Applied Energy 106 (2013) 116–126

Contents lists available at SciVerse ScienceDirect

Applied Energy

journal homepage: www.elsevier.com/locate/apenergy

Performance assessment of producing $Mg(OH)_2$ for CO_2 mineral sequestration

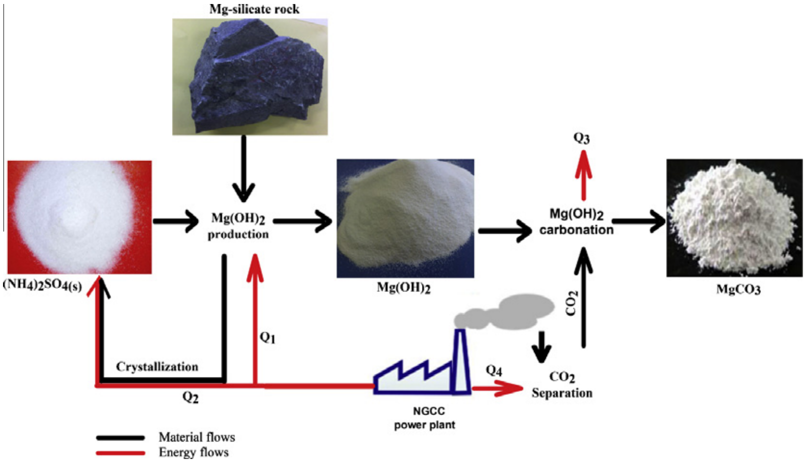
Experience Nduagu [a,*], Inês Romão [a,b], Johan Fagerlund [a], Ron Zevenhoven [a]

[a] *Thermal and Flow Engineering, Åbo Akademi University, Biskopsgatan 8, 20500 Åbo/Turku, Finland*
[b] *University of Coimbra, Coimbra, Portugal*

HIGHLIGHTS

- ▶ Producing $Mg(OH)_2$ for CO_2 mineral sequestration was assessed.
- ▶ Reactor properties, reaction temperature and time affect Mg extraction.
- ▶ A 100% efficient CO_2 mineral sequestration process avoids 567 $CO_2e/t\text{-}CO_2$ net.
- ▶ The process avoids 51% life cycle CO_2 emissions of a NGCC power plant.
- ▶ The process reduces the net power of a NGCC power plant by 20%-points.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 27 August 2012
Received in revised form 14 January 2013
Accepted 16 January 2013
Available online 15 February 2013

Keywords:
Mineral carbonation
Serpentinite
Magnesium hydroxide
Process assessment
Life cycle analysis

ABSTRACT

This study presents the energy and environmental performance assessment of producing magnesium hydroxide ($Mg(OH)_2$) from Mg–silicates for CO_2 mineral sequestration applied to a natural gas combined cycle (NGCC) power plant. $Mg(OH)_2$ produced via a closed loop reaction of serpentinite and ammonium sulfate (AS), precipitation of $Mg(OH)_2$ and AS looping/recovery binds CO_2 into a thermodynamically stable, environmentally benign and leak-free magnesium carbonate ($MgCO_3$). We used results from laboratory, modeling and life cycle assessment (LCA) studies to determine the extent to which magnesium (Mg) from serpentinite rock can be converted to $Mg(OH)_2$, the effects of reaction parameters, scalability and the associated life cycle greenhouse gas emissions (GHGs). We found that reaction temperature positively affects Mg extraction from serpentinite, reaching a maximum yield at different temperatures depending on the reaction time. Also, the reactor properties affect the extraction results as the optimal extraction yield and conditions reported for different reactors differ. While the process of producing $Mg(OH)_2$ is promising, it also possesses a level of energy and environmental burden that cannot be ignored when considering large scale implementation. At 100% conversion and recovery of reagent, the CO_2 mineralization process has a life cycle global warming potential (GWP) of 433 kg CO_2 equivalents per ton CO_2 ($CO_2e/t\text{-}CO_2$). This value increases by 82, 7 and 0.4 kg $CO_2e/t\text{-}CO_2$ for every %-point efficiency loss of

---

* Corresponding author. Tel.: +358 2 215 3223; fax: +358 2 215 4792.
E-mail address: experience.nduagu@abo.fi (E. Nduagu).

http://dx.doi.org/10.1016/j.apenergy.2013.01.049

AS recovery, $Mg(OH)_2$ production and $Mg(OH)_2$ carbonation respectively. Mineral sequestration applied to the 555 MW NGCC plant reduces its net plant efficiency from 50.2% to 38.6%-points (an energy penalty of 30%) but avoids 51% of the GHG emissions to the atmosphere. The results from this study are timely, and could have significant implications on mineral sequestration methods that consider the exothermic nature of the overall mineral carbonation chemistry beneficial.

1. Introduction

CO_2 mineral sequestration (CMS), like other carbon capture and storage (CCS) options, could – while transforming society towards a renewable energy economy – ensure that current and future energy demands are met in a "carbon neutral" manner [1–3]. Unlike the widely advocated storage of CO_2 in geologic formations, CO_2 mineral sequestration will eliminate the fear of a post-storage CO_2 leakage risk and the associated monitoring costs. The reason is that sequestering CO_2 in mineral rocks has the benefit of thermodynamic stability and permanence [4]. But CO_2 mineral sequestration still suffers from two major setbacks: prohibitive energy requirement and poor economics. Sequestration costs are too high compared to other CO_2 storage options, considering CO_2 market prices [5]. Energy is required as an input into the CO_2 mineral sequestration processes to overcome the kinetic barrier and to speed up the reaction via activation. Efforts to address the slow kinetics of mineral carbonation led to employing several activation mechanisms that accelerate liberating reactions of Mg from Mg–silicate rocks. Through activation, the reaction rate and conversion are increased, but the energy and cost penalties increase as well. An additional energy, cost and environmental burden is created by CO_2 capture, and mining and processing large amounts of mineral rocks (2 tons or more mineral per ton CO_2).

CO_2 mineral sequestration processes that employ more than one stage process make simultaneous or step-wise extraction and carbonation of Mg ions in the Mg–silicate mineral rocks possible [6,7]. A staged process of CO_2 mineral sequestration via gas/solid carbonation has been a major focus of over a decade mineralization research initiated at Helsinki University of Technology and currently primarily at Åbo Akademi University (hereafter ÅA), Finland. In this process, we produce $Mg(OH)_2$ from Mg–silicate rocks using a closed loop, staged Mg extraction reaction followed by $Mg(OH)_2$ precipitation and AS looping [8–10]. The $Mg(OH)_2$ thus produced can be used to sequester CO_2 through many routes: (i) direct and indirect reaction with CO_2 or CO_2 containing (flue) gases [11–16] (ii) dry or aqueous reaction with CO_2 from air or flue gases at ambient conditions [17] and (iii) increasing ocean alkalinity by enhancing the capacity of oceans to capture more atmospheric CO_2.

We present here a proof-of-concept and feasibility assessment based on results from optimized laboratory tests, and a systematic evaluation study of environmental performance of producing $Mg(OH)_2$ for CO_2 mineral sequestration using life cycle assessment (LCA). Although previous laboratory test results gave promising results, many questions bordering on several aspects of the process and its potential for scale-up into a pilot process so far remained unanswered. Some of the questions of interest include but are not limited to:

(i) To what extent do identified parameters influence production of $Mg(OH)_2$? What are the realizable maximum yields of extraction of Mg from Mg–silicate rocks? Previous studies [8,10,18] provide a good background to this question, but the results are somewhat inconclusive, especially regarding the effect of reaction time, mixing and reactor type and configuration. We carried out additional laboratory tests to better understand the roles these parameters play, as they give guidelines as to how a given rock should be processed.

(ii) How can scale-up from laboratory to pilot scale be achieved? Here, we expanded on our understanding of process scalability by performing tests with a bench scale equipment, exploring the role of mixing.

(iii) What are the life cycle energy and environmental impacts of producing $Mg(OH)_2$ from Mg–silicate rocks for sequestration of CO_2 emissions from a natural gas (NG) combined cycle power plant?

(iv) Possible options to improve the process efficiency as well as reduce the process greenhouse gas emissions (GHGs) footprint are suggested.

2. Experimental methods

We present here the methods applied to the laboratory studies of producing $Mg(OH)_2$ from serpentinite rock for the sequestration of NGCC-derived CO_2.

2.1. Procedure for Mg extraction and $Mg(OH)_2$ production

$Mg(OH)_2$ was produced from Finnish serpentinite rock (~22 wt.% Mg, ~12 wt.% Si, ~10 wt.% Fe and 1 wt.% Ca) following the concepts and techniques previously described [8–10]. However, new equipment setups were used for the experiments reported here. A Mg extraction stage involves the reaction of pulverized serpentinite rock (75–125 μm) with reagent grade ammonium sulfate (AS) at 250–550 °C to produce primarily magnesium sulfate, silica, water vapor (steam) and ammonia gas according to following equation:

$$\begin{aligned}&Mg_3Si_2O_5(OH)_4(s) + 3(NH_4)_2SO_4(s)\\&\rightarrow 3MgSO_4(s) + 2SiO_2(s) + 6NH_3(g) + 5H_2O(g)\end{aligned} \tag{1}$$

Iron and calcium compounds in the mineral rocks also react with AS to form corresponding sulfates. Hot gases, mainly steam and ammonia, produced from the extraction reaction are recoverable for use in downstream precipitation of FeOOH and $Mg(OH)_2$. The solid products were cooled, weighed, dissolved in water in order to leach the extracted elements.

After separating the residue, mainly silica from the mixture by filtration, 25 wt.% aqueous ammonia solution was then added to the remaining solution to precipitate (oxy)-hydroxides of Fe and Mg. FeOOH was precipitated at a pH of 9–10 while $Mg(OH)_2$ was precipitated at a pH of 11–12.

2.2. Parameters affecting the extraction reaction

The processing of the serpentinite rock entailed pulverizing, separating into particle size fractions (<74 μm, 75–124 μm, 125–249 μm and 250–549 μm) and washing with distilled water to remove very fine "dust-like" fractions not removed during sieving. The 75–125 μm particle size fractions, washed in distilled water