GHGT-10

Carbon capture and utilization: preliminary life cycle CO₂, energy, and cost results of potential mineral carbonation

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Abstract

Mineral carbonation has been identified as a potentially suitable means of CO₂ sequestration in Singapore due to the nation’s lack of land for geological or deep ocean storage of CO₂. In this article, the total energy, CO₂ emissions and costs of mineral carbonation are investigated using a life cycle assessment (LCA) approach. The life cycle investigation took into account energy and greenhouse gas emissions from mineral mining activities and shipment, the recovery of CO₂ based on amine scrubbing technology and simulated scenarios of the net energy requirements for the carbonation process based on ‘ideal’ and worst case energy requirements. The CO₂ avoided results from a total of 4 scenarios were in the range of 106.9 kg to 175.9 kg per 1 MWh. The percentage sequestration effectiveness results are from 32.9% to 49.7%. The life cycle costing results are 105.6 USD/tonne CO₂ avoided and 127.2 USD/tonne CO₂ avoided for two of the most favorable scenarios. However, it is highlighted that various engineering challenges have to be overcome before the ‘ideal’ carbonation reaction conditions represented in the simulation model can be achieved. The results will most likely fluctuate somewhere between the ideal and worst case conditions. The main energy penalties and associated CO₂ emissions come mostly from CO₂ recovery, pre-treatment and mineralization process itself.

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Keywords: Life cycle assessment; CO₂ mineralization; simulation; energy use; CO₂ avoided

1. Introduction

International concerns over global warming have identified the urgent need for large-scale sequestration, reduction, or utilization of CO₂. Any aims to effectively capture and store large volumes of CO₂ should take into account the life cycle of energy use, and overall carbon footprint of the sequestration system itself [1-2]. In Singapore, mineral carbonation has been identified as the most suitable means of CO₂ sequestration due to lack of land for geological storage and ocean territories [3-4]. Moreover, carbon sequestration via mineralization is also suggested as the safest and most stable way of locking away large amounts of CO₂ [5]. Huge deposits of alkaline-
Earth (Mg-based) silicate minerals of the peridotite and serpentinite families exist in countries around Singapore. Two of these sources were identified to be scattered around Kalgoorlie mining areas in Western Australia (WA) and Tasmania, Australia.

2. Mineral carbonation

A simulation study of the energy requirements for carbonation using serpentine is presented here. Thermodynamic calculations were carried out using HSC Chemistry 6.0 [6] and corrected data for MgCO₃ [7] to explore the possible sets of operating conditions, including the range of temperature and pressure for which the carbonation process can be feasible. The following reaction pathway was applied [8]:

\[
\frac{1}{3}[3\text{MgO} \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O}] \rightarrow \text{MgO} + \frac{2}{3} \text{SiO}_2 + \frac{2}{3} \text{H}_2\text{O}(g) \quad (1)
\]

\[
\text{MgO} + \text{H}_2\text{O}(g) \rightarrow \text{Mg(OH)}_2 \quad (2)
\]

\[
\text{Mg(OH)}_2 + \text{CO}_2(g) \rightarrow \text{MgCO}_3 + \text{H}_2\text{O}(g) \quad (3)
\]

The net resultant reaction is:

\[
\frac{1}{3} [3\text{MgO} \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O}] + \text{CO}_2(g) \rightarrow \text{MgCO}_3 + \frac{2}{3}\text{SiO}_2 + \frac{2}{3}\text{H}_2\text{O} \quad (4)
\]

The process flow diagram for the carbonation steps are shown in Figure 1. The overall process is exothermic and hence energy is released in the form of heat which is conveniently used to separate the solid products MgO and SiO₂. This assumption can be made if negligible energy input is required for separation of serpentine into MgO, SiO₂, and H₂O. The stream data for the process is documented in Table 1. The results, displayed as Figure 2, are generated by the application of pinch analysis [9]. From the grand composite curve (Figure 2), the hot and cold utilities required are 1843 MW and 3320 MW respectively. The highest amount of energy required for the carbonation process is formulated according to the hot utility shown in the graph.

![Figure 1 Block flow diagram for carbonation](image)

**Table 1 Stream data for temperature interval analysis (based on 1 tonne CO₂/s and ΔT = 10°C)**

<table>
<thead>
<tr>
<th>Stream</th>
<th>T_in(°C)</th>
<th>T_out(°C)</th>
<th>MCp (MW/K)</th>
<th>Heat Load (MW)</th>
</tr>
</thead>
<tbody>
<tr>
<td>C1 Preheat</td>
<td>25</td>
<td>600</td>
<td>2.8</td>
<td>1596</td>
</tr>
<tr>
<td>C2 Mg-Si → MgO</td>
<td>600</td>
<td>600.5</td>
<td>3630</td>
<td>1815</td>
</tr>
<tr>
<td>H1 MgO → Mg(OH)₂</td>
<td>170</td>
<td>169.5</td>
<td>3609</td>
<td>-1805</td>
</tr>
<tr>
<td>H2 Mg(OH)₂ → MgCO₃</td>
<td>170</td>
<td>169.5</td>
<td>1727</td>
<td>-864</td>
</tr>
<tr>
<td>H3 MgCO₃ cool down</td>
<td>170</td>
<td>45</td>
<td>2.0</td>
<td>-250</td>
</tr>
<tr>
<td>H4 MgO cool down</td>
<td>600</td>
<td>170</td>
<td>1.1</td>
<td>-467</td>
</tr>
<tr>
<td>H5 SiO₂/H₂O cool down</td>
<td>600</td>
<td>45</td>
<td>2.7</td>
<td>-1503</td>
</tr>
</tbody>
</table>
The energy required for the pretreatment of serpentine was estimated with the help of the thermodynamic data reported by King et al. [10] along with the effective heat capacities and temperature provided by Penner et al. [11]. According to the authors, the energy required for pretreatment was calculated by:

$$Q = C_P \Delta T$$  \hspace{1cm} (5)

Where $Q$ = heat (cal/mol); $C_P$ = cal/Kmol @ temperature $T_1$ (K); and $\Delta T = T_1 - T_0$ (298K).

From (5), the heat treatment of serpentine at 630°C ($C_P = 89.26$ cal/K mol) requires 206 kWh/tonne to heat the mineral, while dehydroxylation of the mineral requires another 87 kWh/tonne. The total for energy consumption for the heat treatment process is thus 293 kWh/tonne [11].

The uptake of CO$_2$ by the minerals may be enhanced by taking advantage of the heat released by the exothermic carbonation reaction. Realistically, the conversion rates can be between 80-90% [4-5]. Based on the simulation results (Figures 1 and 2), the net energy requirement for mineral carbonation are as follows:

- **Ideal case**, where it is assumed that the carbonation reaction generates enough heat energy to feed itself (energy input = energy output). The conversion of minerals to carbonate for an ideal case is taken to be 90%.
- **Worst case scenario**, where virtually none of the heat energy can be recovered for use. Therefore the maximum net energy required is simulated to be 1850 MJ/tonne CO$_2$ carbonated. The extent of reaction for this case is taken as 80%.

3. **Life cycle assessment**

The case study of the LCA covers mineral mining, crushing and packaging of mineral rocks, before shipment to Singapore. Next the CO$_2$ recovery from a natural gas combined cycle (NGCC) power plant is taken into account, and finally, CO$_2$ carbonation. The functional unit selected is 1 MWh generated from the NGCC power plant in Singapore. The goal of the LCA is to compare the life cycle CO$_2$ (or carbon footprint), energy requirements and costs of mineral carbonization in Singapore for four scenarios, which are tabulated in Table 2. The life cycle system boundary, along with the input-output flow of energy and CO$_2$, is illustrated in Figure 3. Within the LCA system, the following activities are considered to model the energy requirements and CO$_2$ emissions:

- Mining, crushing and packaging of minerals from Kalgoorlie mining area in Western Australia (WA) and Tasmania, Australia.
- Shipment of mineral rocks to Singapore. The nearest ports from WA and Tasmania to Singapore are Fremantle and Melbourne. The shipment distances are 6991 and 3986 kilometers respectively.
- The recovery of CO$_2$ from the flue gas of an NGCC power plant based on amine scrubbing technology. The CO$_2$ recovery rate is 90% with an energy penalty of 16%.
- The simulated energy and extent of reaction for CO$_2$ mineralization are based on the ‘ideal’ and ‘worst’ cases. In both cases, the ratio of serpentine to CO$_2$ is considered to be 2:1.

The LCA scenarios are as follows:

1. Minerals purchased from WA and processed with ‘ideal case’ mineralization reaction
2. Minerals purchased from WA and processed with ‘worst case’ mineralization reaction
3. Minerals purchased from Tasmania and processed with ‘ideal case’ mineralization reaction
4. Minerals purchased from Tasmania and processed with ‘worst case’ mineralization reaction
The energy requirements for mineral mining, crushing and packaging are extracted from EcoInvent [12] and Hangx and Spiers [13]. These total energy requirements are assumed to be similar to limestone rock mining. The associated CO₂ emissions due to energy usage differ from place to place. The CO₂ inventory is extracted from Hydro Tasmania [14] and CARMA [15]. The energy requirements of CO₂ recovery from a power plant in Singapore is calculated based on amine scrubbing utilizing monoethanolamine or MEA. The energy penalty for an NGCC power plant is 16% with CO₂ recovery rates of 90% [16]. Energy demands (electricity and heat) for amine scrubbing of CO₂ from the power plant flue gas can be as high as 3570 MJ/tonne CO₂ [17]. Carbon dioxide emissions from shipment are also taken from EcoInvent [18].

4. Results and discussions

4.1. Source of mineral

The total CO₂ from mineral sourcing and transportation is shown in Figure 4. It is observed that shipment takes up the main portions of each graph, especially for cases 3 and 4. This is due to the large transport distance travelled for the delivery of minerals to Singapore. Comparatively, emissions from mining activities are less significant. However, it should be highlighted that only CO₂ emissions were considered. A large amount of air pollution from mining activities is dust, which has a detrimental affect on human health [19]. This environmental and health concerns should be taken into consideration for further LCA studies.
4.2. Life cycle CO₂ and energy use

The LCA results for CO₂ and energy use are displayed in Figure 5 and 6 respectively.

![Figure 5 Life cycle CO₂ results](image)

From Figure 5, the inverted peaks represent the amount of CO₂ carbonated and prevented from entering the atmosphere. It is observed that the most preferred case is scenario 1, where the minerals are obtained from WA and the energy requirements for carbonation is ideally supplied by the heat generated from its own exothermic reaction. It is also highlighted that the main concerns of greenhouse gas emissions are actually from, principally, the CO₂ recovery system and, next, the energy used for pre-treatment and carbonation. Compared to these two stages the emissions from mining and transportation are very much less significant. Scenario 4 turned out to be the least favorable option. The benefits are mostly reduced by the large amount of emissions arising mainly from CO₂ recovery, pre-treatment and carbonation.

![Figure 6 Life cycle energy results](image)

While the CO₂ emissions of Figure 5 are mostly influenced by the potential amount of CO₂ carbonated, energy use (Figure 6) is dominated by the energy requirements of the CO₂ recovery system. Before CO₂ can be carbonated, there is an intermediate step of separating and recovering it from the power plant’s flue gases. Highly intensive
energy demands are required for amine scrubbing of CO₂, especially for the heat regeneration process. In this article, heat energy demands of 3570 MJ/tonne CO₂ is used, but even higher energy demands of up to 4500 to 5700 MJ/tonne was reported by Harkin et al. [20] for the same CO₂ removal technology.

Energy requirements for carbonation are process-dependent and it is influenced by a wide range of parameters including the energy required to separate MgO from serpentine, heat, pressure and other reaction kinetics [7-8, 10-11]. The energy requirements for both pre-treatment and carbonation deserve further explorations and experiments to make the entire process feasible.

4.3. CO₂ avoidance and percentage sequestration effectiveness

The amount of CO₂ avoided can be defined as:

\[ CO₂ \text{ avoided} = NGCC_{CO₂} - \Sigma CCS_{CO₂} \]  

(6)

Where \( NGCC_{CO₂} \) = amount of CO₂ emissions from NGCC power plant without any capture system in place. This value is reported to be 380 kg/MWh.

\( \Sigma CCS_{CO₂} \) = the accumulated CO₂ emissions totalled from mineral mining, transportation, CO₂ recovery and carbonation.

From here, the percentage sequestration effectiveness is calculated as:

\[ \frac{Amount \ of \ CO₂ \ Carbonated - [\Sigma CCS_{CO₂}]}{Amount \ of \ CO₂ \ Carbonated} \times 100\% \]  

(7)

The graphical illustration of the total CO₂ avoided is displayed in Figure 7. The results are shown in table 2.

Based on the results, scenario 4 is omitted and the costs of scenarios 1, 2 and 3 are investigated.

4.4. Simplified life cycle costs
Life cycle costing is a method that can be used to take into account the cost components of materials and energy flows within an LCA system. A simplified life cycle costing (LCC) is carried out according to the following equation:

\[
\text{Life cycle cost} = C_{\text{Capital}} + C_M + R_{CO2} + C_T + C_{NG}
\]  

(8)

Where

- \( C_{\text{Capital}} \) = capital cost of equipment, including maintenance
- \( C_M \) = cost of minerals in USD/tonne
- \( R_{CO2} \) = cost of CO\(_2\) recovery from flue gas (USD/tonne CO\(_2\))
- \( C_T \) = cost of transportation/shipment (USD/tonne-km)
- \( C_{NG} \) = cost of energy from natural gas for mineralization process (USD/MJ)

Data for transportation were estimated from shipping rates, distance between ports (and subsequently the duration of time at sea), and shipping rates from Lloyd’s Shipping Economist [21]. As a conservative estimate, the cost of CO\(_2\) recovery and price of natural gas are taken from IPCC [15] and Johnson and Keith [22]. The costs of minerals were taken from U.S. geological survey [23]. The compiled costing data are shown in table 3.

**Table 3  Life cycle cost data**

<table>
<thead>
<tr>
<th>Cost parameters</th>
<th>( C_M ) (USD/tonne)</th>
<th>( R_{CO2} ) (USD/tonne CO(_2))</th>
<th>( C_T ) (USD/tonne-km)</th>
<th>( C_{NG} ) (USD/MJ)</th>
</tr>
</thead>
<tbody>
<tr>
<td>7</td>
<td>33.0</td>
<td>0.0018</td>
<td>0.003</td>
<td></td>
</tr>
</tbody>
</table>

Without considering \( C_{\text{Capital}} \), the results for scenarios 1, 2 and 3 are USD 88, 150 and 106 per tonne CO\(_2\) avoided respectively. IPCC reported that \( C_{\text{Capital}} \) may be estimated as 20% of the total (LCC) [24]. By incorporating this cost component, the LCC results are adjusted to be 105.6 USD/tonne CO\(_2\) avoided for scenario 1, 180 USD/tonne for 2, and 127.2 USD/tonne CO\(_2\) avoided for scenario 3. In comparison, a report from IEA [25] on energy technologies in year 2050 suggests carbon capture and storage projects are projected to cost less than 150 USD per tonne CO\(_2\) avoided. Hence, the cost results for all three scenarios fall within a reasonable range.

5. Conclusions and recommendations

Based on the 4 scenarios, the CO\(_2\) recovery of 90% from the NGCC flue gas resulted in a total of 175.2, 117.6, 166.3 and 106.9 kg CO\(_2\) avoided per 1 MWh delivered to consumers. The range of sequestration effectiveness is from 32.9% to 49.7%. Further analysis estimated the life cycle costing (LCC) results to be 105.6 USD/tonne CO\(_2\) avoided for scenario 1 and 127.2 USD/tonne CO\(_2\) avoided for scenario 3. From the overall results, scenarios 1 and 3 turn out to be the most favourable. However, in reality the ‘ideal’ conditions represented by scenarios 1 and 3 are not easily achievable. The results will most likely fluctuate somewhere between scenarios 1 and 2; or 2 and 3. Apart from simulation work focusing on thermodynamics, experimental studies should target at obtaining the optimal net energy generation in the range between these two scenarios.

The results indicate that main focus for the entire carbon capture and utilization system should primarily be on reducing the energy demands for CO\(_2\) removal technologies [17], as well as, optimizing the use of heat energy generated by the exothermic carbonation reaction. Currently, experimental studies focusing on dry carbonation of Si and Mg silicates have been rather slow and are hindered with various engineering limitations [7-9]. One of the main goals is to optimally utilize the amount of heat produced by the carbonation reaction, which in principal, can be recovered as a high enthalpy system [26]. Another technical challenge lies in the extraction of magnesium from the minerals without the use of chemicals or high energy demands [3-5].

Apart from CO\(_2\), LCA investigations of carbon capture and storage should also expand to include other kinds of pollution, including dusts, acidic gases, and solid wastes. Such studies are exemplified by Khoo and Tan [1-2] and Singh et al. [27]. The authors compared a wide range of environmental aspects from a myriad of options for CO\(_2\) recovery technologies, transportation, and storages. Apart from global warming impacts alone, the environmental concerns of acidification, eutrophication, human toxicity to air and water, and solid waste were taken into account.
6. References


[16] IPCC. *Carbon Dioxide Capture and Storage* - IPCC Special Report. UN Intergovernmental Panel on Climate Change; 2005.


[21] Lloyd’s Shipping Economist; 2009.


