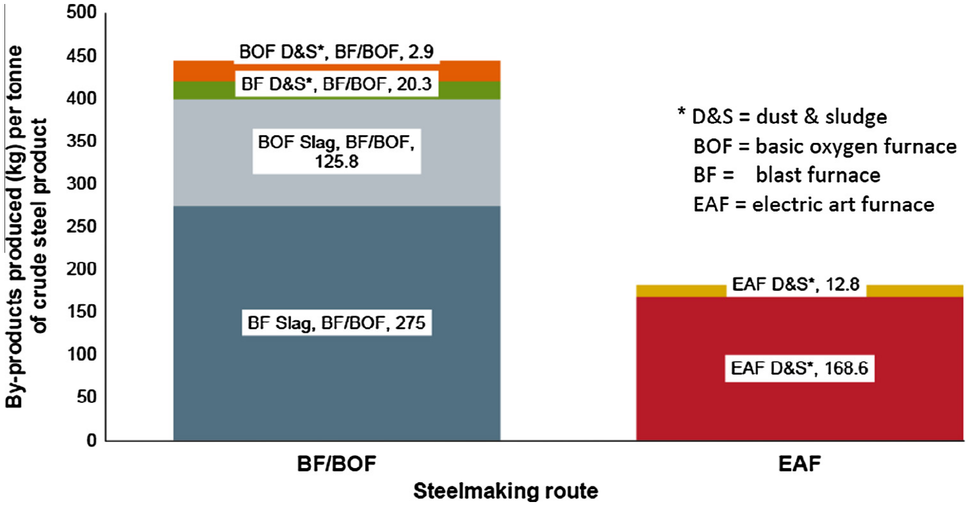

Fig. 1. The average by-products from steelmaking processes [25].