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Literature Thesis

CO$_2$ sequestration and utilization in cement-based materials

An analysis on economic feasibility and environmental impact

by

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Abstract

Reducing mankind’s carbon footprint is important to stabilize the atmospheric CO$_2$ concentration and bring a hold to human-induced climate change. One way to reduce atmospheric carbon is by Carbon Capture and Utilization (CCU). When CO2 is used as a feedstock, the use of fossil feedstock (with associated emissions) is avoided. This report analyzes technologies that aim to convert CO$_2$ into valuable cement-based building materials and assesses their potential of reducing atmospheric CO$_2$. First a general introduction on climate change and CCU is provided, followed by some background information about the cement and concrete industry, about industrial wastes that are reactive to CO$_2$ and about carbonate chemistry. Several methods are discussed and analyzed based on their economic feasibility and potential environmental impact. It was found that several interesting methods and initiatives make use of alkaline waste streams that are reactive to CO$_2$, which is essential for their economically feasible conversion. The usage of steel-making slag to create cement-based construction materials was deemed to have significant potential and it was estimated that this could reduce the carbon footprint by 20 million tonnes of CO$_2$ per year. The utilization of CO$_2$ during the curing process of cement to enhance its properties was also deemed to have significant potential and it was estimated that this technology could reduce the carbon footprint by up to 140 million tonnes of CO$_2$ per year if all cement production in the world adopted this method.
1 Introduction

1.1 Climate change

Carbon dioxide (CO$_2$) is a colourless, odourless and mostly inert gas that naturally occurs in the earth’s atmosphere. It is also a combustion product of all fossil fuels and one of the most debated greenhouse gases. The greenhouse effect is caused by the selective transparency of gases in earth’s atmosphere: visible light passes almost completely but IR is largely absorbed. Because of the greenhouse effect, heat is trapped more effectively on earth. A higher concentration of greenhouse gases in the atmosphere increases this effect and this causes global warming. A graphical representation of the greenhouse effect can be seen in figure 1.

![Figure 1: An animation of the greenhouse effect. Visible/UV light enters the atmosphere where it is partly reflected by earth’s atmosphere and surface and it is partly absorbed as heat. This heat can irradiate via infrared radiation. The greenhouse gases absorb the infrared radiation and partly emit it back towards the surface, preventing much of the heat from radiating out into space.](image)

The total amount of anthropogenic carbon that was emitted globally in 2014 was 9.86Gt (= 9.86 * 10$^9$ tonne, = 36.135 * 10$^9$t CO$_2$) and these emissions have been increasing rapidly as can be seen in figure 2.[1] Since mankind has started burning fossil fuels to supply in its demand for energy, the concentration of CO$_2$ in the atmosphere has steadily increased from about 320 ppm in 1960 up to 400 ppm today.[2] A 2008 paper by Solomon et al. predicts that the changes in climate due to carbon dioxide emissions are largely irreversible over a period of 1,000 years[3]. These changes in climate could lead to higher temperatures, a rise in sea level, loss of biodiversity, more extreme weather events and the list goes on.[4] It is therefore of the utmost importance that the CO$_2$ concentration in our atmosphere is stabilized.

In 2015 the United Nations decided that mankind could not allow the global average
temperature to rise by more than two degrees Celsius above pre-industrial levels. To accomplish this it is important that CO\(_2\) emissions are brought to a halt and a switch to a CO\(_2\) neutral energy supply is made. The next 50 years most of the world will probably keep relying on coal for energy production. Even though there has been a huge rise in sustainable energy sources, additional efforts must be made to keep global warming below 2 degrees as is stated in the Paris agreement.

A measure that could help fix the problem could be to put a tax on CO\(_2\) emissions. A similar thing has been done for sulphur dioxide (SO\(_2\)) pollutions, which causes acid rain among other things. In 1990 the US government made some rigorous changes to the clean air act that established a SO\(_2\) allowance-trading system. A fixed amount of SO\(_2\) was allowed to be emitted and different companies could buy allowances to emit SO\(_2\). The acid rain program was a success and by 2007 the US managed to decrease their SO\(_2\) emissions by more than 9 million tonnes exceeding the initial goal.

Even though an allowance-trading system could in theory be a good regulation to combat the climate problem, it is much harder to implement because the problem is a lot bigger. A coal power plant emits both SO\(_2\) and CO\(_2\), but the amount of CO\(_2\) emitted by power plants is much higher than the amount of SO\(_2\). The concentration CO\(_2\) in coal power plant exhaust gases is 10-20% and the concentration SO\(_2\) is only approximately 0.5%.

One proposed solution to the climate problem is to capture the CO\(_2\) and store it underground; this is called carbon capture and storage (CCS). However, it has many drawbacks and it...
is still far from being applied on a large scale[11]. Technically it is possible to equip the major sources of pollution with CCS equipment, but this would come at a high cost. The additional costs and the lack of any short term benefits make it hard for people, businesses and countries that implement CCS to stay competitive. The cost of emitting CO$_2$ should weigh up to the cost of capturing CO$_2$, which is not the case at the moment.

### 1.2 How to make the most of carbon dioxide?

What if CO$_2$ is not treated as a pollutant that has to be stored deep underground, but as a useful raw material? Nature has been doing so for as long as there has been life on this planet. It should therefore be argued that describing CO$_2$ as just another pollutant is philosophically incorrect. We should instead look at CO$_2$ as one of the most abundant raw materials our planet has to offer. The XPRIZE foundation asked the following question: "How can we make the most of CO$_2$?"[12, 13]

The main issue with using CO$_2$ arises from thermodynamics. Chemically CO$_2$ is very inert, since it is the most stable oxidation state of carbon. The energy cost of separating the carbon atom from the oxygen atoms is very high and therefore turning CO$_2$ into more useful chemicals requires a high input of energy. When converting CO$_2$ into more useful chemicals, it is very important that the energy needed for such a process does not contribute to more CO$_2$ emissions. Therefore, when converting CO$_2$ it is important that the process is not very energy intensive or the necessary energy is derived from sustainable sources with a small carbon footprint.

The production of urea is right now the largest consumer of CO$_2$, accounting for about 60% of the total consumption of CO$_2$.[12] Urea is a nitrogen containing substance that is mostly used as a synthetic fertilizer. First liquid ammonia and gaseous CO$_2$ react at high pressure and temperature to form ammonium carbamate, as can be seen in equation 1a.[14] Then the ammonium carbamate undergoes a decomposition reaction into water and urea, as can be seen in equation 1b. The problem here is that the ammonia used in this reaction is made using fossil fuels and the entire urea synthesis requires a lot of energy. This makes the urea industry a net source of CO$_2$ emissions.

\[
\begin{align*}
2\text{NH}_3 + \text{CO}_2 & \rightleftharpoons \text{H}_2\text{N}--\text{COONH}_4 & (1a) \\
\text{H}_2\text{N}--\text{COONH}_4 & \rightleftharpoons (\text{NH}_2)_2\text{CO} + \text{H}_2\text{O} & (1b)
\end{align*}
\]

A large issue with utilizing CO$_2$ in our atmosphere is that it is very dilute. The minimum amount of energy required to capture CO$_2$ directly from the air is provided by thermodynamics. This minimum energy required to capture and concentrate CO$_2$ is about 0.44 GJ/tCO$_2$. If the energy cost of compressing it to 100 atm, the pressure needed for pipeline transport, is also added the total energy required becomes approximately 1.1 GJ/t CO$_2$.[15] To put this energy requirement into perspective: the best type of coal has an energy density of
32 GJ/t and thus a minimum of 34 kg of coal needs to be burned to capture 1 tonne of CO₂. Burning this much coal will produce another 125 kg of CO₂.

The authors that made the calculations for the minimum energy requirements of direct air capture also estimate that it would be possible to do so for 500$/t CO₂ or less.[15] The price of the European Emission Allowances, a permit that allows the user to emit a given amount of CO₂, can be seen as the cost of emitting CO₂ in the European Union. While this price has steadily climbed the last two years, it is still only about €13/t CO₂ which is a stark contrast with the cost of capturing the CO₂.[16] This indicates that it is much more beneficial to capture CO₂ at the source of emission instead of capturing it directly from the air.

Since the energy requirement to separate the carbon and oxygen atoms in CO₂ is so high, another solution is to create a product where this is not necessary. Mineral carbonation, the process of fixating CO₂ as stable carbonate mineral, is such a process. One example of a company that does this process commercially is Calera. They create calcium carbonate (CaCO₃) from the CO₂ in flue gasses from industrial emitters, calcium and a source of alkalinity. They then use the CaCO₃ to create a range of useful building material products.

1.3 Carbon XPRIZE

On 29 September 2015, the XPRIZE Foundation announced a competition with a prize purse of $20 million that challenges research teams to come up with the best way to turn CO₂ into a useful material.[12] The teams are scored on how much CO₂ they can convert and on the net value of their products. Three of the ten finalists in the XPRIZE competition are focusing on utilizing CO₂ in cement or concrete production. The three teams are: CarbiCrete a Canadian-based team that aims to use industrial waste from the steel-making industry and CO₂ to produce concrete blocks, CarbonCure also a Canadian-based team that aims retrofit the current cement industry to incorporate CO₂ in cement production and lastly Carbon Upcycling UCLA a team from California that aims to create Lego-like bricks from hydrated lime (Ca(OH)₂) and CO₂. The fact that three of the ten finalists of the carbon XPRIZE competition aim to utilize CO₂ in cement-based products is a good indication that there is a lot of potential for CO₂ usage and emissions reduction in this industry.

The aim of this report will be to assess the potential of utilizing CO₂ in the production of cement-based products. Different methods of utilizing CO₂ in different kinds of products will be discussed and methods that show potential for large-scale application are analyzed. The methods will be rated on the cost and the value of the created products, but also on the maximum environmental impact that the method can have.
2 Cement and concrete industry outlined

The global production of cement has grown very rapidly over the past decade and it is the third largest source of man-made CO$_2$ emissions. The CO$_2$ emissions of the cement industry in 2016 were 1.45 ± 0.20 Gt CO$_2$[17], which is roughly 5% of the total CO$_2$ emission.[18]

The production of cement is estimated to keep increasing, especially in developing countries as their economies grow. One article by Global Cement defines a trend between a growth in gross domestic product and an increase in cement consumption.[19] Therefore, when more nations in the world become industrialized, cement consumption will also increase. The cement technology roadmap from the international energy agency (IEA) estimates that cement production will grow to 4.40 billion tonnes in 2050.[20] It is necessary to recognise the urgency to identify new methods to reduce CO$_2$ emissions or find usage for CO$_2$ in the cement production.

Before discussing the possibilities of utilizing CO$_2$ in the cement and concrete production, a brief overview of the cement industry is given and some key steps in cement production are explained.

2.1 Cement production explained

Cement is a binder used in construction that, when mixed with water and aggregates, forms concrete. While any substance that is used as a binder in construction can be called cement, cements commonly used in construction are of an inorganic nature and are usually based on calcium silicate (CaSiO$_4$) and/or lime (CaO). These cements are either called hydraulic when they can set in the presence of water or non-hydraulic when they can not. The non-hydraulic cements set as they dry and undergo a reaction with CO$_2$ and the hydraulic cements set because of a series of hydration reactions between the water and the cement. Almost all modern cements are either Portland cement or a Portland cement blend. Portland cement can be categorized as a hydraulic cement.

The cement technology roadmap by the IEA[20] splits cement production in ten steps, which are shown in figure 3. The raw materials for Portland cement are limestone, which contains CaCO$_3$ and to a lesser degree clay-like materials which provide aluminosilicate minerals. In step 1 the raw materials are mined from a quarry and in steps 2 and 3 they are crushed and ground to a homogeneous mixture. In steps 4 and 5 the mixture of raw materials is heated to a temperature of approximately 600 °C. At this temperature CO$_2$ is driven out of the minerals, as can be seen in equation 2.

\[
\text{CaCO}_3 \xrightarrow{\Delta} \text{CaO} + \text{CO}_2
\]

During step 6 the mixture is heated even further to a temperature of approximately 1450 °C, which partially fuses the material together. The resulting lumps of fused together minerals are called clinkers. The clinkers are stored and cooled during step 7 of the
process. In step 8 other additives are added to the clinkers to achieve the right properties of the finished cement. Finally in step 9, the mixture is ground finely to achieve the finished cement powder.

Figure 3: The cement manufacture process in ten steps.[20]

Most of the CO$_2$ emissions comes from the calcination process in steps 4 and 5. During this process the CO$_2$ trapped in the limestone is released. Generally 60-65% of the total emissions comes from this chemical decomposition.[20] These CO$_2$ emissions cannot be prevented and are inherent to the production of clinkers for Portland cement. This is one of the reasons that CCS is necessary to lower the CO$_2$ emissions of the concrete industry. Almost all other emissions are generated by fuel combustion and electricity usage.

The finished product, Portland cement, consists roughly for 66% of CaO, 20% SiO$_2$ and the rest is varying degrees of iron, aluminium and sulfur oxides. These constituents form several different mineral phases in the cement clinker. The important mineral phases in Portland cement are: tricalcium silicate (also called alite, (CaO)$_3$·SiO$_2$ and accounts for 45 – 75%), dicalcium silicate (also called belite, (CaO)$_2$·SiO$_2$ and accounts for 7 – 32%), tricalcium aluminate ((CaO)$_3$·Al$_2$O$_3$ and accounts for 0 – 13%), calcium aluminoferrite ((CaO)$_4$·Al$_2$O$_3$·Fe$_2$O$_3$ and accounts for 0 – 18%) and calcium sulfate dihydrate (also called gypsum, CaSO$_4$·2H$_2$O which accounts for 2–10%). Alite, is the mineral phase that defines Portland cement from other hydraulic lime containing cements. Alite is formed in the cement kiln at temperatures above 1300 °C when belite fuses with CaO.
2.2 Areas for CO₂ emission reduction or CO₂ usage

The cement technology roadmap by the IEA[20] discusses 4 areas where CO₂ emissions can be reduced: thermal and electric efficiency; alternative fuel usage; clinker substitution; and finally application of CCS during cement production. The developments in these areas are further explained in an article by Schneider et al.[21]

Technologies in the area of thermal and electrical efficiency have matured almost completely and therefore there are not any developments that could contribute significantly to the reduction of CO₂ emissions. Market and economic forces have driven these developments and have already forced many cement companies to improve their own efficiencies to reduce costs.

There are also not any significant new technologies in the area of fuel usage. At the moment coal forms a big part of the fuel source and this inevitably causes a lot of CO₂ emissions, but with the right incentives and regulations companies will convert their fuel usage to natural gas or electricity. Changes in this area will be driven by market and economic forces primarily.

Alternative raw materials or a lower cement:clinker ratio is the third area where CO₂ emissions can be reduced. The cement:clinker ratio states how much of the cement is comprised of clinkers and how much is comprised of alternative additives. Since clinkers are very energy intensive to produce it is beneficial if they can be replaced by other materials. Different kinds of ashes and slags are used for this purpose, for example: steel-making slag[22], waste incineration ash[23], sawdust ash[24], rice husk ash[25], natural volcanic ash and many other solid combustion products are used for this purpose. Section 3 gives some insight in the properties and origin of some important industrial wastes that are discussed in this report. At the moment, utilization of wastes is limited due to strict product quality, regulations and availability of raw materials.

It is also possible to replace Portland cement entirely. Juenger et al. discuss 4 alternative binders for usage in concrete in their paper[26]: Calcium aluminate cement, calcium sulfoaluminate cement, alkali-activated binders and supersulfated cement. Another example is cement made by Solidia Tech, which consists mostly of calcium silicate (CaSiO₄)[27], which is described further in section 5.4.4. All these binders have distinct advantages and disadvantages compared to regular Portland cement. Especially calcium sulfoaluminate cement and alkali-activated cement have a much smaller carbon footprint compared to Portland cement. While the understanding about these alternative binders has improved in recent years, they are still only being applied for niche applications. The main drawback these binders have is that at the moment they have a higher cost than regular Portland cement.

At the moment CCS is not employed in the cement industry on a large scale. However, in order to reduce emissions even further it is necessary to start incorporating CCS in the cement industry. Improvements in the other three areas mentioned can drive down the CO₂ emissions, but CCS will be needed to achieve bigger emission reductions. According
<table>
<thead>
<tr>
<th>Cement usage</th>
<th>Percentage of total cement used</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ready-mix concrete</td>
<td>71.0 %</td>
</tr>
<tr>
<td>Concrete products</td>
<td>13.2 %</td>
</tr>
<tr>
<td>Masonry cement</td>
<td>4.2 %</td>
</tr>
<tr>
<td>Oil, Mining and Waste</td>
<td>1.3 %</td>
</tr>
<tr>
<td>Others/unknown</td>
<td>10.3 %</td>
</tr>
</tbody>
</table>

Table 1: Distribution of cement usage in the US market of 2003.\[30\]

to a report by the global CCS institute\[28\], oxyfuel or post-combustion capture techniques are the preferred CCS methods for cement production as these also allow the capture of all the CO\(_2\) that is released during the calcination process.

### 2.3 Cement market

Earlier in this report it was said that the production of cement causes approximately 5% of the total CO\(_2\) emissions in the world, which is a good indicator that the cement market is very large. A publication by the U.S. Geological Survey\[29\] estimates that the total production of cement worldwide was 4.1 billion tonnes in 2017, or \(4.1 \times 10^9\) t. Of which more than half was produced in China.

Although it is difficult to find accurate and recent data on the end-usage of cement, an estimation will be made. The end usage of cement can be classified in four categories. Firstly, most cement is used to make ready-mix concrete. Ready-mix concrete is made in a batch plant and then either delivered by mixer trucks or it is mixed in on site mixers. Most concrete we see in our daily life was probably made with ready-mix concrete, since its applications are many. Secondly, precast concrete products are also a big cement user. Precast concrete products are products that are cast in a mould in a controlled environment and then sold to an end-user. Thirdly, masonry cement is used to bind building bricks together. Finally, there are many other end users of cement such as oil wells, mining, waste stabilization, and many more.

The only freely available statistics that were found on end-usage of cement where from the U.S. Geological Survey\[30\] and they only had data available until 2003 for the United States. These statistics can be seen in table 1. These statistics will be used to estimate cement usage worldwide, but a few assumptions have to be made. Firstly it was assumed that the distribution of the different kinds of end-usage are comparable with the cement usage in the rest of the world. Secondly, it was assumed that the distribution has remained the same even after 15 years. Finally, it was assumed that the growth in cement usage of the last 15 years can be distributed proportionally over the different end users. Even though these assumptions are far from exact, they are probably good enough for rough estimations.
### Table 2: Common compressive strength values for typical rock-like construction materials.\[33\]

<table>
<thead>
<tr>
<th>Material</th>
<th>Compressive strength (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bricks</td>
<td>7-80</td>
</tr>
<tr>
<td>Brickwork</td>
<td>7-14</td>
</tr>
<tr>
<td>Granite</td>
<td>130</td>
</tr>
<tr>
<td>Limestone</td>
<td>60</td>
</tr>
<tr>
<td>Portland cement less than one month old</td>
<td>14</td>
</tr>
<tr>
<td>Portland cement more than one year old</td>
<td>21</td>
</tr>
<tr>
<td>Portland concrete 28 days old</td>
<td>35</td>
</tr>
<tr>
<td>Portland concrete more than one year old</td>
<td>43</td>
</tr>
</tbody>
</table>

#### 2.4 Producing concrete

As can be seen in table 1, most cement is used to produce concrete. Concrete is an artificial rock-like building material created by mixing wet cement with coarse and fine aggregates and allowing the cement to set and bind the aggregates. Concrete is the world’s most common man-made material and worldwide almost 1 cubic meter of concrete is consumed per person per year.\[31\] Different kinds of cement (e.g. Portland cement, bitumen or organic polymers) and different kinds of aggregates (e.g. sand, gravel crushed rock or recycled materials) can be used to obtain concrete with desired properties.

Generally concrete consists of 60-75% aggregates, 7-15% cement, 14-21% water and 0-8% air (depending on the porosity). It can be derived from this that the worldwide market for construction aggregates is even larger than the cement market. According to a demand and sales forecast done by the Freedonia Group, the world demand for construction aggregates will rise to 51.7 billion tonnes in 2019.\[32\]

#### 2.4.1 Properties of concrete and concrete products

Throughout the rest of this report the quality of created products are most often denoted in the compressive strength of the product. Compressive strength is a value that states how much force can be applied to a material or how much load the material can bear before it cracks or is deformed and in this report it is denoted in \(MPa\); \(1MPa = 10^6Pa = 10^6 \frac{N}{m^2}\). The compressive strength is an important property of construction material because it determines for what purpose the material can be used and how much the material is worth. Some common compressive strength values of building materials are listed in table 2.
3 Industrial wastes for CO$_2$ sequestration

Many of the methods described in the upcoming section for CO$_2$ sequestration make use of alkaline industrial waste streams that contain high levels of calcium or magnesium oxides. These industrial waste streams include among other things ashes, sludges and slags from the steel-making industry and the electric power industry. Because of the high level of CaO and MgO these wastes also exhibit cementitious properties to some degree. Therefore many of them are also used as supplementary cementitious materials and can partially replace the energy intensive clinkers in cement. In this section the properties and availability of these wastes will be discussed.

3.1 Industrial wastes from the steel industry

The global steel production is 1.6 billion tonnes per year.[34] For each tonne of steel that is produced 200-400 kg of solid waste is formed as a by-product. The properties of this solid waste depends on the method of producing steel and the source of the raw-materials that are used. The first main route to create steel consists of creating iron in a blast furnace (BF) and then creating steel in a basic oxygen furnace (BOF). The other route to create steel is to first create direct-reduced iron (DRI) and then create steel in an electric arc furnace (EAF). The BF/BOF route is primarily used to create steel directly from raw iron ore. An EAF is primarily used to recycle scrap metal, but it can also be used to create steel from DRI.

Iron-making and steel-making slags are one of the major by-products of the steel production with the BF/BOF route, but some dusts and sludges are also produced. These slags are a mixture of inorganic components such as silica, calcium oxide, magnesium oxide, aluminium oxides and iron oxides. More than 400 million tonnes of slags are produced each year as a by product. With the BF/BOF route 275 kg iron-making slag is produced in the BF and 125 kg steel-making slag is produced in the BOF.[35] From this it can be concluded that 125 * $10^6$ tonnes of steel-making slag and 275 * $10^6$ iron-making slag is produced each year.

Iron-making slag is already being utilized for nearly 100%, but steel-making slag is only being utilized for approximately 80%.[35] Therefore, it is interesting to investigate how the remaining 20% of steel-making slags that are produced can be utilized and this is exactly what a lot of the articles that are referenced in this report are doing. A total of 20% * 125 * $10^6$ = 25 * $10^6$ tonnes of steel-making slag currently goes to waste. It is hard to recycle steel-making slag due to the high concentrations of calcium oxide and magnesium oxide, which give rise to volume instability and expansive effects.[36] These same high levels of calcium and magnesium oxides make this waste very reactive to CO$_2$.

Huijgen et al. studied the usage of steel-making slag to sequester CO$_2$ and form minerals.[37] They concluded that steel-making slag is a promising waste for CO$_2$ sequestration, because it is cheap since it is considered a waste stream of the steel industry, it is
available near large point sources of CO$_2$ and it reacts easily with CO$_2$. They found that the maximum CO$_2$ uptake capacity of steel slag is about 0.25 kg of CO$_2$ per kg of steel slag on the basis of Ca content in steel slag. They managed to achieve a CO$_2$ uptake of 74% of the maximum uptake capacity under ideal circumstances. These circumstances did require a high CO$_2$ pressure, finely ground steel-making slag particles and 100°C temperature.

Both steel-making slag and iron-making slag are already being utilized for purposes such as construction, fertilizers and metallurgy, it is therefore not a surprise that these wastes still have some value. The cost of steel-making slag will likely depend on the quality and the exact composition. Mahoutian and Shao state in their article that they bought steel-making slag with a high concentration of CaO for $3/tonne from a Canadian steel-making plant[38], which gives a good estimation for the sales value of steel-making slag.

### 3.2 Incineration ashes

Many different kinds of ashes are described to be useful in cement production and they are often reactive towards CO$_2$. Waste incineration ash[23], sawdust ash[24] and rice husk ash[25] are only three examples of many. The most interesting ashes are those produced in large quantities and that are reactive to CO$_2$.

During combustion generally two kinds of ash are distinguished: bottom ash is the non-combustible ash that is leftover in the bottom of the incinerator and fly ash is the ash that is captured from the exhaust of the incinerator. The exact composition of these ashes is highly dependent on the source and type of fuel that is being burned. In general most ashes contain varying concentrations of SiO$_2$, Al$_2$O$_3$, Fe$_2$O$_3$, MgO and CaO.

The ability to sequester CO$_2$ with fly ash has been demonstrated[39], but in their experiment a tonne of fly ash was only able to sequester 26 kg of CO$_2$. Fly ash is also used as an additive in concrete and it can be transformed to aggregates[40, 41], but when they are used as aggregates or additive it does not exhibit the ability to sequester CO$_2$. 

13
4 Carbonate chemistry

The methods that are discussed in this section try to incorporate CO$_2$ in building materials by CO$_2$ mineralization, which creates carbonated minerals. When creating carbonated minerals it is important to know about the chemistry behind these reactions. This section will give a quick summary of carbonate chemistry. The carbonation reaction can create pure carbonates, solid minerals large enough to be used in construction as aggregates or it can solidify cementitious materials.

Carbonation can occur either with gaseous CO$_2$ or with aqueous CO$_2$. Many of the carbonation reactions discussed in this report occur in water. The chemical equations accompanying the dissolution of CO$_2$ in water are shown in equation 3. All these reactions are in equilibrium and this equilibrium is highly dependent on the pH of the solution.

First CO$_2$ is dissolved in the water, as seen in equation 3a. The amount of CO$_2$ that gets dissolved in the water is dependent on the partial pressure of the CO$_2$ and the temperature. A higher partial pressure will dissolve more CO$_2$ and a higher temperature will dissolve less CO$_2$. Although it is possible for some of the methods discussed to use CO$_2$ directly from the atmosphere, the carbonation reaction occurs much faster when a CO$_2$ supply with a much higher partial pressure of is used. This is one of the reasons that flue gases from large industrial processes are most often taken as an example of a suitable CO$_2$ source.

Next the dissolved CO$_2$ can react with water to form carbonic acid, as seen in equation 3b. This molecule is unstable and will quickly lose a proton to form the bicarbonate ion, as seen in equation 3c. Alternatively the carbonic acid molecule can also revert back to dissolved CO$_2$, which is a more stable state than the carbonic acid molecule. The bicarbonate anion can lose one more proton to form the carbonate anion, as is seen in equation 3d.

Figure 4 shows a Bjerrum plot of a system with water and dissolved CO$_2$. This plot shows how the ratio between dissolved CO$_2$, bicarbonate and carbonate are highly dependent on the pH of the solution. These curves are not only dependent on the pH and will also vary with temperature and the concentration of other dissolved ions.

It is possible for these carbonate or bicarbonate ions to precipitate out of the solution when combined with the right cation and under the right reaction conditions. Equation 4 shows how CaCO$_3$ can precipitate when calcium ions are present. The same reactions can occur with some other divalent cations like magnesium.
Figure 4: A Bjerrum plot for a carbonate system. The relative ratios of the different species are plotted on the y-axis. On the x-axis the change in pH is plotted.

\[ \text{Ca}^{2+}(\text{aq}) + \text{HCO}_3^- (\text{aq}) \rightleftharpoons \text{CaCO}_3(s) + \text{H}^+(\text{aq}) \]  \hspace{1cm} (4a)

\[ \text{Ca}^{2+}(\text{aq}) + \text{CO}_3^{2-} (\text{aq}) \rightleftharpoons \text{CaCO}_3(s) \]  \hspace{1cm} (4b)

The effect of temperature on the rate of carbonation is not as straightforward as one might expect. At higher temperatures calcium ions and other components from the cement dissolve more easily and the CO\textsubscript{2} gas diffuses faster in the cement, but CO\textsubscript{2} solubility decreases with an increase in temperature. That is why each carbonation reaction has a maximum reaction rate at a temperature that is determined by the solubility of the involved components.[42] For example, Liu et al. studied the reaction of CO\textsubscript{2} with concrete waste materials and found that the carbonation reaction at atmospheric pressure was enhanced until a temperature of 60°C after which the reaction rate starts declining at higher temperatures.[43]

The partial pressure of CO\textsubscript{2} is also an important factor in the carbonation reaction. A higher CO\textsubscript{2} partial pressure allows more CO\textsubscript{2} to be dissolved and thus the carbonation reaction rate is higher. In a porous medium, like concrete, the carbonation reaction slowly reduces the porosity due to the precipitating CaCO\textsubscript{3} filling the pores. A higher CO\textsubscript{2} partial pressure allows for more CO\textsubscript{2} to be dissolved in the pores before they are closed and can increase the maximum degree of carbonation in this way.[42]

When solid materials react with CO\textsubscript{2}, there are a number of factors that determine the carbonation rate and the total extent of carbonation. These factors include humidity/water content, particle size, porosity of the particles, but also temperature, CO\textsubscript{2} partial pressure and mineral content described above.
The water content of the minerals or the humidity during the reaction is an important factor for the carbonation reaction of solid minerals, because the diffusion of CO$_2$ in water is much slower than in air. When the humidity or the water content is too high and the minerals are saturated with water, the rate of carbonation is limited by the slower diffusion of CO$_2$ to the reactive mineral. However, water also plays a crucial role in dissolving the reactive part of the mineral such as the calcium hydroxide or magnesium hydroxide.

The particle size of the minerals is another important factor for the rate of carbonation. Larger particles will have a smaller surface area for the CO$_2$ to react with the mineral. Furthermore, for larger particles the total extent of carbonation is often smaller because not all of the reactive mineral can be dissolved.

The porosity of the particles is also an important factor for the carbonation reaction. A higher porosity allows for more CO$_2$ to diffuse throughout the particle and react with the active minerals. The total extent of carbonation is often decreased when the pores are closed due to precipitated carbonated minerals. This reduction in porosity due to precipitate carbonated minerals is also an important mechanism how carbonation can increase the compressive strength of materials.

5 CO$_2$ utilization in cement-based materials

Currently utilization of CO$_2$ in cement-based materials is a hot topic. Integrating CO$_2$ in the most widely used building material sounds like an ideal solution to contribute to mitigating the CO$_2$ emissions problem. In this section different methods under development or in use that utilize CO$_2$ in cement-based materials are discussed and explained. For each method the production process and the properties of the created product will be discussed. The methods have to meet the following criteria: the process must produce a cement-based construction material, it must utilize CO$_2$ in the process, it must be applicable on a large scale and it must be economically viable.

To abide by these criteria more easily this report focuses on methods that are already being applied on an industrial scale. The initiatives that are among the finalists of the carbon XPRIZE are considered especially interesting. Some additional interesting initiatives that were considered out of the scope of this report or for which not enough information was found are discussed at the end of this section.

5.1 Blue Planet aggregates

This section explores the method used by a company called Blue Planet which can be used to create aggregates using CO$_2$ and hereby sequestering CO$_2$ inside these aggregates. Blue Planet is a company that is exploring the use of CO$_2$ to produce carbonated rocks that can then be used as aggregates. Blue planet has a patent that describes their methods of
creating different kinds of aggregates with CO$_2$.[44] In this patent they claim that their method is capable of producing aggregates with a wide variety of shapes, surface textures, hardness, chemical resistance, density, porosity and reactivity.

The method described by Blue Planet in their patent creates aggregates by dissolving CO$_2$ in an aqueous solution that contains divalent cations and a base. CO$_2$ forms carbonates and bicarbonates when dissolved in water. These precipitate in combination with divalent cations. Sufficient base is necessary to keep the pH high enough that the dissolved CO$_2$ takes either the carbonate form or the bicarbonate form, as is explained in section 4. In an example of the Blue Planet method described in their patent[44], they added base to the reaction mixture until the pH reached 9.5. Additional details about carbonate chemistry are explained in section 4. The resulting precipitate is washed and dewatered and processed in such a manner that the minerals form solid aggregates. The formed aggregates primarily consists of different hydrated carbonate minerals.

This last processing step could require a lot of additional energy. In the examples stated in the patent by Blue Planet[44] they mention a multitude of methods that are combined to form the precipitated mass into solid aggregates. These methods include filter pressing, using a heated hydraulic press, oven drying, washing with water, compression moulding and spray drying. The processing steps must either be kept as cheap as possible or the resulting product must make up for it in quality.

Although this method is a promising way to capture CO$_2$ and create a useful product at the same time, it does require a lot of added base and a source of calcium or magnesium to precipitate the carbonated minerals. One way to overcome this problem is to use a waste stream that contains both.

5.2 CarbiCrete: steel-making slag blocks

Carbicrete is a Montreal based company that aims to create construction blocks from steel-making slag. CarbiCrete is the first team of the three XPRIZE finalist discussed in the introduction of this report that aims to utilize and sequester CO$_2$ in cement-based products. They have developed a method to create cement-free construction blocks from steel-making slag with a negative carbon footprint.[38]

To create their construction blocks they use a mixture of both EAF slag and BOF slag with a high CaO content (39%). This slag mixture is waste from a Canadian steel plant and was originally destined to be landfilled. They use aggregates from expanded blast furnace slag, which is often used as aggregate in lightweight concrete.[45] These aggregates were probably chosen to show that the blocks can be made using only industrial waste, but other kinds of aggregates are probably also possible. The aggregates, water and the BOF/EAF slag mixture were moulded into construction blocks with a 12 MPa steel mould and then dried in front of a fan. The mixture consisted of 1555 kg/m$^3$ steel slag mixture, 930 kg/m$^3$ slag aggregates and it had a water to slag ratio of 0.2 (water/slag=0.2). The blocks were then carbonated for 24 hours at a CO$_2$ partial pressure of 1.5 bar and
afterwards stored for 35 days in plastic bags to allow subsequent hydration reactions. The created construction blocks achieved a compressive strength of 35.9 MPa. These slag bonded blocks outperformed regular commercial concrete blocks in both compressive strength tests as in durability tests, which makes them qualitatively competitive.

5.2.1 Aggregates from steel-making slag

A recent paper describes a method to produce artificial aggregates from steel-making slag and carbon dioxide.[46] This method is considered complimentary to the CarbiCrete method to create blocks from steel-making slag, because it uses the same raw material with a similar process to create a different type of product.

These artificial aggregates described in the article are created by sieving, milling, moistening and pressing steel-making slag into a cylinder. These cylinders are then carbonated in a chamber with CO$_2$ and water vapour. The cured cylinders were crushed and the crushed aggregates were exposed to CO$_2$ again. The artificial aggregates performed well compared to natural granite.

It was shown that the artificial aggregates could be used to create regular concrete blocks. These concrete blocks were allowed to absorb CO$_2$ once more to increase the total uptake of CO$_2$. A 145 g sample of concrete had a total CO$_2$ uptake of 24 g, of which 20 g was absorbed by the aggregates and the rest was absorbed by the added Portland cement. The article[46] showed with a calculation that a single masonry block (20×20×40 cm) weighing 17.9 kg had a net CO$_2$ balance of -0.68 kg. Because the uptake of CO$_2$ outweighed the total CO$_2$ emitted in the process, they conclude that carbon-negative concrete blocks could be made using their artificial aggregates.

5.3 CarbonCure concrete

The curing process is a process where the cement undergoes a series of reactions that lead to the hardening and solidification of the cement. Hydraulic cements, like Portland cement, cure via a series of hydration reactions. The minerals in the cement clinker undergo a hydration reaction binding them to each other and solidifying them. The interlocked mineral hydrates give rise to the strength of the cement. Non-hydraulic cements cure via a reaction with CO$_2$ in the air. Using CO$_2$ in the curing stage of cement-based products is the focus of this section.

It is conventionally known that the reaction of CO$_2$ with hardened concrete causes durability issues due to effects such as shrinkage, carbonation induced corrosion and a reduced pH of the pore solution. However, during the curing stage it is possible to treat cement with CO$_2$ without loss of durability and in this way upcycle some CO$_2$ into the concrete products. A 2013 paper by Zhan et al. showed that concrete blocks that were CO$_2$ cured had a higher compressive strength than corresponding concrete blocks that were moisture cured[47]. The goal of Zhan’s research was to investigate if CO$_2$ curing
can improve the properties of concrete blocks containing recycled aggregates. Recycling aggregates is of major importance in China right now, because their construction waste has taken enormous proportions.[48] For the CO$_2$ curing process they placed the concrete blocks inside a controlled chamber with a 100% CO$_2$ atmosphere.

While the goal of the research done by Zhan was to find a way to use recycled aggregates and maintain the desired mechanical strength, in actuality they confirmed a theory that has been around since 1972. Klemm and Berger discovered in 1972 that applying CO$_2$ to wet cement can accelerate the curing process and even give rise to an increase in mechanical strength.[49] Recent research has provided even more evidence that applying CO$_2$ to drying cement can increase the mechanical strength and reduce shrinkage of the finished concrete.[50]

A company called CarbonCure is already applying this principle on a large scale to reduce the curing time of concrete and reduce the amount of cement needed to maintain the desired mechanical strength of the finished product. The method CarbonCure applies is to treat concrete with CO$_2$ gas while it is still in a loose state before it is placed into a mould. Their patent states[51]:

\[\text{The addition of carbon dioxide may promote an alternate set of chemical reactions in the concrete resulting in different reaction products. In particular, thermodynamically stable calcium carbonate (limestone) solids may be formed preferentially to calcium hydroxide (portlandite) products. The carbon dioxide may be solvated, hydrated and ionized in water in the concrete to produce carbonate ions. These ions may combine with calcium ions from the cement to precipitate calcium carbonate in addition to amorphous calcium silicates. In this way, carbon dioxide may be sequestered in the concrete blocks as a solid mineral. Excess gas, if any, may be vented away from the treated concrete mass. Otherwise, the production cycle of a given concrete product may remain generally unchanged.}\]

While the CarbonCure patent quoted above states that the addition of CO$_2$ to the wet concrete has some benefits and allows for sequestration of CO$_2$ in the concrete, they do little to back these claims up in their patent. CarbonCure does show a mechanical strength comparison between concrete that had CO$_2$ added and concrete that did not. The control concrete had a mean compressive strength of 9.44 ± 1.23 MPa and the CarbonCure concrete had a mean compressive strength of 10.95 ± 1.35 MPa. This shows that the carbonated concrete was stronger on average than the control sample, but since the two regions of compressive strength overlap it does not exclude the possibility that regular concrete can be stronger than CarbonCure concrete.

To explain the increase in strength and decrease in setting time after addition of CO$_2$ to the wet cement, it is important to know how the CO$_2$ reacts with cement. The gaseous CO$_2$ reacts with calcium oxides in the cement to form finely distributed CaCO$_3$ throughout the cement. A 2010 article by Sato et al. explains that the addition of finely ground CaCO$_3$ to cement can increase the overall strength and shorten the setting
time.[52] But these positive effects only occur when the CaCO$_3$ is evenly distributed and ground to nanometer-size particles. Sato suggested that the nano-CaCO$_3$ acts as nucleation sites for the hydration reaction of the cement. Finally, Sato concludes that it would be too expensive to add nano-CaCO$_3$.

The results of a 2016 article of Monkman et al.[53] combined with the results of Sato[52] described above explain how CO$_2$ can have a positive effect on concrete. Monkman describes how the reaction between CO$_2$ and the wet cement creates finely distributed CaCO$_3$ in the cement. Since the price of CO$_2$ is very low, adding CO$_2$ is cost-effective, compared to adding nano CaCO$_3$ particles. In this article Monkman mainly views CO$_2$ as an additive that can be added to the cement to reduce setting time and increase early strength.

This scientific article by Monkman does conflict slightly with the details of the patent by CarbonCure regarding the amount of CO$_2$ that can be sequestered in the cement. The CarbonCure patent[51] states that: ”the carbon dioxide uptake may be a significant portion of the theoretical maximum uptake, which for conventional cement may be approximately half of the mass of the cement in the mixture.” However, Monkman only added a maximum of 1.5% CO$_2$ by weight of cement and they stated that the CO$_2$ was absorbed into the concrete with an efficiency of about 88%.[53, 54]. Therefore, it is assumed that most of the CO$_2$ used in the CarbonCure process is not absorbed into the final product, contrary to what the patent states.

5.4 Some additional initiatives worth mentioning

This section will highlight some additional initiatives that could either help lower the carbon footprint of cement-based materials, incorporate CO$_2$ in a novel way or both. The methods below were not analyzed thoroughly, because they either fell outside the scope of this report or their development was not far enough that a complete analysis would make sense. Nevertheless, these methods are worth mentioning and clearly show that the field of CO$_2$ utilization in cement-based materials is very broad and constantly developing.

5.4.1 CO$_2$NCRETE: Bricks from carbonated lime

The third and final XPRIZE finalist that will be discussed in this report is the Carbon Upcycling team from the University of California — Los Angeles. They are developing a product called CO$_2$NCRETE that is based on the carbonation reaction of hydrated lime. For their method they mix water and CaO and print it in the desired shape with a 3D-printer. The CO$_2$ comes from flue gases from an industrial emitter. The flue gases are treated in the following order: the gas is diverted, heat is captured, the gases are dehumidified, the gas is filtered, enriched with CO$_2$ and the gas is used to carbonate the 3D-printed bricks.[55]
This process is very interesting because it captures CO₂ and converts it into a useful construction material at the same time. The created CaCO₃ bricks add value to the CO₂ capture process. However, no official patents, reports or articles were found from the developers that explain their invention thoroughly. Because it was not proven that this method can be applied commercially on a large scale, this method is not analyzed further in this report.

5.4.2 Carbon8 aggregates from industrial waste

Carbon8 Aggregates Limited is a British company that has developed a method where they use accelerated carbonation technology to create carbon-negative aggregates from industrial wastes. They won a 7 year contract to recycle the waste incineration ashes into aggregates from a Worcestershire based waste incineration facility.[56]

To produce the aggregates they grind the wastes, add some water, blend them together, create pellets from the mixture in a mould and the pellets are then carbonated in a carbonation chamber for 10 or 20 minutes. In the patent describing their invention[57] they mention that other industrial wastes (e.g. cement bypass dust, biomass ash or steel slag) can also be used. In some of the examples it is stated that they add Portland cement to the mixture to increase the strength of the finished aggregates.

The compressive strength values that were stated for the different kinds of aggregates were in the range of 1.8 - 8.2 MPa, which is quite low compared to other construction materials listed in table 2. Even though Portland cement is known to achieve much higher compressive strength values over more time, a pure Portland cement pellet achieved only 2 MPa compressive strength after 20 minutes of carbonation in the tests done in this patent. This might indicate that the aggregates can achieve a higher compressive strength given a longer curing time and the compressive strength values produced by the tests in their patent are not comparable to the strength of the finished aggregates.

Although this company is successful in converting industrial waste and CO₂ into a useful product, not enough details of their method are known to allow for a complete analysis. However, it is still a good example that proves not only steel-making slag, but also waste-incineration ashes can be converted into aggregates. Apparently this can be done in an economically viable method, since a company is doing it in Great Britain.

5.4.3 Hydrothermal synthesis of CO₂-stored cementitious material

A South Korean research group developed a novel method to create an alkali-activated CO₂-stored cementitious material.[58] The raw materials they use are CaCO₃, sodium aluminate (NaAlO₂), silica fume (micro SiO₂), triethanolamine and sodium hydroxide (NaOH). The novelty of their research is that they managed to create a cementitious material using CaCO₃ as a raw material without releasing the CO₂. They did this by developing a hydrothermal synthesis, which was done at ambient conditions. Because
of the mild conditions the CaCO$_3$ did not decompose and the CO$_2$ was trapped in the final product. They showed that their synthesized cement could reach similar strengths or higher than ordinary Portland cement.

This method shows that a hydrothermal synthesis route can be an interesting alternative to the high-temperature clinkering method. Although the produced cement showed promising physical properties and it had a greatly reduced carbon footprint, the consumption of expensive raw materials will likely make this method to costly to implement. This method was not analyzed further, because it was not proven this method can be applied commercially on a larger scale.

5.4.4 Solidia Tech cement

An example of cement with a reduced carbon footprint is calcium silicate-based cement and Solidia Tech is a company that is commercializing it. The Solidia cement has a reduced amount of CaO and its major mineral phase is belite ((CaO)$_2$·SiO$_2$). Because of the lower amount of CaO, a lot less CaCO$_3$ has to be calcined. Since the cement does not require alite, the temperature for calcination and clinker production can be approximately 250°C lower (1200°C). Solidia cement can be produced in a conventional cement kiln, but because of the reduced amount of CaCO$_3$ that is decomposed and the lower production temperature a lot less CO$_2$ is emitted during the production. During the production of Solidia cement a total of 565 kg of CO$_2$ is emitted per tonne of cement produced.

Furthermore, Solidia cement is non-hydraulic and cures via a reaction with CO$_2$ in the presence of water, as is seen in equation 5. This same reaction is also the cause for most of the strength that regular concrete gains after the first two weeks, because Portland cement is also composed of (CaO)$_2$·SiO$_2$ to some degree. The Solidia Tech cement absorbed between 220 and 236 kg CO$_2$ per tonne of cement used.[27]

$$\text{(CaO)$_2$·SiO$_2$ + 2 CO$_2$ } \xrightarrow{\text{H$_2$O}} 2 \text{CaCO}_3 + \text{SiO}_2$$  \hspace{1cm} (5)

This makes that Solidia cement has a much lower carbon footprint as regular Portland cement. The most efficient cement plants reported 816 kg of CO$_2$ emitted per tonne of Portland cement produced[59], therefore replacing Portland cement with Solidia cement can offer a reduction of the carbon footprint by up to 60%. Although Solidia cement is a good example of cement with a much lower carbon footprint than ordinary Portland cement, it was not analyzed further. The Solidia Tech cement does not show a novel method to utilize CO$_2$ or sequester CO$_2$ in the final product.
6 An analysis on environmental impact and economic feasibility

In this section the environmental impact and the economic feasibility of each method that is discussed in this report will be analyzed. Effort will be made to estimate the maximum environmental impact each method can have. Availability of raw materials, demand of the corresponding product and other factors will be taken into account. The combination of environmental impact and economic feasibility will be used to assess the potential each method has.

To determine the economic feasibility of the methods below it is necessary to look into the details of the production process they use. The costs of the raw materials used and the amount of energy the process requires are the two main factors on which the economic feasibility is determined. When the cost of the raw material was not given, it was estimated by the lowest wholesale price found on Alibaba.com. The lowest price found here is only a decent guess if there is little concern for quality of the raw-materials. The prices in dollars are converted to Euro using the most recent exchange rate, which is $1 = €0.8499. The cost of electricity used in the process is €0.05/ which is the price for large industrial users in the EU known from information inside Avantium.

The cost of CO$_2$ capture and transport of all raw materials including CO$_2$ are estimated to be the same as the costs estimated by Mahoutian for the production of steel-making slag blocks. Mahoutian estimates the cost of CO$_2$ capture and transport as 42.1$/t$ and the transport cost of rock-like materials as 1.38$/t$. In their estimate the aggregates and slags (both rock-like materials) were transported for a distance of approximately 22 km. For all the analyses below it is estimated that the CO$_2$ emissions of transport can be neglected in the total carbon balance of the methods. The costs of transportation are also only calculated for the raw materials and not for the final product, assuming that the customer will pay for the cost of transportation of the final product.

6.1 Blue Planet aggregates

A quick note must be given before continuing with the analysis of the Blue Planet aggregates. This analysis was the first one done for this report and it was done far more extensive than is necessary and in much more detail than the analyses of the other methods. While this does make it harder to directly compare the results of this analysis with the analyses of the other methods, it does not change the overall conclusions. This analysis was ultimately kept in its entirety, because it explains

Blue Planet is a company that produces lightweight aggregates made from precipitated carbonated minerals. The method to make these aggregates is explained in section 5.1. This method shows potential because it captures CO$_2$ and creates a useful product at the same time, while it only requires a source of alkalinity and a divalent cation.

To assess the maximum environmental impact these aggregates can have it is necessary to find out what the limiting factor is for this method. It is unlikely that the limiting
factor is market demand, since the construction aggregate market is one of the largest markets on the world. Therefore, either the cost of the process or the availability of raw materials will be the limiting factor.

In the Blue Planet patent[44] an example of the production of aggregates is explained. A key resource that is used during the production is waste from a plant that creates magnesia (MgO) from sea water. This waste contains 85% Mg(OH)$_2$ and this is the key ingredients for creating the Blue Planet aggregates: a divalent cation and a source of base. Although this waste is the perfect raw material for the production of the aggregates, it is unknown how much of this waste is available.

The production process of creating MgO from sea water does not contain a step where Mg(OH)$_2$ is a waste and if this process where completely optimized all Mg(OH)$_2$ would be converted to MgO.[61] Therefore, it is likely that these kinds of magnesia production plants create only small amounts of Mg(OH)$_2$ waste. The total global magnesia production from seawater or brines in 2015 was only 1263 thousand tonnes.[62] If only a small amount of waste is generated per tonne of produced magnesia than it must be concluded that there is not enough Mg(OH)$_2$ waste being produced to create a significant amount of Blue Planet aggregates with the method that is described. Alternatively it is possible to buy Mg(OH)$_2$ from an industrial supplier. According to the US Geological survey the world reserve of the mineral brucite, which is the mineral form of Mg(OH)$_2$, is several million tonnes.[62]

Giannoulakis et al. describe a life cycle assessment of mineral carbonation for carbon capture and storage in European power generation in their article.[63] An estimation on the cost of aggregate production will be made using the data found in this article. The method of direct aqueous mineral carbonation developed by the National Energy Technology Laboratory is chosen to be the most comparable to the Blue Planet aggregate production, because the method is the most similar.

In the patent by Blue Planet[44] they describe that they used sea water, magnesium hydroxide originating from magnesium oxide plant waste, 50% NaOH solution and CO$_2$ from flue gases of some industrial process. It will be assumed that the sea water can be pumped from the ocean freely and the CO$_2$ will be used directly from an industrial source. The 50% NaOH solution must be bought from a commercial supplier. The patent describes the production process for a single batch, but it is not described how much aggregates can be produced in a single batch.

To find out what the cost of production of the Blue Planet aggregates is it is important to know how much aggregates are created per batch. Although this is not stated in the patent, the details of the production process will be used to estimate the size of the batch. They did state that 30% of the precipitate was converted to solid aggregates. Since the precipitate goes through several drying steps before the aggregates are created, it is assumed that the final product does not contain water. Although it is stated in the results of the patent that the aggregates contain a series of different kinds of mineral carbonates, it is assumed here that only MgCO$_3$ is present in the minerals. It is also assumed that all
present Mg$^{2+}$ will precipitate. To produce a single batch 4500 kg magnesia plant waste containing 3825 kg Mg(OH)$_2$ was added. All the Mg$^{2+}$ can react to form 5525 kg dry MgCO$_3$. If 30% of this precipitate can be converted into solid aggregates than a total of 1658 kg aggregates can be created per batch. Table 3 also shows the estimated costs if the process is more optimized and all precipitate can be converted to aggregates.

The lowest price for the 50% (w/w) NaOH solution is roughly $300/tonne.[64] According to the patent a single batch requires 300 gallons, which weighs 1712 kg. The price of the NaOH of a single batch is: $1712 kg \times \frac{\$300}{1000 kg} = \$513.60$ per batch, which converts to €436.51. The price per tonne of aggregates produced is then: \( \frac{\text{€436.51}}{\text{batch}} \times \frac{\text{batch}}{1.658 t} = \text{€263.28/t} \)

The method described by Blue Planet states that the magnesium hydroxide was jet milled to reduce the particle size. A 2014 paper by Shwarzwalder et al. found that the minimum cost of jet milling metal oxides was 0.34 kWh/kg.[65] The total energy required to jet mill 4500 kg Mg(OH)$_2$ waste is: $4500kg \times \frac{0.34\text{kWh}}{kg} = 1530\text{kWh}$. Using the price of electricity for industrial usage in the EU the added cost of jet milling will be: $1530\text{kWh} \times \frac{\text{€0.05}}{\text{kWh}} = \text{€76.50}$. The cost of jet milling per ton is $\text{€76.50} = \text{€46.14/t}$.

Prices for Mg(OH)$_2$ waste from magnesia plants could not be obtained and there is probably only a small amount of this waste available. However, it is also possible to buy Mg(OH)$_2$ for 100$ per tonne.[66] For each batch 4500 kg waste containing 85% Mg(OH)$_2$ is used and it can therefore be estimated that this can be replaced with 4500 kg * 85% = 3825 kg Mg(OH)$_2$ from an industrial supplier. This 3.825 t * $100 = $382.50 per batch that is created. The added cost per tonne of aggregate would be: $\frac{\text{€382.50}}{\text{batch}} \times \frac{\text{batch}}{1.658 t} = \text{€230.70/t}$. This amount converted to Euro is €190.07.

The patent by Blue Planet also states that they pump 76000 gallons of sea water in their reactor before the reaction and it is assumed that a similar amount of water has to be removed after the precipitate is formed. The energy of pumping water at a rate of 40 gallons/min which is stated in the patent is roughly 1kWh/1000 gallons[67], which brings the total cost of pumping all the water in the reaction tank for a single batch to: $76\text{kWh} \times \frac{\text{€0.05}}{\text{kWh}} = \text{€3.80}$. The cost of pumping water per tonne of aggregates is: $\text{€8.66/1.658} = \text{€2.29}$.

The Blue Planet patent describes how the aggregates are created after the precipitate is taken from the reaction bath. The precipitate is first filtered in a filter press, the energy requirements for this process are very small and will be neglected.[68] After filtration the precipitate is dried in an oven at 40$^\circ$C for 48 hours and in the final step it is stated that the aggregates can optionally be dried for 16 hours at 110$^\circ$C. To calculate the total energy consumption of drying it is assumed that all the water that is contained in the starting precipitate is evaporated and equation 6 is used. The drying efficiency, $\eta$, used in equation 6 is assumed to be 70%.[68] The precipitate described in the patent contains 27.38% water[44], therefore it is necessary to evaporate 377 kg water per tonne of produced aggregates. The latent heat of evaporation of water, $\Delta Q_{\text{vap}}$, is 2256.4kJ/kg. The heat capacity of the aggregates, $C_{\text{agg}}$, is assumed to be the same as that of MgCO$_3$. for
which is $0.897 \text{kJ/(kg} \times \text{K})$. The heat capacity of water, $C_{\text{H}_2\text{O}}$, is $4.184 \text{kJ/(kg} \times \text{K})$. For the difference in temperature, $\Delta T$, it was assumed that the aggregates are dried at approximately $100^\circ \text{C}$ and thus the $\Delta T$ is chosen at 80 (ambient temperature=20$^\circ \text{C}$).

$$Q = \frac{Q_1 + Q_2}{\eta}$$

(6a)

$$Q_1 = \Delta T \times (m_{\text{H}_2\text{O}} \times c_{\text{H}_2\text{O}} + m_{\text{agg}} \times c_{\text{agg}})$$

(6b)

$$Q_2 = m_{\text{H}_2\text{O}} \times \Delta Q_{\text{vap}}$$

(6c)

Using equations 6 it is found that the energy required to evaporate all the water from the precipitate is $1.498 \times 10^6 \text{kJ/t}$ or 416.1 kWh/t. Assuming electricity is used the cost of drying is $18.4 \text{kW} \times 5 \text{min} \times \frac{\text{hour}}{60 \text{min}} = 1.53 \text{kWh}$. The power consumption of the hydraulic press per tonne of aggregate is $1.53 \text{kWh} \times \frac{1000 \text{kg}}{90.6 \text{kg}} = 16.92 \text{kWh/t}$. The cost of this energy consumption is: \[\frac{16.92 \text{kWh}}{t} \times \frac{\text{0.05 kWh}}{\text{kWh}} = \text{8.85/t}.\]

To calculate the costs of transportation of raw materials it is assumed that the estimates by Mahoutian also apply for this process, which is that transport costs $1.38/t$ material. The total weight of the raw materials for the production of a single batch of aggregates is: $3825 \text{kg Mg(OH)}_2 + 1712 \text{kg 50%NaOH} = 5537 \text{kg material}$. The transportation costs for a single batch are estimated to be: $5537 \text{kg} \times \frac{t}{1000 \text{kg}} \times \frac{\$1.38}{t} = \$7.64$. A single batch is either 1658 kg if it is assumed that only 30% of the precipitated mineral can be converted into solid aggregates, which is stated in the patent, or 5525 kg if a more optimistic conversion rate of 100% is assumed. The total transport cost for 30% precipitate to solid aggregate conversion is: $\frac{\$7.64}{\text{batch}} \times \frac{\text{batch}}{1658 \text{ kg aggregate}} = \$4.61/t$, which converts to €3.92/t. For a 100% conversion rate the total transport cost is instead: $\$1.40/t$, which converts to €1.19/t.

It is estimated that this method does not require additional costs for CO$_2$ capture and transport, because the method is specifically designed to use CO$_2$ directly from flue gases without additional capture. The total cost of the Blue Planet process is summarized in table 3. In the patent it was stated that only 30% of the precipitated minerals were
converted to solid aggregates, the total cost of a tonne of aggregates would be £337.29/t. If the process were to be optimized and all minerals could be converted to aggregates the cost would be £116.39/t and it is shown in the table under the column with 100% efficiency. Because the total amount of available Mg(OH)\(_2\) from waste is unknown, but considered to be a small amount in total, an alternative cost was calculated for when Mg(OH)\(_2\) has to be bought from a commercial supplier. This significantly higher cost is shown in red in table 3.

To put these costs into perspective the created product has to be compared with a similar product that is on the market right now. The product created by Blue Planet can be compared to crushed limestone aggregate, which is sold for 10-20$ per tonne on Alibaba.com and for 31-43$ per tonne by a supplier in the USA.[70] Comparing these prices with the prices found in this analysis, it must be concluded that Blue Planet aggregates can not be produced feasibly with the method that was described.

Table 3: A summary of the economics of the Blue Planet aggregates.

<table>
<thead>
<tr>
<th>Process</th>
<th>cost (£/tonne aggregates)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>30% efficiency</td>
</tr>
<tr>
<td>Buying NaOH</td>
<td>263.28</td>
</tr>
<tr>
<td>Buying Mg(OH)(_2)</td>
<td>190.07*</td>
</tr>
<tr>
<td>Jet milling Mg(OH)(_2)</td>
<td>46.14</td>
</tr>
<tr>
<td>Pumping sea water</td>
<td>2.29</td>
</tr>
<tr>
<td>Transport of raw materials</td>
<td>3.92</td>
</tr>
<tr>
<td>Hydraulic press</td>
<td>0.85</td>
</tr>
<tr>
<td>Drying</td>
<td>20.81</td>
</tr>
<tr>
<td><strong>Total cost (incl. buying Mg(OH)(_2))</strong></td>
<td>527.36</td>
</tr>
<tr>
<td><strong>Total cost (excl. buying Mg(OH)(_2))</strong></td>
<td>337.29**</td>
</tr>
</tbody>
</table>

* This is the cost of buying Mg(OH)\(_2\) from an industrial supplier and is added to the total cost although the Mg(OH)\(_2\) used in the example above is from a waste stream.
** It is assumed that the Mg(OH)\(_2\) is taken from a waste stream and can be used free of charge.

### 6.2 CarbiCrete: Steel-making slag blocks

An article by Mahoutian and Shao describe the production of construction blocks from steel-making slag and this section will aim to estimate the maximum environmental impact this method can have and the economic feasibility.[38] This method shows potential because it creates a carbon-negative useful construction product entirely from industrial waste.

Because the demand for standard construction blocks in the US and Canada alone was estimated to be 4.3 billion blocks[71], it is estimated that the availability of steel-making
slag is the limiting factor for the maximum environmental impact of this method. Section 3.1 explains that a total of \(125 \times 10^6\) t of steel-making slag is produced globally each year and that 20% of this steel-making slag is not yet utilized. Since a differentiation between the different kinds of steel-making slag is not made in this estimation, it will be assumed that all available steel-making slag can be efficiently used to create the construction blocks. The method describes that initially a mixture of \(1555 + 930\) kg/m\(^3\) slag and water is moulded and dried. The method states that the moulded blocks are dried until a water/slag ratio of 0.10 remains. If we assume that this water is trapped in the blocks as mineral hydrates, it can be calculated that the final mixture contains \(1555 + 930 = 2485\) kg slag and \(91\) kg water. Therefore, a tonne of blocks contains \(909\) kg slag and \(91\) kg water. If we assume that all available steel-making slag is used to create steel-making slag blocks, then in total \(125 \times 10^6\) t slag \(\times \frac{t \text{ blocks}}{0.909\text{ t slag}} = 137.5 \times 10^6\) blocks can be produced.

Mahoutian explains in his article that the \(\text{CO}_2\) uptake after 24 hours was 4.6% by weight of slag. If all steel-making slag is carbonated to create construction blocks by the method described than the total yearly amount of sequestered \(\text{CO}_2\) is: \(125 \times 10^6\) t slag \(\times 4.6\% \frac{t\text{CO}_2}{t\text{ slag}} = 5.75 \times 10^6\) t \(\text{CO}_2\).

However, the production of these blocks also requires significant amounts of energy for the grinding of slag, \(\text{CO}_2\) recovery, removal, compression and liquefaction. Mahoutian made a carbon balance of the steel-making slag blocks and compared this balance with that of regular cement blocks.\[38\] The production of a single steel-making slag block emitted \(0.27\) kg \(\text{CO}_2\) and this block sequestered \(0.50\) kg \(\text{CO}_2\), making the total carbon balance of this block -0.23 kg \(\text{CO}_2\). From the amount of sequestered \(\text{CO}_2\) it can be concluded that a single block contains: \(0.50\text{kg CO}_2 \times \frac{kg \text{slag}}{4.6\% \text{kg CO}_2} = 10.87\) kg slag. The total amount of blocks that can be produced is: \(125 \times 10^6\) t slag \(\times \frac{1000\text{kg}}{t} \times \frac{\text{blocks}}{10.87\text{kg slag}} = 11.5 \times 10^9\) blocks.

According to Mahoutian’s estimates the production of a regular cement block emits 1.56 kg \(\text{CO}_2\). Therefore the total \(\text{CO}_2\) evaded is the amount of \(\text{CO}_2\) that is sequestered plus amount of \(\text{CO}_2\) that is not emitted for the production of a regular cement block minus the amount of \(\text{CO}_2\) that is emitted during the production of the steel-making slag block:

\[
5.75 \times 10^6\text{t CO}_2 + 11.59 \times 10^8\text{ blocks} \times \frac{1.56\text{kg CO}_2}{\text{block}} \times \frac{t}{1000\text{kg}} - 11.5 \times 10^9\text{ blocks} \times \frac{0.27\text{kg CO}_2}{\text{block}} \times \frac{t}{1000\text{kg}} = 20.52 \times 10^6\text{t CO}_2.
\]

Which is \(\frac{20.52 \times 10^6\text{t CO}_2}{1.45 \times 10^7\text{t CO}_2} \times 100 = 1.42\%\) of the total global \(\text{CO}_2\) emissions in the associated with cement production.

The economics of the steel-making slag construction blocks were already thoroughly described in the article by Mahoutian\[38\] and a short summary of their analysis is given here. The steel-making slag blocks were compared to cement blocks of the same size and comparable physical properties. The cost to produce both types of blocks is summarized in table 4 and this summary concludes that steel-making slag blocks can be produced at a lower price than regular cement blocks.
Table 4: A comparison of the production costs of regular cement blocks and steel-making slag blocks.[38]

<table>
<thead>
<tr>
<th>Item</th>
<th>Steel-making slag block</th>
<th>regular cement block</th>
</tr>
</thead>
<tbody>
<tr>
<td>Normal aggregates</td>
<td>–</td>
<td>17.8</td>
</tr>
<tr>
<td>Expanded aggregate**</td>
<td>23.4</td>
<td>–</td>
</tr>
<tr>
<td>BOF/EAF slag</td>
<td>2.6</td>
<td>–</td>
</tr>
<tr>
<td>Cement</td>
<td>–</td>
<td>14.1</td>
</tr>
<tr>
<td>Transport of raw materials</td>
<td>2.0</td>
<td>2.5</td>
</tr>
<tr>
<td>Grinding of slag</td>
<td>1.5</td>
<td>–</td>
</tr>
<tr>
<td>CO₂ capture + transport</td>
<td>1.8</td>
<td>–</td>
</tr>
<tr>
<td>Steam curing</td>
<td>–</td>
<td>2.4</td>
</tr>
<tr>
<td><strong>Total cost</strong></td>
<td>31.3</td>
<td>36.8</td>
</tr>
</tbody>
</table>

* The costs are expressed in € cents/block and not in €/tonne as is the case for the other analyses in this report.
** Aggregates from expanded blast furnace slag have considerably more value than the BOF/EAF slag that was bought, because they are commonly used in construction applications.
All prices were converted from US$ to €.

6.2.1 Aggregates from Steel-making slag

This section will estimate the maximum environmental impact the aggregates made from steel-making slag, CO₂ and water described in the article by Ghouleh et al.[46] can have. This method shows potential because it creates a useful construction material from industrial waste that sequesters CO₂ during its production.

The aggregates they made compared favourably with natural granite and the concrete blocks produced with these aggregates compared favourably with regular concrete blocks. Since the market for both natural granite and regular concrete blocks is very large, it is assumed that the limiting factor for the maximum environmental impact will be the available steel-making slag.

To estimate the maximum environmental impact and costs per tonne of aggregates produced it is necessary to know how much aggregates are made per tonne of steel-making slag that is used. According to the article the particles with a diameter greater than 212µm were discarded and this was 9.6% of the total weight. They also state that they mixed the milled slag with water with a water-to-slag ratio of 0.15, which results in the addition of 0.15 kg of water per kg of slag. Then finally it is stated that the CO₂ uptake is 17% by weight of slag.

Per tonne of steel making slag 1000 * 9.6% = 96 kg is discarded when sieving initially and 904 kg is leftover for aggregate production. To this smaller particle fraction of the slag 904 * 0.15 = 135.6 kg of water is added, it is assumed that this water all ends up in the final product. This results in 904 + 135.6 = 1039.5 kg of aggregate per tonne of slag.
before CO₂ absorption. When the aggregates have fully reacted with CO₂ their weight has increased by 17%, thus resulting in 1039.5 * 117% = 1216 kg aggregate per tonne of slag used. The amount of aggregates created per tonne of slag used is used to estimate the price of aggregates, shown in table 5.

The total available steel making slag is 125 * 10⁶ t, which was explained in section 3.1. The overall CO₂ uptake of the artificial aggregates averaged out at 17% CO₂ by weight of the dry steel-making slag, but 9.6% of the steel-making slag was discarded.[46] This allows us to estimate the total possible CO₂ uptake if all available steel-making slag is used to produce artificial aggregates: 17% * (100 − 9.6%) * 125 * 10⁶ t = 19.21 * 10⁶ t/y. Which is \( \frac{19.21 \times 10^6 \text{tCO}_2}{1.45 \times 10^7 \text{tCO}_2} \times 100 = 1.32\% \) of the total global CO₂ emissions in the associated with cement production.

The only raw material used in the production process is steel-making slag and the only step in the production process that requires a significant amount of energy is the milling of the steel-making slag. It is estimated that the price of steel-making slag is $3/t.[38] The milling of the steel-making slag required 36.7 kWh per tonne of slag according to the method described in the article by Ghouleh.[46] The cost of this electricity is per tonne of steel-making slag used: 36.7 kWh \( \times \frac{0.05 \text{€/kWh}}{1} = 1.84 \text{€}. \) The cost per tonne of aggregates produced can be calculated by the fact that 904 kg of slag has to be milled to create 1216 kg of aggregates:
\[
0.904 \frac{\text{t slag}}{1216 \text{t aggregate}} \times \frac{1.38 \text{€}}{1 \text{t aggregate}} = 1.36 \frac{\text{€}}{1 \text{t aggregate}}.
\]

As was calculated in above paragraphs one tonne of slag finally produces 1.216 tonne of aggregates, which is the same as \( \frac{1 \text{t slag}}{1216 \text{t aggregate}} = 0.82 \frac{\text{t slag}}{1 \text{t aggregate}} \). From this we can calculate the transport cost for the slag assuming the distance and the cost of transportation are the same as in the process described by Mahoutian: 0.82t slag*1.38$/t = $1.13. This converts to 0.96 transport costs of steel-making slag per tonne of aggregates produced.

To calculate the cost of CO₂ capture and transport it is assumed that the estimates by Mahoutian are valid for this process. It is also estimated that all captured and transported CO₂ is efficiently absorbed. The aggregates absorb 17% CO₂ by weight of slag. In the calculations above it was calculated that 904 kg slag creates 1216 kg of aggregates, which is the same as \( \frac{904 \text{kg slag}}{1216 \text{t aggregate}} = 0.74 \frac{\text{t slag}}{1 \text{t aggregate}} \). The total amount of required CO₂ per tonne of aggregates is: 0.74t slag * \( \frac{17\% \text{CO}_2}{\text{slag}} = 0.13 \text{t CO}_2 \). The cost of transport and capture of this amount of CO₂ is: 0.13t CO₂ * 42.18$/t CO₂ = $5.32. So the total cost of capture and transport of CO₂ per tonne of aggregate produced converted to Euro is: 4.52/t aggregate. If the transport distances are smaller than these costs will be significantly less. For example when the aggregate production and CO₂ capture are both done at the location of a steel plant then both these costs can be reduced and the entire process becomes more efficient.

The final cost of producing aggregate from steel-making slag is summarized in table 5. The cost of these artificial aggregates will be compared to the cost of natural crushed granite aggregates, because the created artificial aggregates compared favourably to natural granite aggregates. The minimal price of natural crushed granite found was 9-29$ per tonne, but much higher prices were also stated. The artificial aggregates created compared
favourably with natural granite, therefore a slightly higher price can be warranted. The cost of creating these aggregates leaves enough room to generate profit and therefore it is concluded that it is possible to create artificial aggregates utilizing CO$_2$ in an economic feasible manner.

<table>
<thead>
<tr>
<th>Process</th>
<th>cost (€/tonne aggregates)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Buying steel-making slag</td>
<td>2.47</td>
</tr>
<tr>
<td>Milling steel-making slag</td>
<td>1.36</td>
</tr>
<tr>
<td>Transport of slag</td>
<td>0.96</td>
</tr>
<tr>
<td>Capture and transport of CO$_2$</td>
<td>4.52</td>
</tr>
<tr>
<td><strong>Total cost</strong></td>
<td><strong>9.31</strong></td>
</tr>
</tbody>
</table>

Table 5: A summary of the economics of steel-making slag aggregates of Ghouleh et al.[46]

6.3 CarbonCure concrete

The method by CarbonCure to add CO$_2$ to concrete during the curing stage allows for some incorporation of CO$_2$ in the final cement product and it allows for a decrease in total cement usage due to higher cement strength.[50] The only raw material that is used is compressed CO$_2$. The method shows potential because it does not require significant input of energy or a high capital investment and it utilizes CO$_2$.

The total environmental impact from this method comes from the combination of a reduction in total cement usage and upcycling of CO$_2$ into the final product. The 2016 case study done by CarbonCure company only mentions a reduction in total cement usage, which corresponds to a reduction in CO$_2$ emissions. Their proposed method allows for a 5% cement reduction. The amount of CO$_2$ that absorbed in the final product is not mentioned in the case study done by CarbonCure. But a 2016 paper by Monkman et al. shows that approximately 1.3% CO$_2$ by weight of cement can be absorbed and incorporated into the final product.

To estimate the maximum total environmental impact it is necessary to know for which products this method can be used and how large these corresponding markets are. The patent by CarbonCure describes that their method to cure concrete with CO$_2$ is meant for concrete products that are cast in a mould.[51] These products include concrete blocks, planar products (such as pavers) and hollow products (such as concrete pipes). While only these examples are given, the patent also states that their method is not limited to these products and a later case study done by CarbonCure shows them implementing their method to ready-mix concrete.[72] It will be assumed that all precast concrete products and all ready-mix concrete can be treated with CO$_2$ in the manner described by CarbonCure.

In section 2.3 an approximation to the global cement usage was made. From this estimation we can approximate that the worldwide cement usage in concrete products
is 3.44 billion tonne. If all these concrete products would be produced with the method described by CarbonCure, then it is possible to reduce the total cement usage by $0.05 \times 3.444 \times 10^9 = 172.2 \times 10^6 t$ of cement. The total amount of CO₂ emitted per tonne of cement produced is 800 kg/t according to the cement technology roadmap from the IEA.[20] This reduction of cement usage avoids $172.2 \times 10^6 \times 0.8 = 137.8 \times 10^6 t$ CO₂.

There is also some CO₂ stored inside the concrete as carbonated rock. The amount of CO₂ that is stored is equal to $1.32\% \times 88\% = 1.32\%$ of the used cement by weight[54]. The total CO₂ stored is: $1.32\% \times 3.44 \times 10^9 = 44.8 \times 10^6 t$. Which is only a small amount compared to the CO₂ that is avoided. The total carbon balance for this method would be: $44.8 \times 10^6 + 137.8 \times 10^6 = 182.6 \times 10^6 t$ CO₂/y. Which is $\frac{182.6 \times 10^9 t CO₂}{1.32\% \times 88\% \times 3.44 \times 10^9 t CO₂} = 100 = 12.59\%$ of the total global CO₂ emissions in the associated with cement production.

The CarbonCure method to incorporate CO₂ in concrete is very straightforward and therefore determining the economic feasibility is should not be hard. During the mixing of the concrete CO₂ is added to the wet concrete and the reaction CO₂ has with the cement increases the overall strength of the cement and accelerates the setting of the concrete. The increase in strength allows for a more durable product or a reduction in cement usage. The accelerating behaviour of CO₂ allows for a faster setting concrete or it allows for a reduction of other types of accelerating admixtures.

The only raw material that is used in the process is CO₂ and the only additional costs are the cost of CO₂, extra labour cost and cost of CarbonCure gas injection hardware. No data is available on the amount of extra labour that is required and the prices of the required hardware. Therefore, the only extra costs that can be calculated is the cost of the used CO₂. The cost of CO₂ for industrial usage is currently $100-150 per tonne, but if more carbon capture facilities will become operational in the future this price might drop significantly. The exact amount of CO₂ added is unknown, but from the laboratory trials discussed in a CarbonCure presentation[72] and an article discussing CO₂ uptake in concrete products[54] it is assumed that 1.5% CO₂ by weight of cement is added to the concrete.

The amount of cement used can be reduced by 5% reduction of total cement usage, while maintaining the same level of strength. The minimum price of Portland cement is roughly $50 per tonne.[73] So if originally 100 tonnes of cement are used by adding CO₂ a reduction of 5 tonnes of cement can be made, but $95 \times 1.5\% = 1.425$ tonne CO₂ are added. Assuming both the CO₂ and the cement are bought from an industrial supplier at the lowest price then: $5t \times \frac{$50}{t} = $250 is saved and $1.425t \times \frac{$100}{t} = $142.5 is spent. This is a net saving of $107.50 for every 95 tonnes of cement used, which is $1.50/t or €1.27/t. Because applying the CarbonCure method to utilize CO₂ in the production of concrete results in a net saving of money, it can be concluded that this method is economically feasible.
7 Summary of discussed methods

The methods discussed in section 5 and 6 are quickly summarized in table 6. The method described by Blue Planet is an interesting method to utilize CO$_2$ and create a useful product at the same time, but the process that was described and analyzed is considered too expensive and the lack of enough available raw materials decreases the possible environmental impact it can have.

The method utilized by CarbiCrete and the method described by Ghouleh et al. show that it is possible to create both blocks and aggregates from steel-making slag utilizing CO$_2$ in an economically feasible manner. Because, both these methods use steel-making slag as a raw material their total environmental impact is linked and is roughly 20 * $10^6$ t CO$_2$/y if each year all steel-making slag is used to create these CO$_2$ sequestering building materials.

The method described by CarbonCure is the most simple to implement directly, because it can be incorporated without much interference in the production of most concrete. According to the analysis above implementing this method reduces the cost of produced concrete directly, therefore the method is assessed to be economically feasible. The environmental impact is limited, because only very little CO$_2$ is utilized and sequestered per tonne of product that is created. The total environmental impact this method can have is 182.6 * $10^6$ t CO$_2$/y, but this is mostly due to a possible reduction in total cement usage and only for a very small part due to CO$_2$ utilization. Although the calculated maximum environmental impact is quite high, this can only be achieved if every concrete producer in the entire world starts implementing this method.

<table>
<thead>
<tr>
<th>Method</th>
<th>Environmental impact</th>
<th>Economic feasibility</th>
</tr>
</thead>
<tbody>
<tr>
<td>Blue Planet aggregates</td>
<td>−</td>
<td>−</td>
</tr>
<tr>
<td>CarbiCrete blocks</td>
<td>+</td>
<td>+</td>
</tr>
<tr>
<td>Steel slag aggregates</td>
<td>+</td>
<td>+</td>
</tr>
<tr>
<td>CarbonCure concrete</td>
<td>+/−</td>
<td>+</td>
</tr>
</tbody>
</table>

Table 6: Summary of the methods discussed in this report. A minus sign (−) indicates that the method can only have small environmental impact or that it could not be economically feasible. A plus-minus (+/−) indicates that the method can have a limited environmental impact or that the economic feasibility is limited. A plus sign (+) indicates that the method can have a significant environmental impact or that it can be economically feasible.
8 Conclusion and outlook

The methods discussed in this report show that there is a lot of potential to utilize and sequester CO$_2$ in cement-based materials. The method used by Blue Planet shows that it is very hard to create cement-based construction materials from precipitated carbonated minerals in an economically viable way. This method is likely only viable when it is possible to obtain the necessary raw materials at a very low costs through the utilization of wastes.

The methods to utilize steel-making slag to create either aggregates or blocks showed that it is possible to utilize and sequester approximately $20 \times 10^6 t CO_2/y$. These methods are deemed economically viable and this is largely due to the fact that the steel-making slag is a very cheap raw material.

The method used by CarbonCure shows that it is possible to utilize and sequester $44.8 \times 10^6 t CO_2/y$ and avoid $137.8 \times 10^6 t CO_2/y$ if all cement production would adopt their technology. This method is deemed economically viable and this is due to the fact that no raw materials besides CO$_2$ are used. The method can also be incorporated into the current cement production rather easily. However, due to the large scale of the worldwide cement industry it is quite unrealistic to assume that all concrete production will adopt this method and the total possible environmental impact will likely never be achieved.

Other methods that were mentioned in this report with both a high possible environmental impact and that were economically feasible have one thing in common: they make use of a waste stream that is reactive to CO$_2$. Therefore, it can be concluded that there is potential to convert waste streams that are reactive to CO$_2$ to useful construction materials. The use of steel-making slag was discussed thoroughly, but different types of ashes and other industrial residues should also be investigated in more detail.

The methods that were discussed in this section all utilize CO$_2$ and have some extra costs accompanied to them because of this. This cost either comes from buying CO$_2$ from an industrial supplier or it comes from capturing and transporting it from an industrial emitter. But this might change in the future. When more countries start capturing CO$_2$ it is likely that a large surplus in the supply of CO$_2$ will arise and this will drive the price of CO$_2$ down making all the methods discussed more economically viable.

It can also be interesting to investigate how much can be gained from combining carbon capture with CO$_2$ utilization for the production of cement-based materials. Blue Planet is one example of a company that combines carbon capture directly at the source of emission with the production of a cement-based construction material. It can be especially interesting to combine the capture of CO$_2$ and production of the cement-based materials that utilize the CO$_2$ at the site where the wastes are produced. For example: capture the CO$_2$ from a steel-mill, utilize the steel-making slag they produce and create the products all at the same site.
Acknowledgement

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