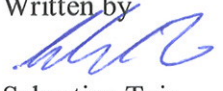

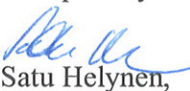




Evaluation of concepts for CO₂ neutralisation using feldspar

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<p>Summary</p> <p>The target of this work was to evaluate the feasibility of a new carbon dioxide (CO₂) neutralisation concept by literature review and mass and energy balance calculations. In this concept, CO₂ is captured from a gas stream, either by dissolution in pressurized water or using conventional CO₂ separation processes. An acidic CO₂-water solution is obtained, which is neutralized with feldspar. The neutralization of the feldspar is expected to result in a bauxite by-product that could be used as raw material for aluminium production. Three embodiments of the concept are given in a patent application related to the concept, of which two embodiments were assessed.</p> <p>None of the CO₂ neutralisation concepts assessed were found economically feasible, not even when taking into account revenues from by-product sales. The slow reaction kinetics of feldspar with CO₂ is the main bottleneck of the concept. The scarce experimental data available indicates that pressures over 100 bar CO₂ and temperatures between 100-300 °C are required to even reach a conversion as low as 9% of the theoretical maximum in 6 h. Completion of the reaction is therefore expected to take days, which is far too slow for an industrial process. The water consumption and the need for dumping of millions of tonnes of bicarbonate waste water is another hurdle for the concepts studied. This can be overcome by using only Ca-plagioclase for neutralisation, resulting in solid calcium carbonates, and not K-feldspar or Na-plagioclase, resulting in a bicarbonate solution.</p> <p>An alternative process for simultaneous CO₂ neutralisation and aluminium ore production by leaching feldspar in mineral acids was also briefly assessed. It was found technically feasible and less demanding to perform than the main CO₂ neutralisation concepts evaluated, but due to the large consumption of chemicals it was also found economically unfeasible.</p>		
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Preface

This evaluation was performed as a commission from Fortum during December-February 2012. The target of the project was to evaluate the feasibility of CO₂ neutralisation using feldspar, a concept that has received a lot of media attention lately. An objective assessment of the concept has not previously been made. The progress of the evaluation was monitored and directed by Risto Sormunen and Antti Arasto, with whom several fruitful meetings and discussions were held.

Espoo, 10.4.2012

Sebastian Teir

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1 Introduction

The urgent need to reduce greenhouse gas emissions for mitigating the on-going climate change has given birth to many new concepts. Of these, carbon capture and storage (CCS) is seen as the most promising concept that could contribute with roughly one fifth of the total reduction need (50-90%) by 2050 (IEA, 2008). In CCS, carbon dioxide is separated at the source (power plant or industry) and transported for final storage, where it needs to stay isolated from the atmosphere for millennia to come.

Currently, the only technologically and economically feasible option for long-term storage of CO₂ is storage in deep, underground geological formations, such as depleted oil and gas fields or saline aquifers (porous, sedimentary rock formations containing brine). However, only those formations that meet certain requirements (depth over 800 m, high permeability and porosity, impermeable layer of cap rock or clay on top of the formation etc.) can be considered for storage of CO₂. Also, the formation has to be monitored for tens of years after the injection of CO₂ has ended to verify that the CO₂ is securely trapped.

Due to the vast amounts of CO₂ that needs to be stored (number and citation here) there are only a few alternatives to underground geological storage that can be considered. One is mineral carbonation, i.e. conversion of CO₂ into environmentally stable carbonate minerals by reacting CO₂ with calcium silicates and magnesium silicates. So far, the concept has not reached the piloting stage, since the reaction rate of the minerals is far too slow for an industrial process. Although many different concepts have been suggested to speed up the reaction, most of these require either too much energy or chemicals for being feasible. Another alternative that has been suggested is the injection of either gaseous or liquid CO₂ into the deep ocean or deposition of CO₂ as a bicarbonate solution into the ocean. However, the ocean surface water is already being acidified by the higher CO₂ concentrations in the atmosphere and this has a negative effect on the organisms and ecosystem in the oceans. Therefore, recent legislations and regulations in Europe have forbidden injection of CO₂ into the oceans (particularly the OSPAR Commission and the EU CCS directive). Still, some countries (e.g. Japan) continue to develop ocean storage of CO₂.

Recently, a new Finnish invention for reducing CO₂ emissions by neutralising CO₂ with feldspar has received a lot of media attention. Nuclear physicist Matti Nurmia is the inventor of the concept and a company, Cuycha Innovations, has been established to develop this method and other inventions of Matti Nurmia. Except for neutralising CO₂ the emissions the concept is claimed to facilitate simultaneous extraction of valuable industrial metals and minerals from feldspar, such as alumina, lithium and tantalum. The concept is currently set to be piloted in South Africa (Cuycha Innovation, 2011).

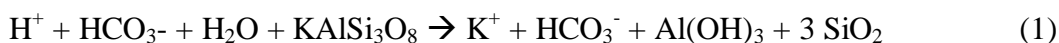
2 Goal

The target of this work was to evaluate the feasibility of the CO₂ neutralisation concept presented by Matti Nurmia and Cuycha Innovations. The chemistry

involved in the concept was initially assessed by Huttunen (2009). After that, the concept was preliminarily evaluated by Rajamäki (2011), who had discussed with the inventor and Cuycha Innovation for gaining additional information regarding the concept and newer innovations related to this. The present study focuses on the neutralisation concept and the three embodiments presented in the published patent application by Nurmia (2011).

3 Description of the concept studied

The concept of carbon dioxide neutralization is presented in a patent application by Nurmia (2011). The objective of the invention is a process, where CO₂ is captured from a gas stream by dissolution in pressurized water. The acidic CO₂-water solution obtained is neutralized with the help of feldspar minerals of gravel, sand or crushed rock material, which is essentially the same as the chemical weathering phenomenon that happens to feldspar minerals in nature caused by CO₂ dissolved in rainwater (example with orthoclase, i.e. potassium feldspar):



According to the patent the neutralization of the feldspar minerals results in bauxite, which is a mixture of gibbsite, Al(OH)₃, and kaolinite, Al₂Si₂O₅(OH)₄. Further on, the patent states that the bauxite can be separated from the rock material by washing and further processed into aluminium oxide, an important raw material for aluminium production. The neutralized solution, in which the hydrogen ions have been replaced with ions of alkali or alkaline earth metals, can according to the patent be passed to the sea or rivers without increasing the acidity of the waters. Three embodiments of the innovation are given in the patent application.

3.1 Embodiment 1

Embodiment 1 describes a process for use with a flue gas or other gas at normal pressure. The flue gas passes through a heat exchanger (Figure 1, item 21) and a compressor (item 22) equipped with water injection that pressurises the flue gas (e.g. 5 bar). The pressurized flue gas is led into a washing column (item 23), into which cold water is sprayed. The main part of the CO₂ in the flue gas dissolves into the water flow and the CO₂-water solution is passed into a “neutralization space” (item 24), containing the mineral matter. The neutralized solution exits into

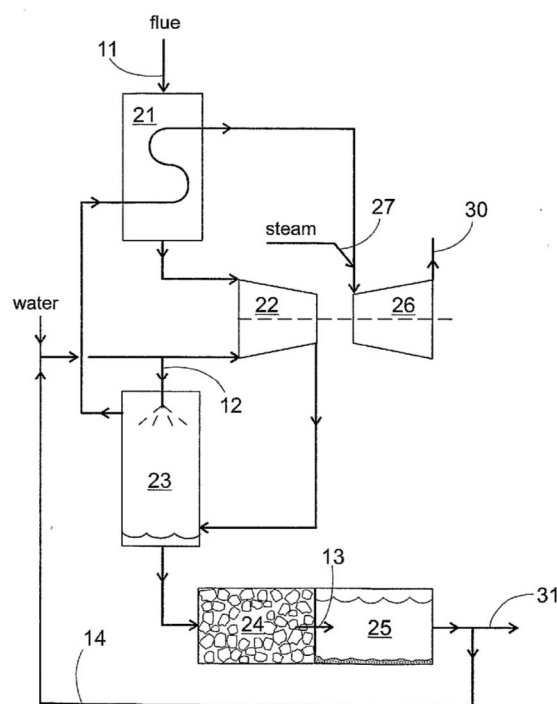


Figure 1. Process flow chart of Embodiment 1

a bauxite settling pool (item 25), from where the bauxite precipitate is collected at regular intervals. Part of the solution is recycled back. The crushed mineral matter is exchanged at regular intervals. To minimize power consumption the stripped flue gas is led to a turbine (item 26). According to the patent, the combination of turbine and compressor may produce surplus energy depending on the CO₂ content, pressure and temperature of the flue gas.

3.2 Embodiment 2

In Embodiment 2 the flue gas is divided into two streams, of which one (Figure 2, stream A) is pressurized (e.g. 20 bar). According to the patent, flue gas stream B is only used for covering for the heat requirements of the process. The other main differences to Embodiment 1 is the use of two-stage compression and expansion and the use of an underground reactor chamber functioning both as a neutralization reactor and washing column. The neutralized solution exits through stream 31.

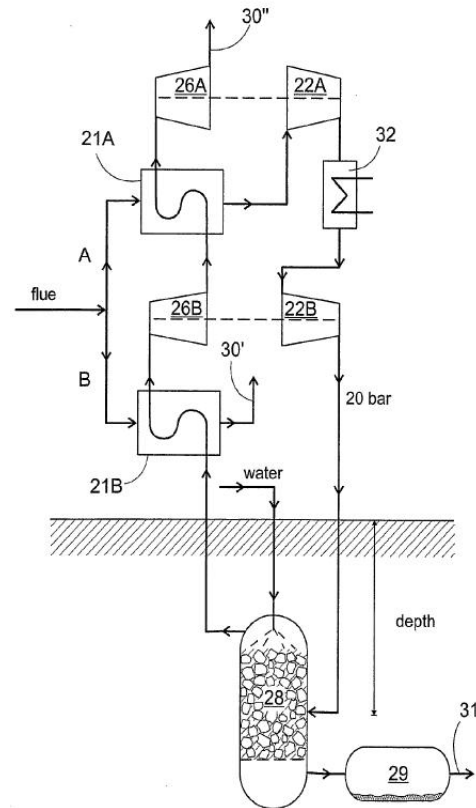


Figure 2. Process flow chart for Embodiment 2.

3.3 Embodiment 3

Embodiment 3 uses liquefied carbon dioxide under pressure (e.g. 20-40 bar). Similarly to Embodiment 2 the dissolution and neutralization takes place in an underground reactor chamber. CO₂ is dissolved into the water in the lower part of the reactor (Figure 3, section 28'), which flows through a fluidized bed filled with crushed rock (section 28'') and from there to the settling pool (29), where the bauxite is separated.

4 Resources

This chapter focuses on determining whether the feldspar resources for

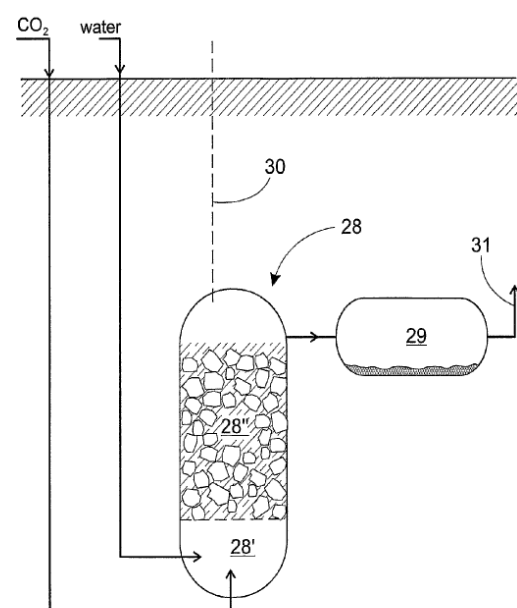


Figure 3. Flow chart of Embodiment 3.

implementing a CO₂ neutralisation process are available in such a large scale as needed for a full-scale process.

4.1 Feldspar

Feldspars (KAlSi_3O_8 – $\text{NaAlSi}_3\text{O}_8$ – $\text{CaAl}_2\text{Si}_2\text{O}_8$) are a group of rock-forming minerals, which make up as much as 60% of the Earth's crust (Wikipedia). Compositions of feldspars can be expressed in terms of three endmembers: Potassium-Feldspar - KAlSi_3O_8 , albite - $\text{NaAlSi}_3\text{O}_8$, and anorthite $\text{CaAl}_2\text{Si}_2\text{O}_8$ (Figure 4). Solid solutions between K-feldspar and albite are called *alkali feldspar*. Solid solutions between albite and anorthite are called *plagioclase feldspar*. Feldspar minerals are found in both intrusive igneous rocks (e.g. granite) and extrusive igneous rocks (e.g. certain basalts), as veins, and are also present in many types of metamorphic rock (e.g. gneiss). Rock formed almost entirely of calcic plagioclase feldspar is known as anorthosite. Feldspars are also found in many types of sedimentary rock.

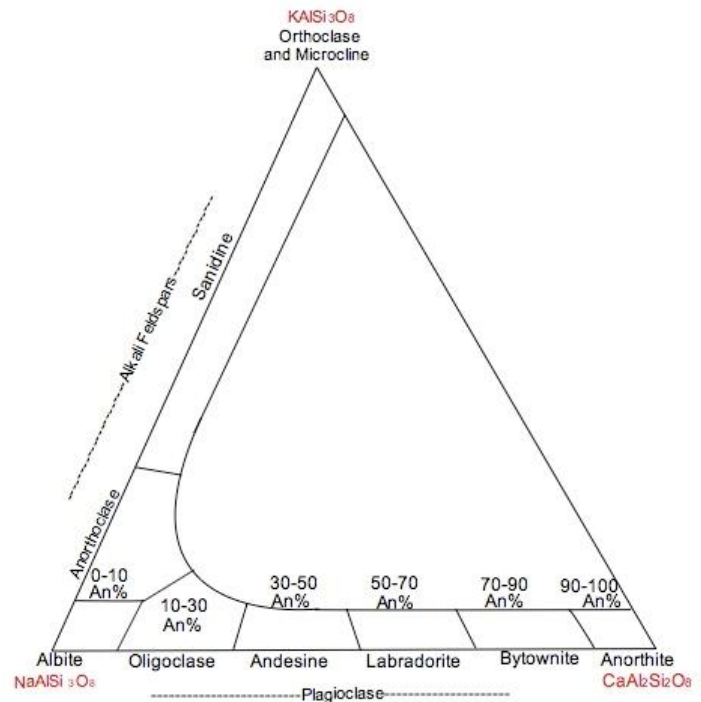


Figure 4. Phase diagram that shows the different minerals that the feldspar solid solution can consist of.

4.2 Use and production of feldspar

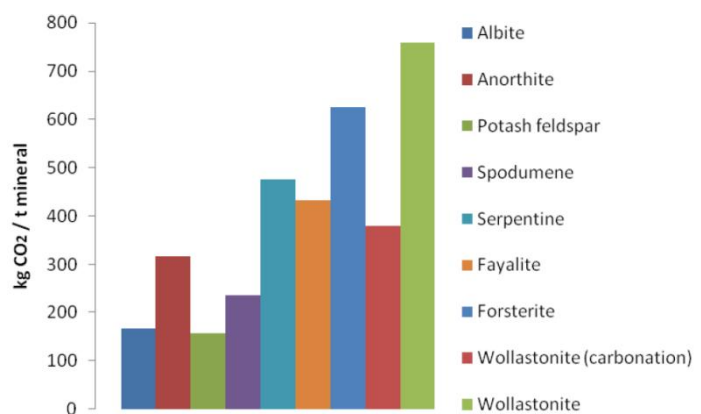
Pure feldspar has commercial uses. Feldspar is a common raw material used in glassmaking (66% of the feldspar consumption in the U.S.), ceramics, and also to some extent as a filler and extender in paint, plastics, and rubber (Wikipedia). About 20 million tonnes of feldspar was produced (i.e. mined) in 2010 (USGS, 2011). The largest producers were Italy (4.7 Mt), Turkey (4.5 Mt), and China (2 Mt). According to USGS the identified and hypothetical resources of feldspar are more than adequate to meet the anticipated world demand. Quantitative data on resources of feldspar existing in feldspathic sands, granites, and pegmatites have generally not been compiled. In Finland, the production of feldspar between 1967-2001 has been on an average 50 000 t per year (Puustinen, 2003). According to the latest available price list in Industrial Minerals (2011) crude Na-feldspar (Turkish, -10mm size bulk, FOB) costs 22-23 US\$/t while glass grade Na-feldspar (Turkish, -500 microns, bagged, FOB) costs 70 US\$/t.

4.3 CO₂ reduction requirements

According to IPCC, a greenhouse gas emission reduction of 80-95% by 2050 compared to 1990 levels is required to stabilize the rise of the global average temperature to an acceptable level of +2°C (IPCC, 2007) and the long-term strategy of the European Council supports this target. For Finland's part, the target has been evaluated to be achievable by 2050 and CCS could contribute to abate 10-20 Mt CO₂/a by 2050 (Teir et al. 2011). The ten largest CO₂ emission sources in Finland in 2008 ranged from 0.8 Mt/a to 4.4 Mt/, which gives an indication on the maximum required capacity for a CO₂ neutralization plant.

4.4 Estimation of feldspar resources

According to USGS (2011) the identified and hypothetical resources of feldspar are more than adequate to meet the anticipated world demand considering current uses. However, if feldspars were used for neutralizing CO₂ the quantities needed would be on a different scale than today's uses. In theory, neutralizing 1 tonne of CO₂ according to Equation 1 requires 3.1 – 6.7 t of pure feldspar



mineral, depending on the feldspar composition (Figure 5). In practice, more rock is required for a neutralisation process depending on the contents of feldspar minerals in the rock used and to what extent the neutralization process consumes the rock used.

Figure 5. Theoretical neutralization capacities of various minerals (Rajamäki, 2011).

Anorthosite rock consists mostly of plagioclase feldspar (90–100%) and would therefore be the ideal resource for a CO₂ neutralisation process. Available data seem to indicate that resources are enough for flue gas CO₂ neutralisation on a large scale. For instance, total U.S. resources of anorthosite is estimated at 599 Gt (National Research Council, 1970). This would equal a maximum CO₂ neutralisation potential of 90-190 Gt CO₂ in the anorthosite resources alone. For comparison, the stationary CO₂ sources in the U.S. have total annual emissions of more than 3.4 Gt annually, while the CO₂ storage capacity of saline aquifers in the U.S. has been estimated at 1,600 – 22,000 Gt (NETL, 2010a). Norway has also large anorthosite deposits of industrial significance. One of the major deposits is the Gudvangen deposit that contains 500 Mt anorthosite with a chemical composition of 29% Al₂O₃, 48% SiO₂, and 14% CaO (Industrial Minerals, 2011).

Although the current feldspar production in Finland is low in comparison to the scale required for CO₂ neutralization, it is possible that existing waste rock from mining operations in Finland could contain feldspar of a sufficient quality for a

CO₂ neutralisation process. For instance, in 2008 the operational mines in Finland extracted a total of 39 Mt of rock, of which 17 Mt was leftover (i.e. waste) rocks (Tuusjärvi et al. 2008). Of this, the extraction and use of natural stone produced 2.8 Mt of granite and shales as leftover rocks. In total 1.3 Gt of rock has been extracted in Finland, of which 430 Mt is leftover rock (Puustinen, 2010). However, a brief literature review did not result in any large feldspar resources in leftover rocks. An investigation of the tailings¹ and waste rocks of five large-scale operational mines in Finland (Pyhäsalmi, Hitura, Talvivaara, Siilinjärvi and Ihalainen) found a significant concentration of feldspar in portions of the waste rock from the Ihalainen quarry. The waste rock production at Ihalainen was 740 000 t in 2005 (Toropainen and Heikkinen, 2006) and a total of 13 Mt of waste rock has been produced since the opening of the mine (Puustinen, 2010). The feldspar concentration of other tailings and waste rocks varied between 0 – 24%.

*Table 1. Mineralogy of waste rock from the Ihalainen quarry (Toropainen and Heikkinen, 2006). Feldspar marked with **bold** type.*

K-Feldspar, KAlSi₃O₈	62 %
Quartz, SiO ₂	31 %
Muscovite, KAl ₂ (Si ₃ Al)O ₁₀ (OH,F) ₂	2 %
Plagioclase, (Na,Ca)Al(Al,Si)Si₂O₈	1 %
Epidote, Ca ₂ (Al,Fe) ₃ (SiO ₄) ₃ (OH)	1,5 %
Carbonate	0,5 %
Opagues	2 %

4.5 Other minerals

Except for feldspars, also spodumene, LiAl(SiO₃)₂, is listed as a suitable mineral for the neutralization process (Rajamäki, 2011). In theory, lithium carbonate could be extracted from this material in conjunction with a CO₂ neutralization process. Magnesium-, calcium- and iron silicates are also listed as potential neutralization minerals, but these have been and are being extensively studied in the field of mineral carbonation processes and won't be covered here.

5 Technology

This chapter focuses on reviewing the technology required for the CO₂ neutralisation concept.

5.1 CO₂ capture with water

In Embodiment 1 (see Chapter 3) of the CO₂ neutralization concept CO₂ is separated from the flue gas using pressurized water. The absorption of carbon dioxide in water at elevated pressures was formerly an important industrial process, but has now been replaced by more efficient systems that use chemical or

¹ the materials left over after a process that separates the valuable fraction from the uneconomic fraction of an ore

physical solvents (Kohl & Nielsen, 1997). To the best of our knowledge, there is currently no CCS concept being developed where pressurized water is used as a CO₂ solvent. According to Herzog et al. (2009), water is much more soluble to CO₂ than to N₂, but its capacity for CO₂ capture is so low that capturing industrial-scale amounts of CO₂ would require the circulation of very large water flows. In addition to this, the flue gas needs to be pressurized, which would demand much more compression power than current CCS concepts demand, where only the separated CO₂ fraction is compressed.

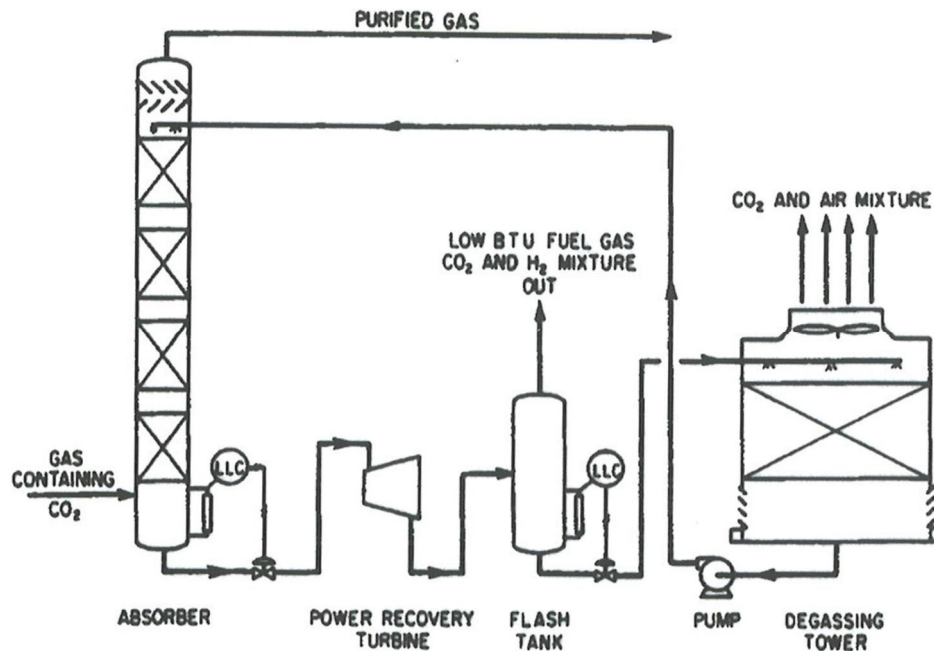


Figure 6. A simplified flow diagram of a water-wash process for absorption of CO₂ from gas streams (Kohl & Nielsen, 1997).

Figure 6 shows a flow diagram of a water-wash process for CO₂ capture. The plant consists in its simplest form of an absorption tower operating at elevated pressures, a flash chamber where CO₂ is released from the water after pressure reduction, and a pump to return the water to the absorber. In the flow diagram a power recovery turbine has been added as well as a degassing tower for removing more CO₂ from the water than a simple flash enables. According to Kohl & Nielsen, the process is in general limited to gas streams containing CO₂ at a partial pressure greater than 3.5 bar due to the low CO₂ capacity of water. The main advantages of a water-wash process over a monoethanolamine (MEA) - based capture process are:

1. A simple plant design without the need for heat exchangers or reboilers
2. No heat load
3. Inexpensive and environmental friendly solvent
4. Solvent is not reactive with COS, O₂ and other possible trace components
5. No nitrogenous solvent vapour losses into the gas stream.

The main disadvantages are a very high pumping load, a poor CO₂ removal efficiency, an impure CO₂ product, and risk for corrosion due to the acidic water. Due to these disadvantages the water-wash process is rarely used nowadays for CO₂ removal.

The CO₂ absorption capacity of water is depended largely both on pressure and temperature (Figure 7). In the patent application by Nurmia, an example is given for washing a flue gas, containing CO₂ with a partial pressure of 1.5 bar, with water at 5 °C. At these conditions the solubility of CO₂ is stated to be 4.5 kg/t water, which seems to correspond well to the value that can be read from the graph in Figure 7. In Embodiment 1 a flue gas pressure level of 5 bar is suggested. Assuming a typical coal fired power plant flue gas with a CO₂ concentration of 13.5%, the partial pressure of CO₂ would be 0.68 bar, corresponding to a CO₂ capacity of about 2 kg/t water. For comparison, MEA has a theoretical CO₂ capacity of 0.5 mol CO₂ per mol MEA (Zhang & Farthing, 2009). In practice, up to 30 wt-% MEA solutions are used for CO₂ scrubbing, which would correspond to a theoretical capacity of 170 kg/t solvent.

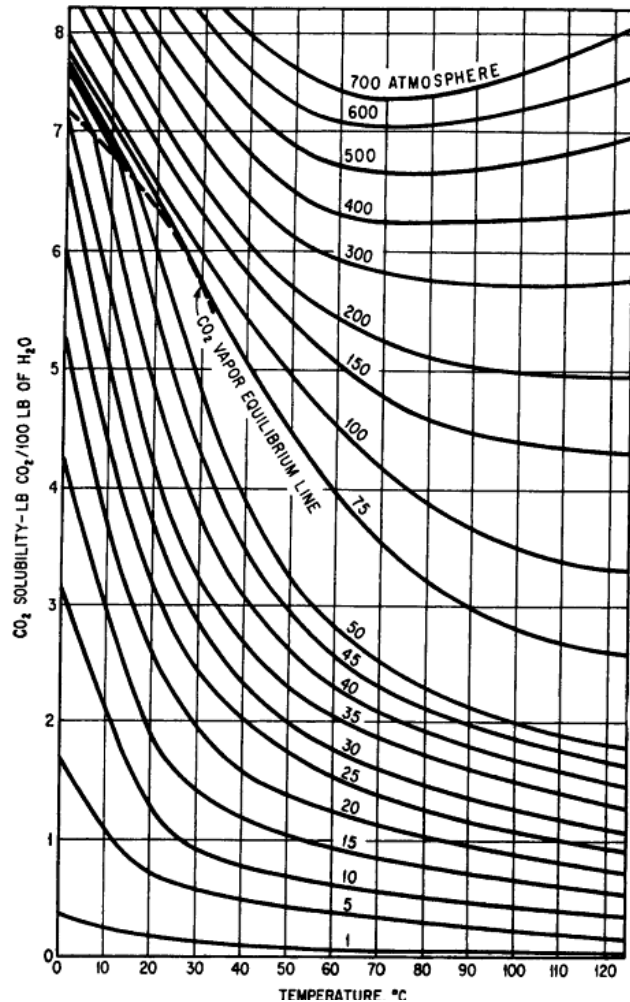


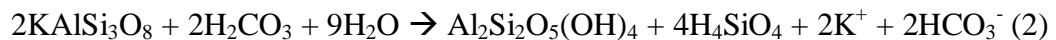
Figure 7. Solubility of CO₂ in water at partial pressures of 1 atm and higher (Kohl & Nielsen, 1997).

Kohl & Nielsen (1997) presents also operational data for a plant, where CO₂ is removed from synthesis gas by water scrubbing: The scrubber was operated in July at 17 bar with a feed water temperature of 27 °C, which cooled the gas down to 29 °C. The CO₂ concentration of the feed was 16.7 % and the removal efficiency was as high as 95 %. From the operational data presented, the CO₂ capacity of the feed water was calculated to ~0.01 kg CO₂ per kg H₂O. In another example with the same scrubber operating in December with a feed water temperature of 11 °C, which cooled the gas down to 13 °C, the CO₂ capacity of the feed water was ~0.02 kg CO₂ per kg H₂O. This is peculiar, since it is over twice the CO₂ solubility in water at equilibrium conditions (Figure 7). Since the data

was taken from an ammonia production plant it is likely that traces of ammonia in the gas stream improved the CO₂ capacity of the water scrubber. Data from another example in Kohl & Nielsen performed at a 12 bar total pressure (61 vol-% CO₂) and 15 °C shows a CO₂ removal efficiency of 99 % with a capacity of 0.005 kg CO₂/kg H₂O – roughly half the equilibrium capacity. The higher removal efficiency that is required the more excess water will naturally be required.

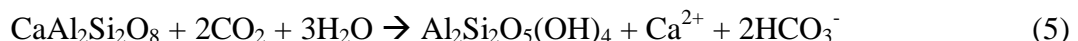
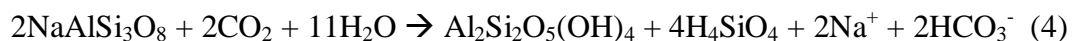
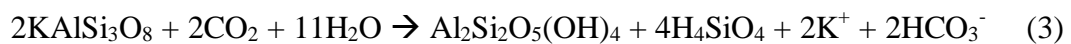
5.2 Neutralisation of feldspar with CO₂

The neutralisation of feldspar with CO₂ is the most important process step that each embodiment in the concept studied is built around. The idea comes from the natural process known as weathering of rocks. The process can further be divided into mechanical (disintegration of rocks without chemical change) and chemical weathering. Chemical weathering changes the composition of rocks, often transforming them when acidic waters interacts with minerals to create various chemical reactions, causing secondary (i.e. new) minerals to develop from the original minerals of the rock. Rainfall is slightly acidic by nature, because atmospheric carbon dioxide dissolves in rainwater, producing weak carbonic acid (H₂CO₃). Aluminosilicates, such as feldspar, produce when subjected to the hydrolysis reaction a secondary mineral rather than simply releasing cations (Wikipedia):



The weathering reactivity of feldspar can be compared to other minerals by using the Goldich dissolution series (Figure 8). Goldich (1938) found that minerals that form at higher temperatures and pressures are less stable on the surface than minerals that form at lower temperatures and pressures. This pattern follows the same pattern of the Bowen's reaction series, with the minerals that are first to crystallize are also the first to undergo chemical weathering (Wikipedia). According to the Goldich dissolution series, the most reactive feldspar is calcium-rich plagioclase, which should have a similar reactivity to olivine. The reactivity weakens as the sodium content of the plagioclase increases in relation to the calcium content. Orthoclase, or potassium feldspar, is the least reactive feldspar.

Huttunen (2009)² has assessed the chemistry involved in the CO₂ neutralization concept. Simplified weathering reactions for feldspar endmembers (orthoclase – 3, albite – 4, anorthite – 5) are presented in the following equations:



According to the chemical weathering process occurring in nature, solid kaolinite as well as solute silica and sodium/potassium/calcium carbonate are expected to

² Note: there are errors in the reaction equation balances for K-feldspar both in Huttunen (2009) and Rajamäki (2011)

form. Both sodium carbonate and potassium carbonate are highly soluble in water (216 g/l and 1120 g/l, respectively, at 20 °C) and are therefore expected to remain in solution. On the other hand, calcium carbonate is sparingly soluble in water (0.015 g/l at 25 °C) and is therefore expected to form a solid precipitate. Similarly, silica is sparingly soluble (0.079 g/l) and is also expected to precipitate.

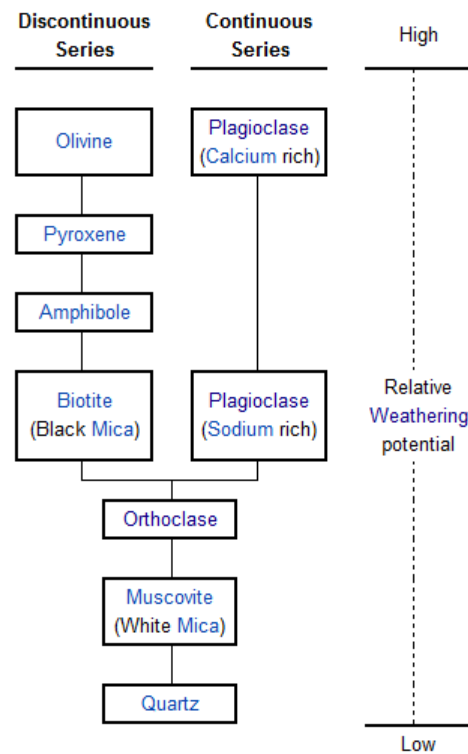


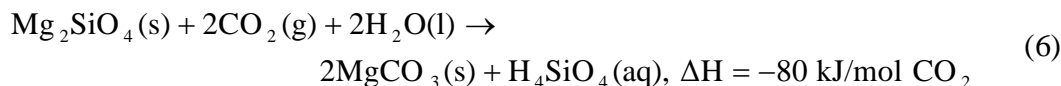
Figure 8. The weathering rate of various minerals (Goldich, 1938).

The weathering of feldspar in nature is extremely slow. For a CO₂ neutralisation process the weathering can be speeded up by raising temperature and CO₂ pressure as well as lowering the pH. The reaction surface can be increased by grinding the material finer, thus also increasing the speed of the reaction.

However, there is currently very little experimental information available related directly to this concept. Huttunen (2009) performed a set of experiments, but the results were far from promising. Nordic Mining and StatoilHydro tested in cooperation with the Norwegian Institute for Energy Technology (IFE) new technology for processing of anorthosite and binding of CO₂ (Nordic Mining, 2009). According to their press statement, the tests verified that aluminium feedstock (gibbsite) of satisfactory quality can be produced from anorthosite, but the reaction rates in the tests were too low for industrial scaling of the technology. Unfortunately, no data has been published related to these results. Nordic Mining has now proceeded with developing a process for processing the anorthosites with acid (see Chapter 5.3).

As already implied by the Goldich dissolution series (Figure 8) the reactivity of feldspar is expected to be similar to those of olivine. Aqueous carbonation of olivine and several other magnesium and calcium silicates is the most

comprehensively studied carbonation process (O'Connor et al. 2005), and can therefore be used for comparison. The process and reactions involved resembles that of the CO₂ neutralisation concept, but instead of producing aqueous bicarbonate solid magnesium and calcium carbonates are formed. In this process a slurry of water and olivine is reacted with pressurised carbon dioxide to produce magnesium carbonate:



Although the conversion chemistry involves three steps, it takes place in a single reactor. Carbon dioxide is dissolved in water to form carbonic acid (H₂CO₃), which dissociates to hydrogen cations (H⁺) and bicarbonate anions (HCO₃⁻):



The hydrogen cations react with the mineral, liberating magnesium cations (Mg²⁺):



The cations react with the bicarbonate to form solid carbonate and silicic acid (which in turn becomes silica and water):



According to O'Connor et al. (2005), the same process could be used for carbonating Ca-silicates as well. Correspondingly, the carbonate product would be calcium carbonate (CaCO₃):



Preliminary tests conducted at ambient temperature and sub-critical CO₂ pressures (below 74 bar) resulted in very slow carbonate formation. In later tests, using an aqueous solution of sodium bicarbonate (NaHCO₃) and sodium chloride (NaCl) at elevated temperatures and pressures, several silicate minerals were successfully carbonated to a large extent in one hour (Table 2). The lowest costs reported from a case-specific feasibility study regarding storing CO₂ using this method were 54 US\$/tCO₂ with olivine as feedstock, but later reviews have assessed the lowest cost to 80 euro/t CO₂ stored (Styring et al. 2011). The study included pre-treatment costs, but excluded CO₂ separation and transport costs.

Feldspar was tested by O'Connor et al. only in two experiments. Similar conditions were used as for the other minerals, resulting in a low carbonate conversion (9 %), although the residence time had been extended to 6 h (Table 2). Due to the scarcity of publically available data, this is currently the best reported carbonation conversion of feldspar.

There has been some other public research activity related to the reactivity of feldspar with CO₂ at pressures and temperatures related to geological storage of CO₂. Feldspar is namely also found in sedimentary rock and the behaviour of feldspar in contact with CO₂ is therefore of considering how it affects the geological storage of CO₂. For instance, Hangx and Spiers (2009) tested anorthite and albite in a plagioclase–CO₂–water system at temperatures in the range 200–300 °C, using carbon dioxide pressures from 0.4 to 15 MPa. Although the experiments were performed for 7–21 days, hardly any carbonate formation was detected. Lagache (1975) tested the dissolution of alkali feldspar at 100–200 °C at 6 bar CO₂ pressures. Also Sorai and Sasaki (2010) studied the dissolution kinetics of anorthite in a supercritical CO₂–water system. Suto et al. (2007) studied the behaviour of granite at over a temperature range of 100–350 °C at up to 250 bar. Although Ca-plagioclase have in most studies been reported to react relatively fast, the reactions rates were measured in days and are therefore not promising for an industrial process. The results from most of the studies seem to follow the Goldich dissolution series, even at high temperatures and pressures, with Ca-plagioclase being the most reactive and K-feldspar being the least reactive of the feldspars.

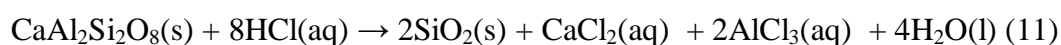
Table 2. Best carbonate conversion achieved with aqueous carbonation with the following test conditions (batch autoclave with continuous stirring): 80% <37 μm feed; 1 hour residence time; T = 185 °C; PCO₂ = 150 atm; 15% solids; 0.64 M NaHCO₃, 1 M NaCl (O'Connor et al., 2005)

Rock / Mineral group	Mineral	Formula	Conversion to carbonate (%)
Feldspar	Anorthite	CaAl ₂ Si ₂ O ₈	9 ^a
Serpentine	Antigorite	Mg ₃ Si ₂ O ₅ (OH) ₄	92
Pyroxene	Augite	CaMgSi ₂ O ₆ +(Fe,Al)	33 ^a
Basalt			15
Olivine	Fayalite	FeSiO ₄	66
Olivine	Fosterite	Mg ₂ SiO ₄	81
Serpentine	Lizardite	Mg ₃ Si ₂ O ₅ (OH) ₄	40
Oxide	Magnetite	Fe ₃ O ₄	08
Ultramafic	Talc	Mg ₃ Si ₄ O ₁₀ (OH) ₂	15
Ultramafic	Wollastonite	CaSiO ₃	82

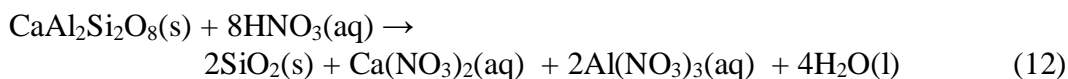
^aThe residence time was 6 h for anorthite and augite.

5.3 Alternative technology

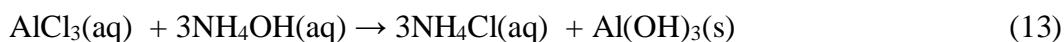
An alternative concept for producing aluminium from anorthosite (a rock formed almost entirely of calcic plagioclase feldspar) was presented by Råheim et al. (1998). The concept is visualized in Figure 9. In this concept anorthosite is leached using hydrochloric acid or nitric acid³:



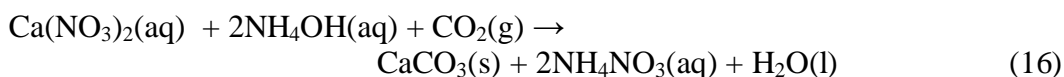
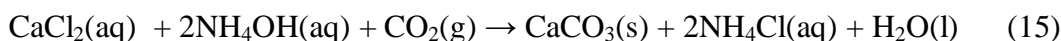
³ Reaction equations were not presented by Råheim et al. (1998), so simplified equations were derived based on the explanations in the original text.



The leaching residue from this type of acid process is a white, highly porous, low-density, amorphous silica gel, which could be used for instance as a filler in paint, plastics and paper, as well as a binding material in cement production (Wanvik, 2000). After the residue has been filtered out, ammonium hydroxide is added to raise the pH of the solution to 2.5 – 5, resulting in precipitation of aluminium hydroxide:



After the aluminium hydroxide has been filtrated from the solution, the pH of the remainder of the solution is regulated to 6 – 12, while the solution is brought into contact with CO₂ from an industrial source, causing calcium carbonate to precipitate:



Precipitated calcium carbonate is used as a filler material in paint, paper and plastics. Ammonium nitrate is commonly used as a fertilizer.

A similar kind of process was studied by Teir et al. (2009) for use with serpentine mineral. The benefits of such a “pH-swing” process is that it can operated at room temperature and make use of CO₂ in flue gases – flue gas can either be directly injected into a solution containing calcium chloride/nitrate and ammonium hydroxide, or the flue gas stream can be “washed” in a spray tower using an ammonium hydroxide solution, after which the resulting ammonium carbonate/bicarbonate solution can be brought in contact with the calcium chloride/nitrate solution, resulting in precipitation of CaCO₃. The main drawback of such a process is the consumption of large amounts of chemicals. Also, experimental data from a single extraction example presented by Råheim et al. (1998) gave an extraction of 80-90% of the various elements involved after 12 h leaching time, which is relatively slow for use in a continuous process.

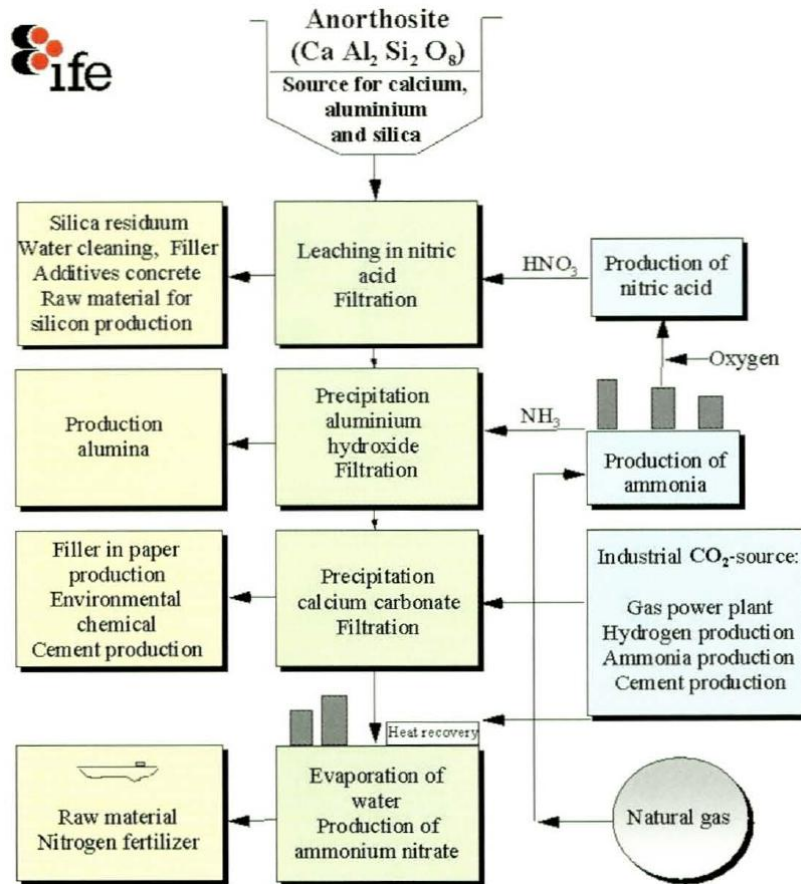


Figure 9. Alternative process for CO₂ neutralisation using anorthosite (Wanvik, 2000).

According to a recent press release by Nordic Mining promising results has recently been achieved for production of alumina from anorthosite (Nordic Mining, 2011). The process is based on usage of a common mineral acid where alumina is leached and recovered in different process steps. Precipitated calcium carbonate is made as a bi-product with addition of CO₂. The project is a joint development project between Nordic Mining's and Institute for Energy Technology's (IFE). Although the concept sounds similar to the concept presented above, no details of the process are given.

6 Case studies

The various embodiments of the concept have been assessed in more detail. Embodiment 1 was selected for a mass and energy balance study. Embodiment 3 is also discussed but in less detail. Embodiment 2 was not evaluated, since it is a minor refinement of Embodiment 1 attempting to reduce the energy requirements by utilising intercoolers for the compressor and turbine stages while employing the underground reaction chamber of Embodiment 3.

6.1 Methods

Mass and energy balances for the processes were calculated using Excel. For the calculations, it was assumed that the power plant supplying the flue gas was a modern supercritical (SC) pulverized-coal (PC) fired condensing power plant. The reference example by NETL (2010b; Case 11) provided data for the power plant and flue gas composition and physical attributes (Table 3).

Table 3. Flue gas data for a SC PC power plant with a net output of 550 MWe.

Parameter	Value
Flue gas flow	2137881 kg/h
CO ₂ concentration	13.5 vol-%
H ₂ O concentration	15 vol-%
Pressure	1 bar
Density	1.1 kg/m ³
Temperature	57 °C
Molar mass	28.86 g/mol
Operating hours	8000 (assumption)
CO ₂ emissions	3.36 Mt/a

To simplify the calculations, several assumptions were made. The compressors and expanders were assumed to work at isentropic conditions with an isentropic efficiency of 80 %. The specific heat capacity for the flue gas was calculated as an ideal gas and assumed to be constant, although CO₂ removal would affect this slightly. The condensation heat needed for the water in the flue gases was not calculated. The efficiency for the water pumps was assumed to be 85 %. The pinch temperature for the heat exchangers was set to an optimistic 10 K.

6.2 Assessment of Embodiment 1

Embodiment 1 presents a process, where CO₂ is captured from a flue gas stream using pressurised water and neutralised with feldspar (Figure 1). In the original embodiment a heat exchanger is placed in the flue gas stream before the flue gas enters the compressor. But since the compressor heats the flue gas stream and the scrubbing should be performed at as low temperatures as possible, the heat exchanger should be placed after the compressor instead. The corrected flow chart is shown in Figure 10, where also the solid material flows have been added.

Since Embodiment 1 requires the flue gas to be as cold as possible the scrubber system is inserted just before the flue gas enters the smoke stack. The flue gas (Figure 10, stream 1) passes through a series of compressors (simplified as one compressor) that pressurises the flue gas. The hot, pressurized flue gas (stream 2) is led into heat exchangers that cool the flue gas down close to the operating temperature of the pressurized spray tower, into which cold water is sprayed, removing most of the CO₂ in the flue gas. The CO₂-lean flue gas (stream 5) is led back to the main heat exchanger exchanging heat with the CO₂-rich flue gas. The pressurized and heated CO₂-lean flue gas (stream 6) is expanded in a turbine back to the entrance temperature (that of stream 1) and the mechanical power from the expander is used for driving part of the compressors.

The CO₂-rich water solution (stream 10) is passed into a neutralisation reactor where it is mixed with the crushed and fine-ground feldspar (stream 11). For simplifying the calculations, it is assumed that the neutralisation reaction proceeds according to Equation 1, making all the Al ions precipitate as gibbsite, Al(OH)₃, and all Si ions end up in silica in the neutralisation reaction. It is assumed that any unreacted material and silica can be removed from the reactor at this stage (stream 12), while the extracted aluminium ions flow with the solution to the settling pool, where gibbsite precipitate and can be removed (stream 13). The any CO₂ that has not reacted in the neutralisation reactor exits at this stage (stream 14).

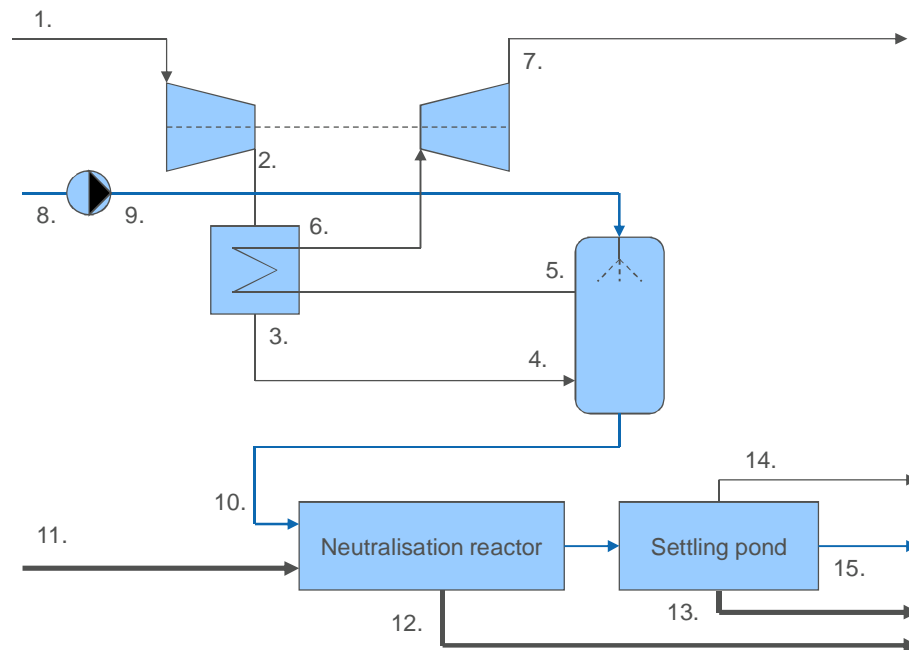


Figure 10. Embodiment 1 with added material streams and adjusted process equipment.

6.2.1 Assessment of the CO₂ capture stage

In order to assess the potential of the CO₂ capture stage of the concept, the neutralisation reactor is assumed to work ideally: the conversion is set to 100 % and the reaction is assumed to require no heating (i.e. takes place at 20 °C). These reactor parameters are completely unrealistic in view of the available experimental data, but are used here only for assessing the water capture step separately. It is also assumed that the feldspar source is 100 % Ca-plagioclase. In this case, all of the water could in theory be recycled, assuming that the extracted calcium ions precipitate as calcium carbonate.

It was assumed that the water entering the process is already at the same temperature as required by the scrubber. The scrubber is operated at 5 bar total pressure (0.68 bar CO₂) and ambient temperature (equilibrium capacity ~0.001 kg CO₂/kg H₂O at 20°C). Data on the performance of water scrubbing systems is scarce, but the examples provided by Kohl & Nielsen (1997) show that 95-99 % removal efficiencies are possible to achieve in commercial scale applications with

a CO₂ capacity of water close to the equilibrium condition. But the total pressures were typically higher (12-17 bar) and the CO₂ partial pressure also higher. Therefore, a CO₂ removal efficiency of 90% is assumed using the equilibrium carrying capacity of water.

Table 4. Summary of base case input parameters (above the middle line) and results (below the middle line).

Parameter	Value
Total pressure (bar)	5
Temperature (°C)	20
CO ₂ solubility (kg/t H ₂ O)	1.24
Capture efficiency	90 %
Water for scrubber (t/s)	94
CO ₂ captured (Mt/a)	3.0
Power requirem. (MW)	
compressor	224
expander	-101
water pump	44
	167
Power req. (MJ/t CO ₂)	1.6
Cooling requirements (MW)	48
Net power loss	31 %

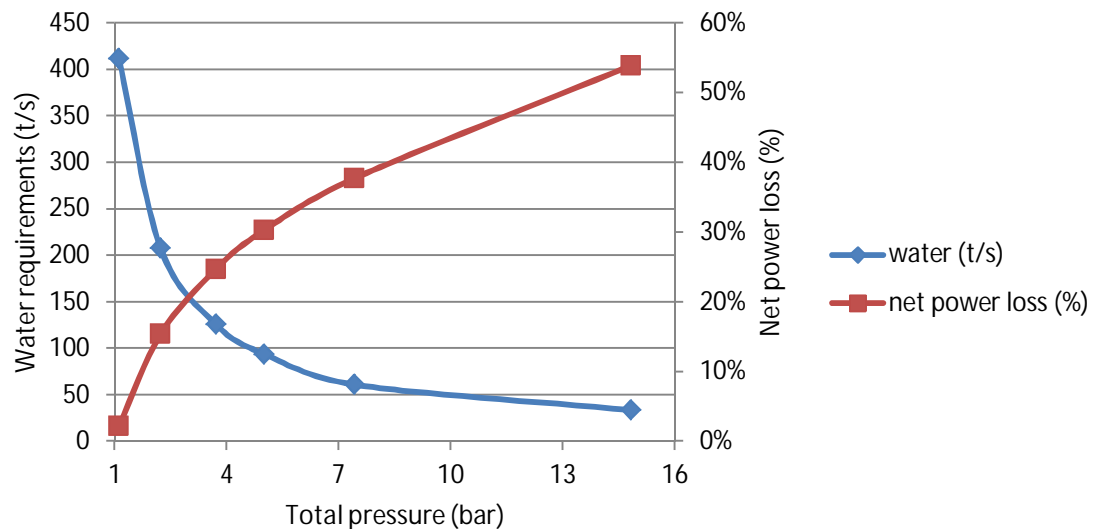


Figure 11. Scrubber water requirements and power loss as a function of pressure (at 20°C).

The results are summarised in Table 4 and Figure 11 – Figure 14. Table 4 summarises the base case input parameters and the calculation results. At these conditions, the net pressurisation and pumping power requirements would cause a drop of the net power output of the power plant with 31 %. However, at these conditions the low CO₂ solubility in water (only 1.24 kg/t H₂O) would require 94 tonne scrubbing water per second. The solubility grows with higher partial pressures of CO₂ and reduces the power need for water pumping, but increases the compressor power need, which leads to a net increase of the power requirements (Figure 11 and Figure 12). Lowering the temperature of the water entering the

scrubber will increase the CO₂ solubility in water, but due to the high compression power requirements in relation to pumping power it will not affect the power requirements much (Figure 13). Still, a higher CO₂ capacity will reduce the size of the scrubber and lower the investment costs.

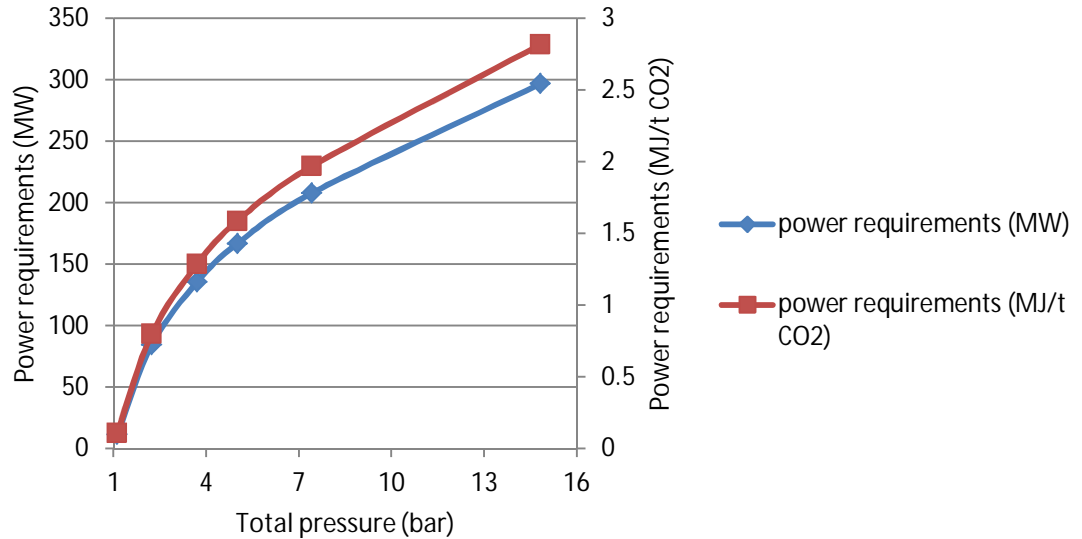


Figure 12. Power requirements as a function of pressure (at 20°C).

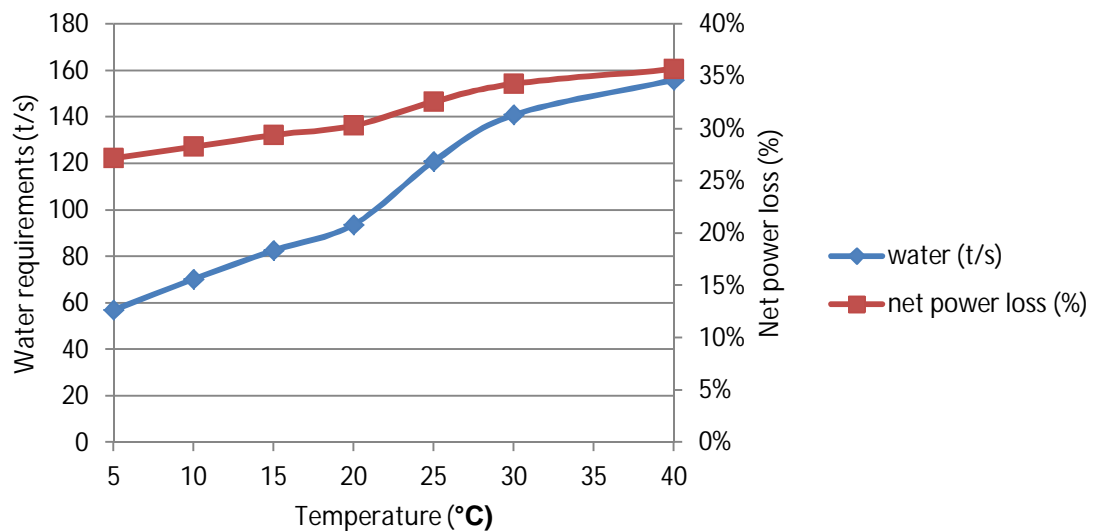


Figure 13. Scrubber water requirements and power loss as a function of temperature (at 5 bar total pressure).

However, this sensitivity analysis was performed assuming equilibrium CO₂ solubility of water. The actual CO₂ capacity is expected to be much lower. As can be seen in Figure 14, the lower the CO₂ capacity is the water and power requirements are.

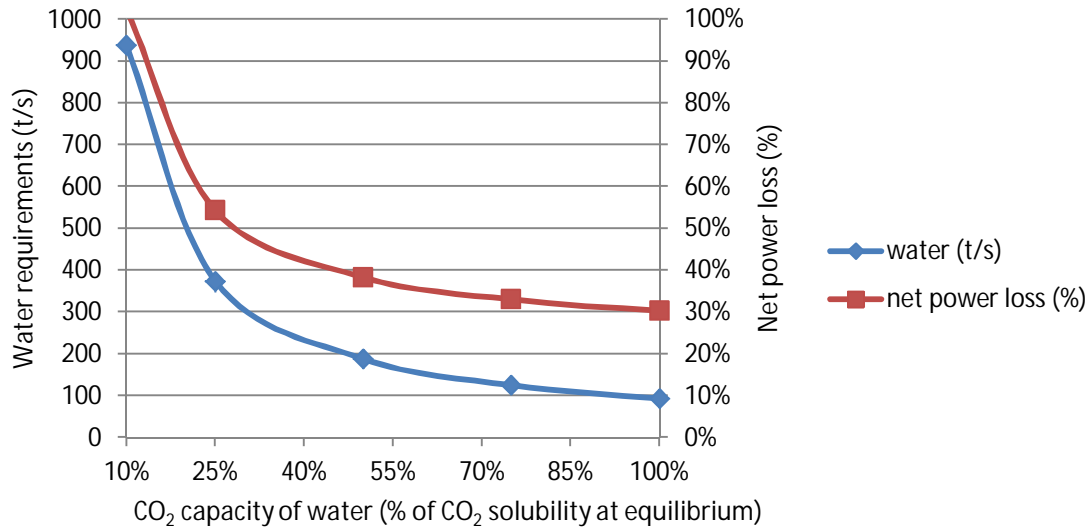
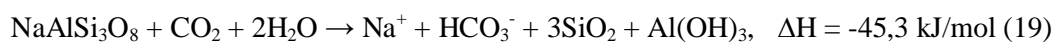
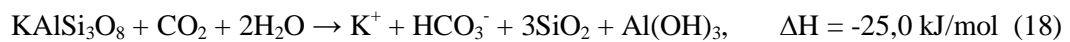
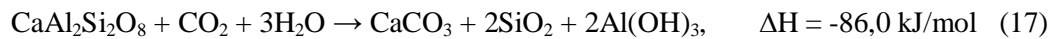


Figure 14. Impact of the CO₂ capacity of water upon scrubbing water requirements and the net power loss.

6.2.2 Assessment of the CO₂ neutralisation stage

After the CO₂ has been captured into the pressurized water stream, the CO₂ saturated water stream enters the CO₂ neutralisation reactor. To simplify the calculations it is assumed that the reaction with feldspar results in silica and gibbsite (formation energies calculated at 20 °C):



As mentioned above, in the base case it is assumed that the feldspar source is 100 % Ca-plagioclase. In this case, all of the water could in theory be recycled, assuming that the extracted calcium ions precipitate as calcium carbonate. However, as mentioned previously, the main problem is the slow reaction rate. All previous experimental data indicate that practically no reaction will occur at 20 °C. According to O'Connor et al. only 9 % conversion was achieved in 6 h at 185 °C and 150 bar partial pressure of CO₂. Assuming that the water flow was pressurised and heated to that level the requirements for the process power and heat requirements for the base case become completely utopic. However, to illustrate the impossible requirements for this process the data has been listed in Table 5. Note that the heat and power requirements come from the water heating alone and is not dependent upon the conversion of the mineral. The heat needed to heat the rock to the same temperature has not yet been taken into account, neither the power required to pressurize water and CO₂ nor the power required to grind the mineral, since these are negligible in comparison to the heating requirements for the water in Embodiment 1. Although the neutralisation reactions are

exothermic, even a 100% conversion of the material will only raise the heat of the reactor water by 0.2 °C due to the large flow through of water. Similarly, heating the reactor water with the flue gas exiting the expander (at ~200 °C) the reactor water will only be raised by 0.2 °C.

Since the heating requirements in the very optimistic base case will be 100 times the power output of the plant, it is evident that Embodiment 1 is unfeasible even at a theoretical level.

Table 5. Summary of base case input parameters (above the middle line) and results (below the middle line).

Parameter	Value
Neutralisation	10%
CO ₂ captured (Mt/a)	3.0
Temperature (°C)	185
Pressure (bar)	150
Feldspar requirements	57 t / t CO ₂
	192 Mt/a
Water heating requirements (MW)	~65000
w. complete heat recovery, 10K pinch (MW)	~3900
Pressurized reactor volume (m ³)	2000000

6.3 Assessment of Embodiment 3

In Embodiment 3 carbon dioxide under pressure, is injected into a reactor, filled with crushed rock and water. The reactor is according to the embodiment a fluidized bed that is located underground. After the reactor, the rock-water slurry is led to a settling pool, where the bauxite is separated.

As described above, the pressures required for neutralising feldspar with CO₂ are high – far over 100 bar. In order to have an overlaying rock burden thick enough to withstand that pressure the reactor would have to be located several hundred meters underground. For instance, the pressure gradient in sedimentary rock formations is typically approximated by calculating the hydrostatic pressure of water at a similar depth. That means that approximately a 1000 m thick sedimentary rock burden is necessary to achieve a pressure of 100 bar. Having a pressurized fluidized bed operating at this depth below ground is therefore not a feasible concept.

It is possible that rock feldspar located at this depth could be neutralised by injecting pressurized CO₂, but in this case the rock formation must meet the same requirements as those for geological storage of CO₂, i.e. high porosity and permeability, depth of more than approximately 800 m, overlaying cap rock formation, high enough storage capacity (millions of tonnes of CO₂) etc. As mentioned above, feldspar minerals occur in sedimentary rocks and their role in geological storage of CO₂ as a mineral trapping mechanism has been studied. But geological storage of CO₂ is a concept already being demonstrated at several parts of the world and is not the focus of the present study.

It is not specified in the embodiment whether the reactor is a batch reactor or a continuous reactor. A pressurized fluidized bed with the bed material consisting of rock it would likely be a batch reactor. However, preventing unreacted CO₂ from escaping the reactor during removal of spent feldspar residue and loading of fresh feldspar would be technically very difficult and costly to implement. Also, a large, pressurized fluidized bed reactor operating at 100 bar would be technically very challenging and extremely costly to implement. In this assessment, it is therefore assumed that the process is located above ground in a continuously operated pressurized reactor, such as a plug flow reactor. The technical implementation of this reactor is not addressed here further, but the material and energy requirements are assessed.

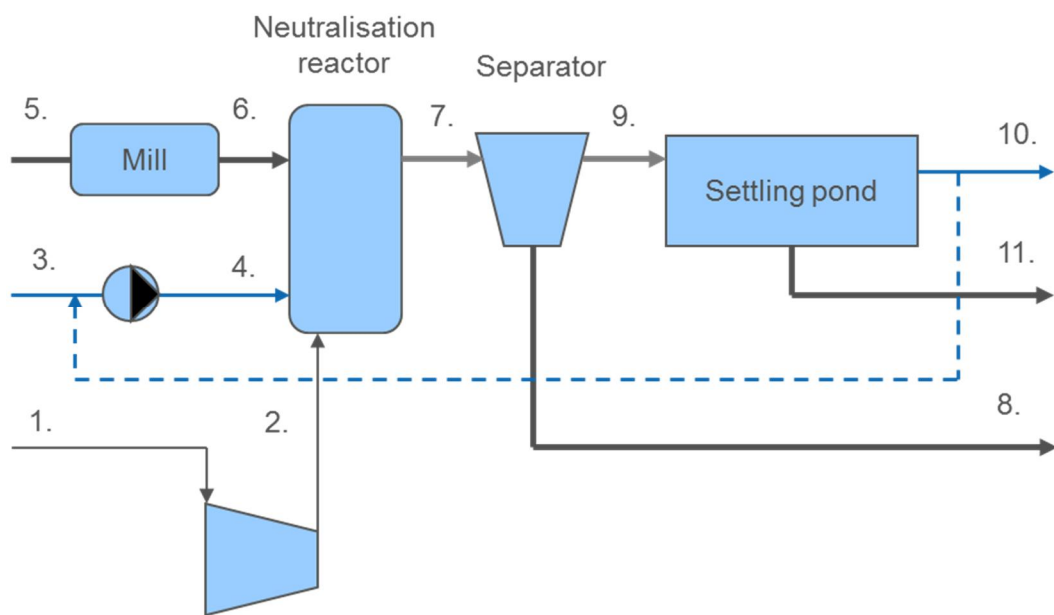


Figure 15. Modified process flow chart of the CO₂ neutralisation process.

In this case, the water requirements will be much less than that of Embodiment 1 and therefore the neutralisation process in this embodiment is assessed in more detail. Figure 15 shows the modified process flow chart of Embodiment 3. It is assumed, that CO₂ at 100 bar pressure (stream 1) is delivered from a capture facility (removal efficiency 90%) located at the same power plant as that used for the calculations in Embodiment 1. Therefore, it is assumed that the neutralisation plant is also operated 8000 h per year. Since the best available data for a neutralisation process using feldspar is the data presented by O'Connor et al. (2005) this data is used as a basis for the assessment (Table 2). The pressurized CO₂ has to be further pressurized up to 150 bar (2) and the incoming water (3) needs also to be pressurized up to 150 bar (4). The feldspar rock (5) is assumed to consist of 100% Ca-plagioclase in the base case and is ground from 10 cm lumps to fine powder (80% <37 μm) and fed to the reactor (6). The reactor is kept at a temperature of 185 °C. The reaction is assumed to proceed according to Equation 11. The reacted slurry needs to be fed to a separator, where the unreacted solid material and silica (8) is separated from the liquid (9). Gibbsite is allowed to

precipitate in a settling pond with calcite, from where it can be collected (11). Although O'Connor et al. used a water solution containing 0.64 M NaHCO₃ and 1 M NaCl it is for simplicity assumed here that all the water (10) can be recycled since Ca-plagioclase is assumed to not form any water soluble compounds. In practice, this would at least require some make-up chemicals as part of the chemicals are lost with moisture in the solid products. For simplicity, it is assumed that all CO₂ is consumed in the reaction and a neutralisation conversion of 10% is calculated so that 10 times more feed material is used to consume all CO₂ that is fed to the system.

6.3.1 Base case

The experimental data presented by O'Connor et al. (2005) was first used as input to determine the parameters for the base case. The results from the first calculation using O'Connor's experimentally verified data (Table 6, left column) show that the low feldspar conversion (neutralisation) combined with the relative low solid contents makes the concept unfeasible already from a material and energy balance view: both material requirements and heating requirements, mainly for water, are huge. Since the water is recycled, large savings in the water heating can be achieved. Assuming that the temperature of the water drops with 30 K during the separator step and settling pond step is very optimistic, but shows that the heating requirements can be cut back if water is recycled. The pumping power required for water could be reduced as well, if the water could be kept pressurized throughout the process. However, this would require pressurized

Table 6. Summary of base case input parameters (above the middle line) and results (below the middle line).

Parameter	O'Connor	Base case
Neutralisation	10%	50%
CO ₂ captured (90 % efficiency)	3.0 Mt/a	3.0 Mt/a
Temperature	185 °C	185 °C
Pressure	150 bar	150 bar
Solid content (mass basis)	15%	50%
Reaction time	6 h	6 h
<hr/>		
Feldspar requirements; stream 5 (per t CO ₂)	57 t	11 t
	192 Mt/a	38 Mt/a
Feedwater; stream 3 (recycled)	1085 Mt/a	77 Mt/a
Power for grinding	643 MW	129 MW
Power for pumping water	660 MW	23 MW
<u>Power for pumping CO₂</u>	<u>1.1 MW</u>	<u>1.1 MW</u>
Total power consumption	1305 MW	153MW
Power loss for power plant	237 %	28%
Heat from reaction (as negative value)	-206 MW	-206 MW
Heating requirements for CO ₂	38.5 MW	38.5 MW
Heating requirements for rock	973 MW	195 MW
(Heating requirements for water	~26000 MW	~917 MW)
<u> w. complete heat recovery, 30K losses</u>	<u>4720 MW</u>	<u>167 MW</u>
Net heat consumption	5531 MW	194 MW
	63 MJ/kg CO ₂	1.8 MJ/kg CO ₂
Pressurized reactor volume	~960000 m ³	60000 m ³

separation of the by-products and unreacted feldspar, which would add to the investment costs.

However, the experimental conditions were not optimised for feldspar and it is possible that a higher feldspar conversion can be achieved by varying the process parameters. For simplicity, the same process parameters are assumed, but the conversion is raised to 50% for the base case. Since it is highly likely that the solid to liquid ratio can be raised also in a plug flow reactor, the solid to liquid ratio is set to 50% for the base case. This lowers the power and heat consumption of the neutralisation reactor somewhat (Table 6). The consumption is still too high, since a typical CO₂ capture facility alone will reduce the power output of the power plant with 20-30 %.

6.3.2 Sensitivity analysis

The comparison in Table 6 already shows that a high feldspar conversion and high solid to liquid ratio is essential for bringing down the power and heat requirements. It is possible that chemical additives can speed up the reaction further, but the additional expenses are likely to make the process economically unfeasible.

In order to see if there is a window of feasibility, if the conversion could be improved, the power and heat requirements were plotted as functions of various parameters (Figure 16 – Figure 20). Figure 16 show that feldspar conversion has a high impact on both power requirements and heat requirements. A higher conversion means that less feldspar is needed, which leads to reduced power requirements for grinding and water pumping as well as the less heating requirements for rock and water.

Figure 17 and Figure 18 show that a higher solid content in the reactor helps to reduce the heat requirements of the significantly, since less water needs to be heated. However, the power requirements are not reduced as drastically, because for a solid content is increased over 15 % the pumping power for CO₂ and water combined becomes is lower than the grinding power, regardless of the feldspar conversion.

As Table 6 shows the largest part of the power consumption in the base case comes from the mill. The results in Figure 19 show that the power consumption from milling could be significantly reduced, if the neutralisation would work with a larger particle size. However, this is highly unlikely, since reactivity is usually directly related to the particle size, i.e. larger particles have a smaller surface area per gram material and the conversion is therefore slower. The grinding power has been calculated by assuming that the power required to grind the feldspar used is similar to that of granite. If the power required would be similar to grinding limestone, the power need would be only one third lower.

In Figure 18 the conversion was fixed at 75%, which leads to a negative heat requirement (i.e. cooling requirements instead of heating) for solid contents higher than 75%. However, for such a fine powder material as in this case it is likely that the solid content of the slurry needs to be relatively low in order to prevent the

slurry from clogging up the reactor. Negative heat requirements means that the heat of reaction is higher than the requirements for heating the incoming feed streams up to the reaction temperature. Similarly, it can be seen from Figure 20 (left image) that in the base case heating would only be needed for a reactor temperature of 50 °C and higher. The CO₂ transported from the capture facility is already in a supercritical state at a pressure of 100 bar and further pressurisation requires only pumping power. Therefore, the reactor pressure (Figure 20, right image) has a relatively small impact on the energy requirements.

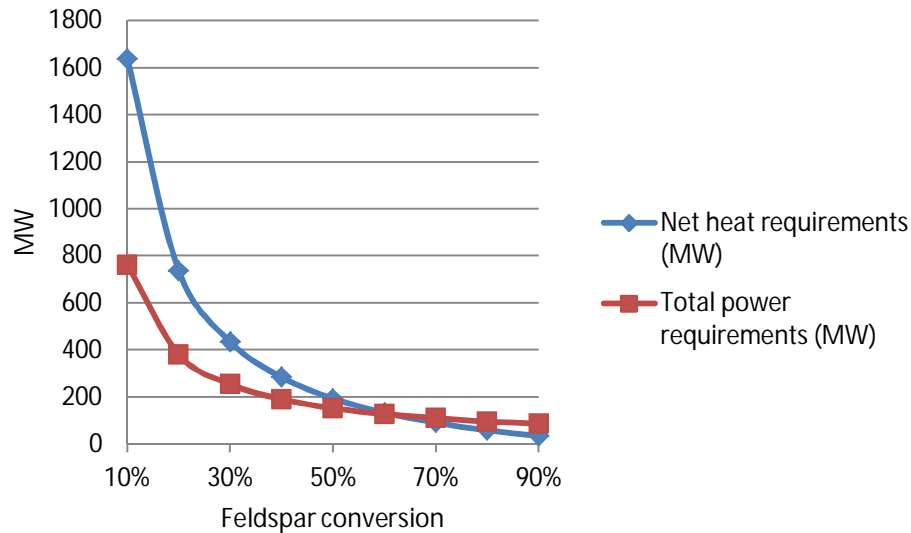


Figure 16. Power and heat requirements as a function of feldspar conversion. Other parameters according to the base case in Table 6.

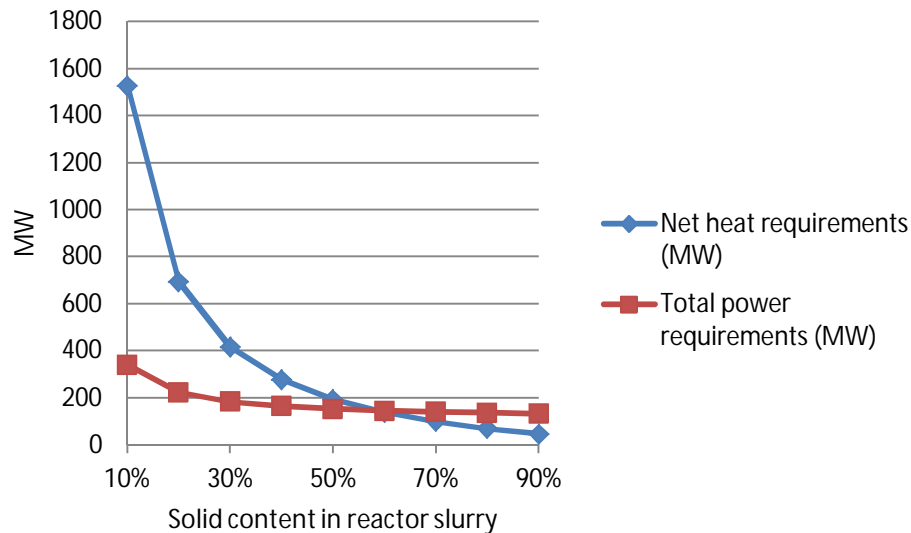


Figure 17. Power and heat requirements as a function of solid content when conversion is set to 50 %. Other parameters according to the base case in Table 6. °

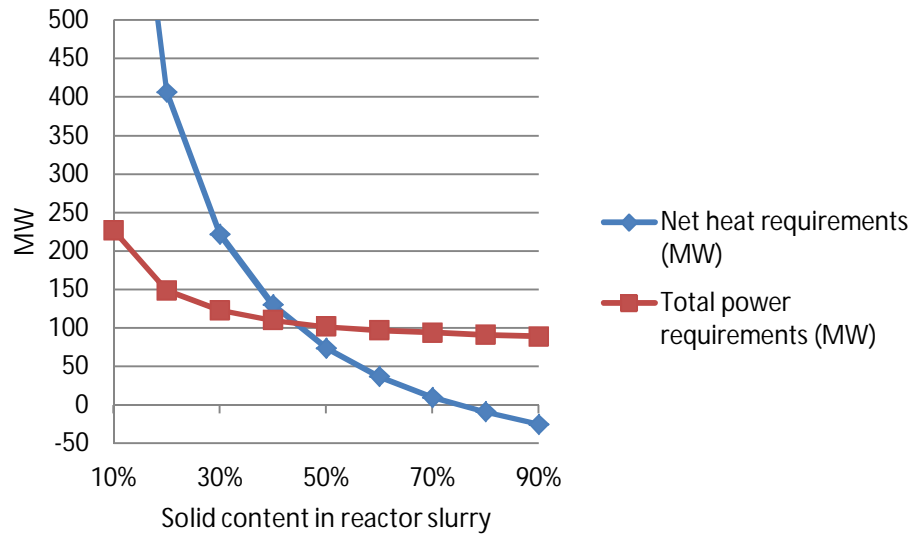


Figure 18. Power and heat requirements as a function of solid content when conversion is set to 75 %. Other parameters according to the base case in Table 6.

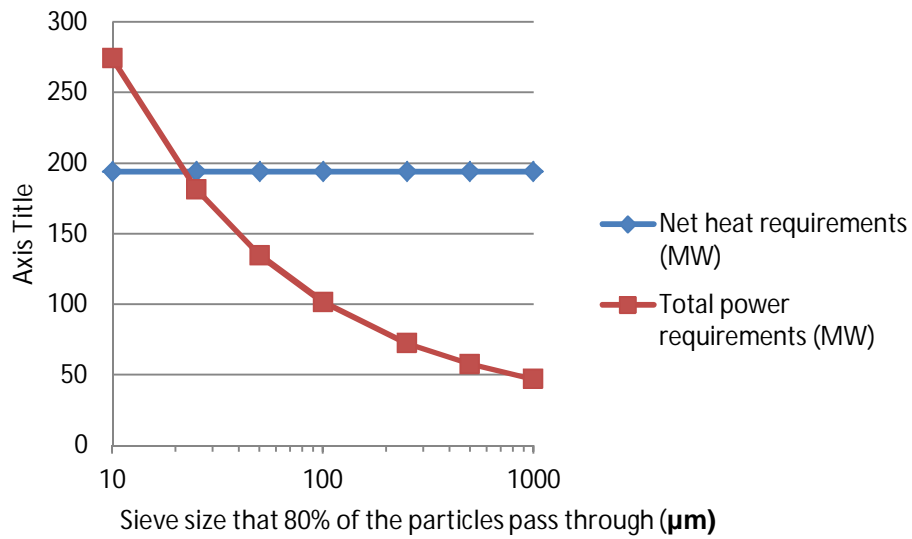


Figure 19. Power and heat requirements as a function of the particle size of milled feldspar as fed to the reactor. Other parameters according to the base case in Table 6.

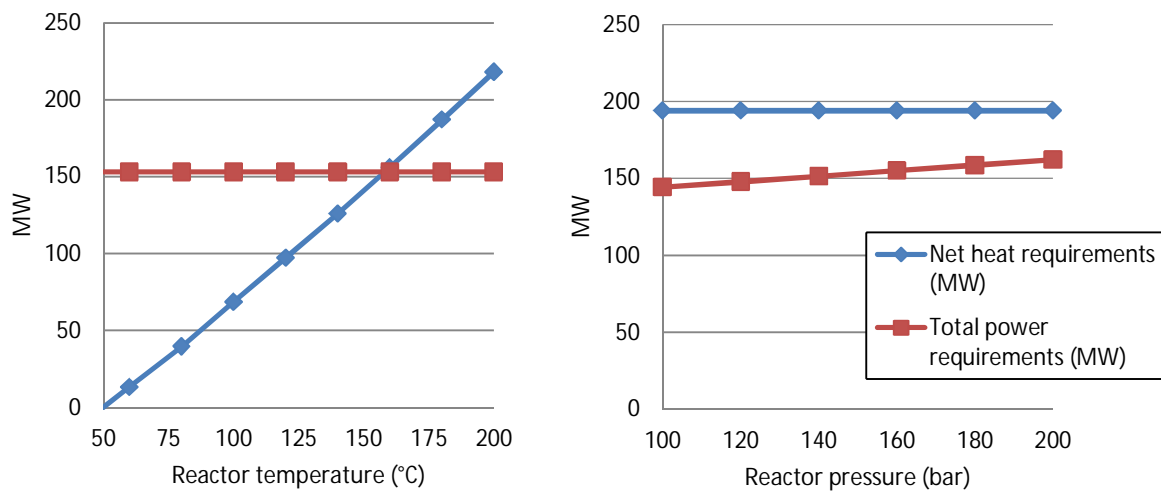


Figure 20. Power and heat requirements as a functions of reactor temperature and pressure. Other parameters according to the base case in Table 6.

7 Feasibility of the CO₂ neutralisation concepts

7.1 Technical feasibility

As the literature review and case calculations above show, the bottleneck of the process is the slow reactivity of feldspar with CO₂. Although publicly available experimental data on feldspar-CO₂ reactivity in conditions relevant for an industrial process (i.e. process parameters allowing a significant conversion in hours) are few, all available data show that very demanding process conditions are required (CO₂ pressure of >100 bar, very fine ground rock material, several hours residence time in reactor) to achieve any notable conversion. Even at these conditions, the reported conversions have been low: less than 10% conversion.

Embodiment 1 makes an attempt at an integrated CO₂ capture and neutralisation concept using pressurized water for absorbing CO₂ flue gases and feldspar for neutralising the CO₂-saturated water. However, these two concepts seem to be incompatible, because CO₂ absorption requires large amounts of cold water, which needs to be heated to temperatures over 100 °C in order for the neutralisation process to work even at remotely acceptable reaction rates. The excessive heat requirements for heating the cold water makes Embodiment 1 therefore unfeasible in practice.

The modified version of Embodiment 3 assessed here presents a “best-case scenario” for the CO₂ neutralisation concept, but although the heat and power requirements are less than for Embodiment 1 since less water is needed the requirements are still very large. Using experimentally verified data the power requirements of the process exceeds over twice the power output of the power plant, from which the CO₂ is neutralised. Also, the amount of feldspar needed due to the low conversion (10 %) is unacceptable. Although the sensitivity analysis show that significant improvements can be achieved if a more reactive material is

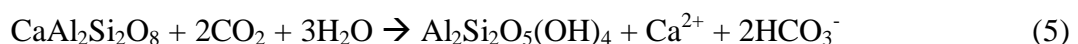
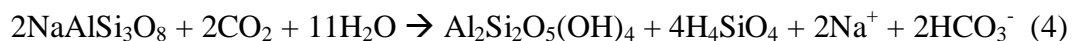
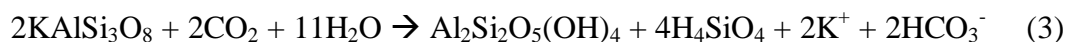
used, the neutralisation process would still require a separate CO₂ capture unit at the power plant for supplying CO₂ to the neutralisation process. Also, this type of CO₂ neutralisation concepts, where pressurized CO₂ is reacted with a slurry of mineral and water to fix CO₂ as a carbonate mineral, has been studied extensively in the U.S. for magnesium and calcium silicates (Table 2; O'Connor et al., 2005). The lowest costs reported from a feasibility study regarding storing CO₂ using this method were 54 US\$/tCO₂ with olivine as feedstock, 64 US\$/tCO₂ with wollastonite as feedstock, and 78 US\$/tCO₂ using serpentine as feedstock, excluding CO₂ capture and transportation costs (O'Connor et al., 2005; Gerdemann et al., 2007). The focus of the research has since then migrated towards geological storage of CO₂, since this is still the most cost effective method for storage of CO₂.

7.2 Scale of operation

The size of operation for a CO₂ neutralization plant would be on a similar scale of other large-scale mining operations. To illustrate this, the Talvivaara Sotkamo mine is used here as an example. The Talvivaara Sotkamo mine is located at one of the largest known nickel sulphide deposit in Europe, estimated to 1,550 Mt ore containing 0.22% of nickel, 0.49% of zinc, 0.13% of copper and 0.02% of cobalt (Talvivaara, 2012). The planned scale of the mine is to process 24 Mt of ore annually. In 2010, 16.7 Mt of rock was mined. The size of a mine that would produce enough feldspar for neutralising CO₂ from a 550 MWe coal-fired power plant, as used in the calculations above, would require a mine of similar scale, assuming that most of the rock mined is feldspar.

7.3 By-products purity

The CO₂ neutralisation concept is being promoted as a process that allows for both production of valuable minerals (silica and alumina) and neutralisation of CO₂. However, it is not clear in what form the minerals are after the processing of the feldspars with CO₂. According to the chemical weathering process occurring in nature, kaolinite as well as solute silica and sodium/potassium/calcium carbonate:



As mentioned previously, both solute silica and calcium carbonate are expected to precipitate as well. However, it is unclear how well silica, calcium carbonate and kaolinite could be separated after formation. Rajamäki (2011) made a preliminary evaluation of the business potential of the concept, but the potential is evaluated assuming that the byproduct quality is of commercial quality. According to previous experience in the field of mineral carbonation, the product from an aqueous, pressurized reactor will be a mixture of both unreacted mineral and solid products.

Hangx and Spiers (2009) analysed the product from the CO₂-plagioclase-water experiments at 200–300 °C, using carbon dioxide pressures from 4–150 bar. They reported formation of clays (kaolinite and smectite or illite), boehmite, and a Mg, Ni, Fe–hydrotalcite-like phase. No silica and very little carbonates were reported. Although kaolinite and boehmite are minerals that are typically part of bauxite ore, it is not certain that the quality would be sufficient for aluminium processing. The alumina content of commercial bauxite ores ranges typically from 30%–65%. Refractory grade bauxite (essentially all diasporic, i.e. α -AlO(OH)) can cost as much as 400–500 USD/t (Industrial Minerals, 2011), but bauxite ore used for aluminium production is typically around 20–30 USD/t (CapeAlumina, 2011). The prices depend on the alumina content and the type of bauxite (for example gibbsite, being easier to process), with deductions for reactive silica content. Silica sand is typically about \$20–26/t. If the resulting material is being used for earth works or deposited as waste it has to meet certain regulatory standards. More on waste regulations of interest for this concept can be found in Huttunen (2009).

7.4 Economic feasibility

7.4.1 Embodiment 3

Based on the assessment made for the modified process of Embodiment 3 and using the number presented in Chapter 7.2 a coarse breakdown of the operating costs can be made based on the annual costs for power, heat, and rock material, taking into account revenues from gibbsite and CO₂ emission trading. Table 7 shows the input parameters as well as the results from the calculations. It was assumed that silica would be mixed with the reacted feldspar and would likely not have an economic value. The calculations were performed based on the two cases presented in Table 6. As can be seen from the table, in the case based on real experimental data the operational costs are almost twenty times the revenue. Using the optimistic base case data the operational costs come within range of the revenue. It can be seen from the table that the cost for feldspar and revenue from gibbsite largely dictate the operational costs for the base case, while the revenue from CO₂ trading is negligible. Therefore, the assumptions used for feldspar cost and gibbsite revenue has also a large impact. Using these input parameters, an operational break-even points (i.e. annual costs = 0) were calculated for various feldspar conversions and solid contents of the slurry using two different reactor temperatures. The results (Figure 21) show that in order for the revenue to break even with the operational costs a fairly high feldspar conversion and solid content is required.

However, in addition to these a high investment cost is needed. A pressurized plug flow reactor over 60000 m³ is likely to be technically very challenging to make and probably very expensive. It is also unclear how the alumina compounds can be separated from the feed material in practice and what the quality and economic value of such alumina compounds would be. Considering the results from these calculations, it is highly likely that feldspar is not reactive enough for making the CO₂ neutralisation concept even remotely feasible.

Table 7. Breakdown of the economic feasibility based on main annual operational costs and revenues.

Input parameters		Results	O'Connor	Base case
Power	0.08 €/kWh	Power	835 M€/a	98 M€/a
Heat	0.08 €/kWh	Heat	3540 M€/a	124 M€/a
Feldspar	10 €/t	Feldspar	1915 M€/a	383 M€/a
Gibbsite	30 €/t	Gibbsite	-322 M€/a	-322 M€/a
CO ₂ (EU ETS ave. price) ¹	10 €/t	CO ₂	-30 M€/a	-30 M€/a
		Total costs	5937 M€/a	253 M€/a

¹These calculations do not include the costs related to CO₂ separation, pressurisation and transportation, which would add about 40-100 €/t CO₂ to the operational costs. The ETS price has been included mainly for showing the cost in relation to the current CO₂ trading price.

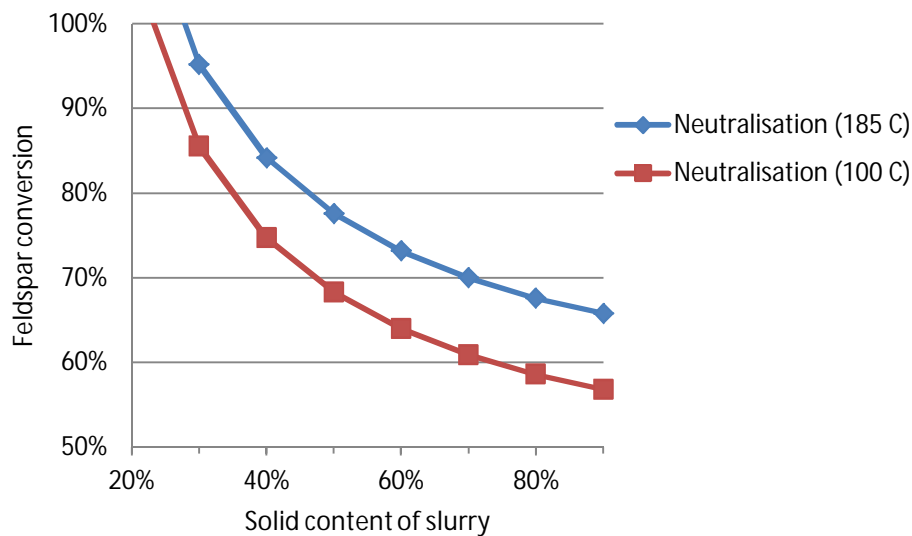


Figure 21. Break-even points for different values of the solid content and feldspar conversion, using two different reaction temperatures.

7.4.2 Alternative process

Using the commodity price list for minerals in Industrial Minerals (2011) and that for chemicals in ICIS (2012), the economic feasibility of the alternative process presented in Chapter 5.3 can be preliminarily assessed based on the cost for the chemicals consumed. The base assumption is 80% conversion of all reactions (see Chapter 5.3) and the use of nitric acid is assumed, since the ammonium nitrate formed could possibly be sold. As the results in Table 8 show, even when assuming that all products would have a high enough purity to be sold the cost for the chemical consumption is higher than the estimated incomes from the products. It must be pointed out that various additional filtration and purification processes are likely to be needed in order for the products to have a commercial grade quality, which would add to the operational costs. Also, any investment costs have not either been taken into account, so this concept does not either seem

economically feasible. In order to make the concept feasible, the solvent would need to be of a different type that could be recycled and reused in the process.

Table 8. Estimation of the economic feasibility based on commodity costs and revenues.

Commodity	Unit cost	Amounts	Cost
Nitric acid	220 USD/t ¹	54 Mt/a	9385 M€a
Feldspar	10 €/t	30 Mt/a	299 M€a
Ammonia	600 USD/t	12 Mt/a	5532 M€a
Silica	~100 €/t	10 Mt/a	-1000 M€a
Ammonium nitrate, fertilizer grade	170 USD/t	55 Mt/a	-7370 M€a
Gibbsite	30 €/t	13 Mt/a	-402 M€a
Precipitated calcium carbonate	~100 €/t	7 Mt/a	-5506 M€a
CO ₂ (EU ETS ave. price)	10 €/t	3 Mt/a	-30 M€a
Net cost			908 M€a
			300 €/t CO ₂

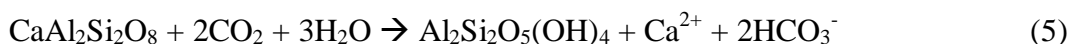
¹EUR/USD average in 2006 = 1.27

7.5 Deposition of bicarbonate water

The largest environmental issue of the CO₂ neutralisation process is likely to be related to the large amounts of bicarbonate water that has to be deposited of, in case K-feldspar or Na-feldspar is used. Even if K-feldspar or Na-plagioclase could be converted to 100% at a solid content of 50% in the reactor, the water consumption would be one tonne per tonne feldspar. This means that for a 550 MWe coal-fired power plant (as used in the calculations above) a minimum of about 20 million m³ of bicarbonate water would need to be dumped in the sea each year.

As mentioned in the introduction, recent legislations and regulations in Europe has forbidden placement of CO₂ into the water-column of the sea and on the seabed, because of the potential negative effects (Ospar, 2007). Although it is very improbable that dumping of large amounts of calcium, potassium and sodium bicarbonate waters into the sea would be allowed, it may also be equally challenging to acquire the feedwater for the process.

In order to minimize the use of water and eliminate the need for dumping of bicarbonate waste water, the neutralisation of CO₂ could be restricted to using only Ca-plagioclase (mostly CaAl₂Si₂O₈) as raw material. In this case, the reactor conditions could be controlled to enable precipitation of the bicarbonate solution as solid calcium carbonates:



Both potassium carbonates and sodium carbonates are water soluble and precipitation is therefore not possible with Na-plagioclase or K-feldspar. Since Ca-plagioclase is the most reactive of the feldspars it is also the most likely candidate for a CO₂ neutralisation process. However, storing CO₂ as carbonates instead of bicarbonates requires twice the amount of minerals, as the stoichiometry of the reactions above shows.

CO₂ neutralisation of feldspar could possibly mobilise other valuable metals as well, but these are likely to be released in concentrations too low for economic recovery. However, these trace elements would still need to be removed for enabling recycling or deposition of the used process waters.

8 Conclusions

Based on the assessment above, the following conclusions can be drawn from this assessment.

- None of the CO₂ neutralisation concepts assessed were found economically feasible, not even if all the mineral by-products could be sold.
- The slow reaction kinetics of feldspar with CO₂ is the main bottleneck of the CO₂ neutralisation concept. The scarce experimental data available indicates that pressures over 100 bar CO₂ and temperatures between 100-300 °C are required and the highest verified conversion was as low as 9% of the theoretical maximum in 6 h. The completion of the reaction is therefore expected to take days, which is far too slow for an industrial process, due to the gigantic pressurized reactor volume. Therefore, neutralisation of feldspar with CO₂ was found both technically and economically unfeasible.
- CO₂ capture with water is a known technology, but integrating water-capture of CO₂ with CO₂ neutralisation makes the energy economics of the process worse: CO₂ capture with water requires large amounts of water, while in order to minimize the heat requirements for the CO₂ neutralisation reactor the water usage needs to be minimized.
- Known deposits of feldspar contain enough rock for large-scale CO₂ neutralisation operations at least in U.S. and Norway. It is questionable if deposits in Finland have enough resources with sufficient feldspar concentrations.
- The water consumption and the need for dumping of millions of tonnes of bicarbonate waste water is another hurdle for the process. Due to new European legislations that forbid dumping of CO₂ in the sea water it is likely, that K-feldspar and Na-plagioclase cannot be accepted for CO₂ neutralisation, since neutralisation of these result in a liquid bicarbonate solution. However, this does not apply to neutralisation of Ca-plagioclase (as found in anorthosite). Neutralisation of Ca-plagioclase results in solid calcium carbonates that could be filtered out and the process water could be recycled.

- For aluminium ore production, the studied CO₂ neutralisation processes were also found too expensive. An alternative process for processing feldspar into aluminium ore by using mineral acids was also assessed, but although technically feasible and less demanding to perform than the CO₂ neutralisation concepts, it was still found economically unfeasible due to the large consumption of chemicals.

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Appendix A: Process calculation sheet for Embodiment 1, with input data set for base case

White cells: input data
 Grey cells: data from NETL(2010b), Case 11.

Molar volume of a gas	
V_m0	22.71
V_m	0.0274 m ³ /mol
V_CO2	262376 m ³ /h

Compressor	
n	1.4
R	8.3145 J/molK
R_fg	0.288 kJ/kgK
n_is.eff	80 %
p2	5 bar
pCO2	0.675 bar
T2s	355 C
T2	430 C
Cp	1.0085 kJ/kgK
w_comp	376.1 kJ/kg
P_comp	223351 kW

Heat exchangers	
T4 = T5 = T8	20 C
m5,6,7	1759358 kg/h
	489 kg/s
	14.1 Mt/a
T_pinch	10 K
T6	420 C
W_hx_23	197113 kW*
T3	101 C

Turbine	
T7s	164 C
T7	216 C
P_turb	100735 kW

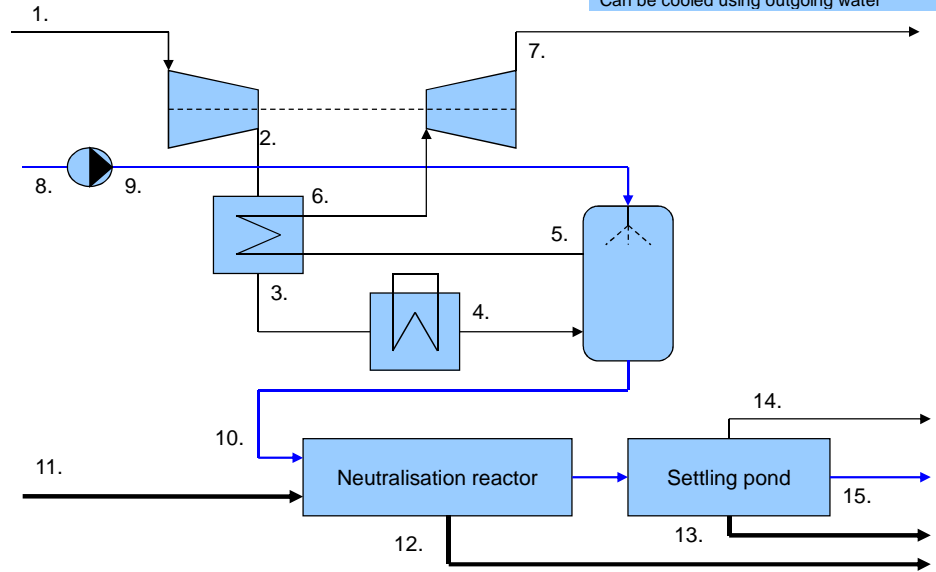
Flue gas	
CO2	13.5 % vol
T1	57 C
p1 = p7	1 bar
dens	1.1 kg/m ³
m1,2,3,4	2137881 kg/h
	594 kg/s
	17.1 Mt/a
M_fg	28.86 g/mol
V	1943528 m ³ /h
m_CO2	420581 kg/h
	117 kg/s
Op. hours	8000 h/a
CO2 em.	3.36 Mt/a

Scrubber	
n_scrub	90 %
m10	337923783.5 kg/h
	93868 kg/s
	2703 Mt/a

Net power requirements	
P_capt	166740 kW
	1.59 MJ/kg CO2
P_plant	549990 kW
P_plant_net	383250 kW
P_penalty	30.3 % net power output loss
W_hx_34	48398 kW cooling required*
	0.46 MJ/kg CO2

*Can be cooled using outgoing water

Water	
at 5 bar	0.001246 kg CO2/kg H2O
m8,9	337545260 kg/h
	93763 kg/s
	2700 Mt/a
T8 = T9	20 C
p8	1 bar
p9 = p2	5 bar
n_pump	85 %
dens_H2O	1000 kg/m ³
W_pump	44124 kW



Waste rock, Ihalainen mine	
m11	2393471 kg/h
	665 kg/s
	19.1 Mt/a
	5.7 t/CO2
Composition	
KAlSi3O8	0 %
CaAl2Si2O8	100 %
SiO2	0 %
Other	0 %
Sum	100 %
m_kfeld	0.0 Mt/a
m_cafeld	19.1 Mt/a

Neutralisation	
$CaAl_2Si_2O_8 + CO_2 + 3H_2O \rightarrow CaCO_3 + 2SiO_2 + 2Al(OH)_3$ $KAlSi_3O_8 + CO_2 + 2H_2O \rightarrow K^+ + HCO_3^- + 3SiO_2 + Al(OH)_3$	
n_neutr	100 %
Temp up to boiling point of water, practically <90 C	

Heat of reaction	
H_Ca	205511 kW
H_K	0 kW
tot	205511 kW
Reactor:	20.5 C

CO2 neutralized	
Gross	3.03 Mt/a
gross eff.	90 %

Byproducts and wastes	
m12	8.3 Mt/a unreacted & SiO2
m13	10.7 Mt/a gibbsite, Al(OH)3
	6.9 Mt/a calcite, CaCO3
m14	0.00 Mt/a CO2
m15	2697 Mt/a water out

Heating of reactor water with flue gases	
W_hx	78142 kW
Cp_H2O	4.18 kJ/kgK
T*10	20.2 C

Mass balance delta	
neutr.react	0.00
system	0.00

Heating requirements	
W_add	392367 kW per 1 C
T pinch	10 C
	3923671 kW

Appendix B: Process calculation sheet for Embodiment 3, with input data set for base case

White cells: input data

Grey cells: data from NETL(2010b), Case 11.

Molar volume of a gas

V_m0	22.71
V_m	0.0274 m ³ /mol
V_CO2	262376 m ³ /h

Flue gas

CO2	13.5 %	vol
T1	57	C
p1 = p7	1	bar
dens	1.1	kg/m ³
m_fg	2137881	kg/h
	594	kg/s
	17.1	Mt/a
M_fg	28.86	g/mol
V	1943528	m ³ /h
m_CO2	420581	kg/h
	117	kg/s
Op. hours	8000	h/a
CO2 em.	3.36	Mt/a
capt_eff	90 %	
m1,2	3.03	Mt/a
p1	100	bar

Water

Solid content	50 %	
m3,4	4786943 kg/h	
	1330 kg/s	
	38 Mt/a	
T8 = T9	20	C
p8	1	bar
p9 = p2	150	bar
n_pump	85 %	
dens_H2O	1000	kg/m ³
W_pump	23309	kW
Cp_H2O	4.18	kJ/kgK

Waste rock, Ihalainen mine

m5,m6	4786943 kg/h
	1330 kg/s
	38.3 Mt/a
	11.4 t/tCO2
cp_albite	0.233 kJ/molK
	0.887 kJ/kgK
Composition	
KAISi3O8	0 %
CaAl2Si2O8	100 %
SiO2	0 %
Other	0 %
Sum	100 %
m_kfeld	0.0 Mt/a
m_cafeld	38.3 Mt/a

CO2 pump

n	1.4
R	8.3145 J/molK
R_fg	0.288 kJ/kgK
n_eff	75 %
p2=p	150 bar
d_CO2	630 kg/m ³
cp_CO2	2 kJ/kgK
P_comp	1113 kW

Mill

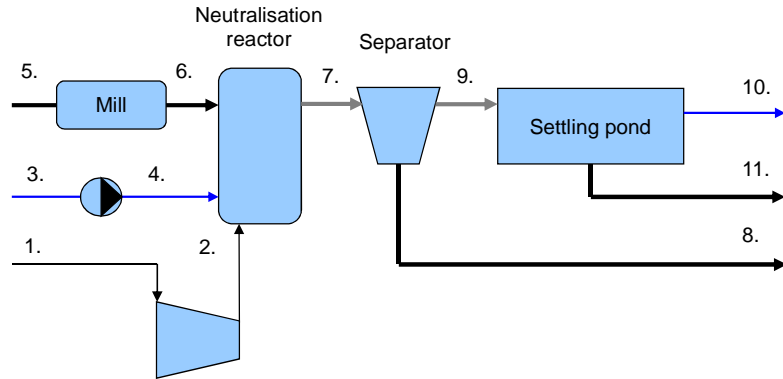
Feed	100000	um
Product	37	um
W_index	15.13	(granite)
W_Bond	24.39511417	kWh/ts
	26.89648751	kWh/t
	96827	kJ/t
P_mill	128752	kW

Power consumption

P_tot	153174 kW
	2.91 MJ/kg CO2
P_plant	549990 kW
P_plant_net	396816 kW
P_penalty	27.9 % net power output loss

Heat consumption

Q_tot	194382 kW heating needed
	1.85 MJ/kg CO2



Neutralisation

T	185	C
p	150	bar
t	6	h
$\text{CaAl}_2\text{Si}_2\text{O}_8 + \text{CO}_2 + 3\text{H}_2\text{O} \rightarrow \text{CaCO}_3 + 2\text{SiO}_2 + 2\text{Al}(\text{OH})_3$		
$\text{KAISi}_3\text{O}_8 + \text{CO}_2 + 2\text{H}_2\text{O} \rightarrow \text{K}^+ + \text{HCO}_3^- + 3\text{SiO}_2 + \text{Al}(\text{OH})_3$		
n_neutr	50 %	

Byproducts and wastes

m8a	27.4 Mt/a unreacted & SiO2
m8b	6.9 Mt/a calcite, CaCO3
m11	10.7 Mt/a gibbsite, Al(OH)3
m10	35 Mt/a bicarbonate water

Heat of reaction

H_Ca	205511 kW	
H_K	0 kW	
Q_react_tc	205511 kW	
delta_T	30 K, assumed heat loss	
Q4	-166745	-817088 kW
Q6	-194625 kW	
Q2	-38553 kW	
	-194382	-1150277 kW

Reactor volume	59967 m ³
mass balance delta	0.00