



TAMPERE UNIVERSITY OF TECHNOLOGY

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EVALUATION OF A METHOD FOR “CARBON CAPTURE AND
NEUTRALIZATION”

ABSTRACT

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23 pages, 4 Appendix pages

August 2011

Keywords: CCN, CCS, carbon dioxide, (rock) weathering, (rock) mineral, feldspar, aluminum, mineral carbonation

Carbon dioxide has for long been considered to be the most important greenhouse gas causing global warming and climate change, and therefore extensive demand for Carbon Capture and Storage (CCS) (methods) has been arisen. Possibly the most challenging task in CCS is to assure the reasonable and long-term safe storage of captured carbon dioxide. A new, path-breaking application of CCS, Carbon Capture and Neutralization (CCN), an invention by Matti Nurmi, provides a new alternative for solving the problem. In the CCN method, carbon dioxide is transformed into bicarbonates that can be released into natural waters and no storage is thus needed. The transformation is based on neutralizing the carbon dioxide with rock minerals, imitating natural rock weathering.

The objective of this report is to evaluate the feasibility of the CCN method. The theoretical neutralization capacities of the most appropriate minerals are studied. A few examples of estimated mass flows in neutralization of one ton of carbon dioxide by rocks with known mineral compositions are presented as well, combined with rude cost approximations.

The basic idea of the CCN method is to dissolve carbon dioxide into water and then neutralize the acidic solution by passing it through neutralizing minerals. The possible applications of the method are numerous. In addition to bicarbonates, the neutralization produces valuable by-products, such as aluminum compounds and silicon oxide that can be sold to partially cover the neutralization costs. This is precisely what makes CCN profitable compared to other CCS methods. The minerals suitable for neutralization can be found in common rocks, and their acquisition should thus be relatively simple.

In order to get a better idea of the actual feasibility of CCN, neutralization reactions have to be studied in detail. Especially the reaction speed and the completeness of the reactions should be observed. Furthermore, environmental risks and costs have to be examined in depth as well. The use of both laboratory and pilot scale test reactors would be reasonable in further research. Since the first pilot plants are already about to be built abroad in South Africa and Botswana, rapid actions for further research are necessary in order to get the best results from the CCN method.

PREFACE

The following text is a summary of the “Carbon Capture and Neutralization” report written during a summer employment in 2011 at the Department of Energy and Process Engineering at Tampere University of Technology. It also contains some additional cost estimations. The report is a part of the Carbon Capture and Storage Program (CCSP) of the CLEEN Ltd., the Cluster and Strategic Centre for Science, Technology and Innovation (CSTI, in Finnish SHOK) for energy and environment, and it is funded by Fortum Power and Heat Oy, a consortium member of the CCSP. The objective of the report is to give a general techno-economical evaluation of the Carbon Capture and Neutralization (CCN) method invented by Matti Nurmia. The original report also serves as my Bachelor of Science Thesis.

The supervisor of the work was Professor Risto Raiko, and I want to thank him for his help and constructing feedback during the writing process. I would also like to thank Matti Nurmia for both the invention, and answering my questions about it. Associate professor Elina Vuorimaa-Laukkanen from Department of Chemistry and Bioengineering deserves my gratitude for answering my questions about reaction chemistry in CCN.

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ABBREVIATIONS AND NOTATION

CCN	Carbon Capture and Neutralization
CCS	Carbon Capture and Storage
e	Euro
GTK	<i>Geologian tutkimuskeskus</i> , Geological Survey of Finland
IFE	Institution for Energy Technology
IPCC	Intergovernmental Panel on Climate Change
rpm	Revolutions per minute
VTT	<i>Valtion teknillinen tutkimuskeskus</i> , Technical research Center of Finland
w- %	Per cent in weight
A	Exponential constant
E_a	Activation energy (kJ/mol)
k	Reaction speed constant
k_H	Henry's constant (atm)
P	Partial pressure (atm)
R	Molar gas constant (8,314 J/molK)
x	Mole fraction

1 INTRODUCTION

As is widely known, carbon dioxide is an important greenhouse gas, and thus its emission rates are related to global warming. So far, means for reducing carbon dioxide emissions are, for example, improvements in energy efficiency and replacing coal with less pollutant energy sources, such as natural gas, and even more preferably, renewable energies. Nevertheless, these measures are not enough. That is why the capture and storage of carbon dioxide is needed to reduce the emissions, and to curb the most intensive peak of climate change.

Even though carbon capture technologies have lately been developed outstandingly, the mere capturing of carbon dioxide, however, is not a satisfactory solution to the problem, since the captured carbon dioxide also has to be stored somewhere. Hitherto, underground storage in geological formations and in oceans has been seen as the most feasible alternative. (IPCC, 2007) There is also industrial need for carbon dioxide, but the scale is not enough to expend all the captured carbon dioxide. Moreover, the majority of carbon dioxide spent in industrial processes most likely finally ends up into the atmosphere. (Koljonen, 2002)

Besides the fact that there are no geological formations suitable for carbon dioxide storage in Finland (VTT, 2011), the greatest problem of carbon storage is probably still the long-term safety of it. When storing carbon dioxide underground, there is always a possibility that it might leak back into the atmosphere. Carbon dioxide can also have other harmful effects on the ecosystem, such as ocean acidification.

A potential way to safely get rid of carbon dioxide for good is to convert it into a harmless form with the help of mineral neutralization. In neutralization, carbon dioxide chemically reacts with rock minerals, producing inoffensive bicarbonates while also forming valuable by-products. The by-products can then be sold to partially cover the neutralization costs. The Carbon Capture and Neutralization (CCN) method was invented by Matti Nurmi, and its chemical basis has been studied by Jussi Huttunen. Jens Kohlmann and Ron Zevenhoven, in co-ordination with Arun B. Mukherjee, have studied mineral carbonation that also offers ideas for carbon dioxide transformation.

The objective of this report is to describe and evaluate the CCN method on a general level, and also offer some rough estimates about the mass flows in neutralization. First, the most important neutralizing minerals are introduced in Chapter 2. Chapter 3 then concentrates on the chemistry of neutralization reactions, and Chapter 4 on giving a short description of the overall process and its placement. The neutralization costs are discussed in Chapter 5, and two examples of neutralizing rocks are finally presented in Chapter 6.

2 NEUTRALIZING MINERALS

Carbon dioxide can be neutralized with rock minerals that chemically react with carbon dioxide, converting it into inoffensive bicarbonates. The neutralization is based on chemical reactions similar to natural weathering, which causes the transformation of geological formations.

In addition to converting carbon dioxide into a harmless form, neutralization creates various by-products, many of which have financial value. The composition of minerals used for neutralization determines the by-products formed, and some minerals produce more valuable by-products than others. Aluminum compounds and silicon oxide are the most typical by-products of CCN.

2.1 Feldspars

A significant group of silicate minerals is called feldspars; they are aluminum silicate minerals that also contain sodium, potassium, calcium or barium. (Keramidas & Barbayiannis, 2005) The three feldspars suitable for carbon dioxide neutralization are anorthite, albite, and potash feldspar.

Anorthite or calcium aluminum feldspar, $\text{CaAl}_2\text{Si}_2\text{O}_8$, and albite or sodium aluminum feldspar, $\text{NaAlSi}_3\text{O}_8$, are generally found together forming plagioclase. (Hytönen, 1999) Potash feldspar is found in three different subtypes: orthoclase, anidine and microcline. The subtypes have the same chemical formula, KAlSi_3O_8 , and they are usually all called simply potash feldspars. (Hytönen, 1999)

2.2 Other special minerals

Perhaps one of the most suitable non-feldspar mineral for neutralizing carbon dioxide is spodumene $\text{LiAl}(\text{SiO}_3)_2$. (Hytönen, 1999) It is especially useful in neutralization on account of the lithium it contains, because through neutralization lithium can be extracted and exploited. Lithium is an extremely valuable element used, for example, in batteries and fusion reactors. (Nurmia, M., 2011)

Minerals containing magnesium oxides are also suitable for neutralizing carbon dioxide: they are able to cause carbonation reactions that, in addition to neutralizing carbon dioxide, produce magnesium carbonate. The most abundant and thus the most convenient magnesium silicates for neutralization are serpentine, $\text{Mg}_3\text{Si}_2\text{O}_5(\text{OH})_4$, and olivine, $(\text{Mg,Fe})_2\text{SiO}_4$. (Kohlmann & Zevenhoven, 2002) Olivine is a compound of two minerals: forsterite, Mg_2SiO_4 , and fayalite, Fe_2SiO_4 . (Hurlbut & Wyllie, 2008) Iron and calcium oxides are also able to neutralize carbon dioxide (Marini, 2006), and thus also

fayalite and olivine as such are suitable for neutralization, as well as the calcium silicate mineral wollastonite, CaSiO_3 , for example. Calcium minerals are actually even better for carbonation than magnesium silicates, since calcium silicates are more reactive than magnesium silicates. (Huttunen, 2009)

There are also many other minerals that have neutralization capacities, such as muscovite, biotite, and hornblende. However, the concentration of these minerals in rocks is usually so minimal that they cannot be considered as reasonable neutralizers in large scale operations. As minor components of the rocks containing feldspars, for instance, they may still have a role to play. In addition to aluminum, silicon, magnesium, and lithium already mentioned, minerals may also contain certain other valuable or rare elements such as rare-earth elements or heavy metals. These substances can also possibly be extracted during the neutralization process, after which they can be sold to partially cover the expenses of the neutralization.

2.3 Occurrence

In general, silicate minerals are extremely abundant. Plagioclases are the most common minerals on Earth and they are found in almost all rocks (Tuisku, 2002), 40% of Earth's continental crust actually consists of plagioclase (Lehtinen et al., 1998). Potash feldspar is also a widely appearing mineral in igneous rocks, such as granites and pegmatites (Hytönen, 1999) The continental crust contains approximately 12% potash feldspar. (Lehtinen et al., 1998) Feldspars for carbon dioxide neutralization could even possibly be acquired from other mining activities, for instance, as waste rocks or tailings.

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A significant deposit of spodumene has been found in Leviäkangas in Kaustinen, situated in Ostrobothnia, Finland. (Ahtola et al., 2010) The Central Ostrobothnia area has other spodumene deposits as well. (Keliber, 2011)

Magnesium silicates are very common on Earth, and Finland especially holds remarkable deposits of them. (Kohlmann & Zevenhoven, 2002) Moreover, wollastonite is mined in a wide scale already. (GTK, 2009b)

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2.4 Susceptibility for weathering

The susceptibility for weathering, or the relative stability of a mineral tells how easily the mineral weathers. Susceptibility differs from mineral to mineral. In general, the minerals are the more stable, the higher temperatures they need to crystallize.

When it comes to feldspars, potash feldspar is more stable than plagioclases albite and anorthite, since it has high potassium content. From plagioclases, albite is more stable than anorthite because the stability decreases with decreasing sodium content. (Huttunen, 2009) Olivine is even more eager to weather than anorthite (Schaetzl & Anderson, 2005), and the weathering of wollastonite is relatively rapid as well (Toropainen, 2006).

3 NEUTRALIZATION REACTIONS

When carbon dioxide is neutralized with aluminum silicate minerals, the mineral reacts with carbon dioxide and water and forms a cation-poor clay mineral, a cation, as well as hydrocarbonate HCO_3^- , and silicic acid H_4SiO_4 . (Murray, 2004) In reactions with metal oxides, the neutralization occurs via carbonation, forming carbonates.

The neutralization reaction is supported by the acidity of the solution. (Murray, 2004) The acidity is the result of carbon dioxide dissolution, since the dissolved carbon dioxide forms carbonic acid in the solution. (Appelo & Postma, 2005)

3.1 The dissolution of carbon dioxide

The dissolution rate of carbon dioxide in water is highly dependent on the partial pressure of carbon dioxide, and on the temperature of the reaction environment. The correlation between the solubility and partial pressure is presented according to Henry's law in Equation 1:

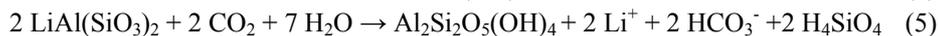
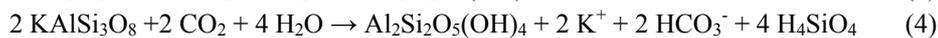
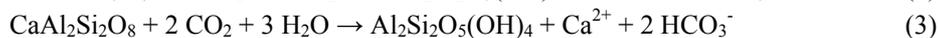
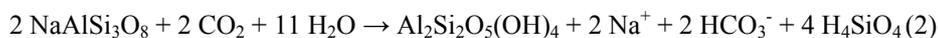
$$P = k_H \cdot x, \quad (1)$$

where P is the partial pressure, k_H is a constant called Henry's law constant, which is $1,64 \cdot 10^3$ atm for carbon dioxide, and x is the mole fraction of the dissolved gas. (Zumdahl, 2005)

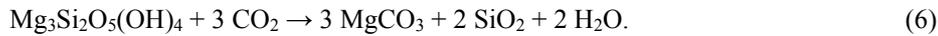
The temperature dependence of the solubility is not as straightforward. Increase in temperature causes the dissolving reaction to become more rapid, but it decreases the total rate of dissolution. (Zumdahl, 2005; CRC, 2011) The reason for this is the increase in the partial pressure of water vapor with increasing temperature; for example in 100 °C the dissolution rate of carbon dioxide in water is already approaching zero (CRC, 2011).

3.2 Neutralization reactions

The acidity of the solution causes weathering in the minerals. Depending on the circumstances, secondary products such as kaolinite, $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$, are formed. The following Equations 2–5 describe the kaolinite producing neutralization reactions for albite, anorthite, potash feldspar, and spodumene. (Huttunen, 2009)



Carbon dioxide neutralization with magnesium compounds is based on mineral carbonation. For serpentine, the carbonation reaction is (Kohlmann & Zevenhoven, 2002)



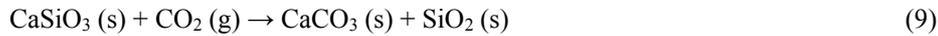
Olivine is a mixture of forsterite, Mg_2SiO_4 , and fayalite, Fe_2SiO_4 . The neutralization reaction of forsterite is (Kohlmann & Zevenhoven, 2002)



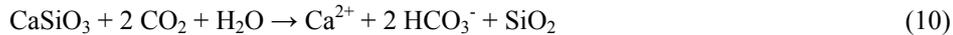
Correspondingly, the neutralization reaction of fayalite is (Marini, 2006)



Calcium carbonation is similar to the reactions with magnesium. Equation 9 presents the carbonation of wollastonite. (Huttunen, 2009)



Another possibility for wollastonite weathering is presented in Equation 10. (Huttunen, 2009)



The maximum amounts of carbon dioxide that can be neutralized with the help of various minerals can easily be determined from stoichiometric reaction equations. The maximums for the neutralization with albite, anorthite, potash feldspar, and spodumene are approximately 168 kg, 316 kg, 158 kg, and 236 kg per one ton of pure mineral. One ton of serpentine is able to neutralize 476 kg of carbon dioxide at the maximum, whereas the neutralization capacity of olivine ranges from 432 kg to 626 kg per one ton of pure mineral, depending on whether it consists of fayalite or forsterite. The carbonation capacity of one ton of wollastonite is 379 kg of carbon dioxide, whereas the neutralization capacity through feldspar-like weathering according to Equation 10 is 758 kg. The neutralization capacities of the minerals as kilograms of carbon dioxide neutralized per one ton one pure mineral are presented in Figure 3.1.

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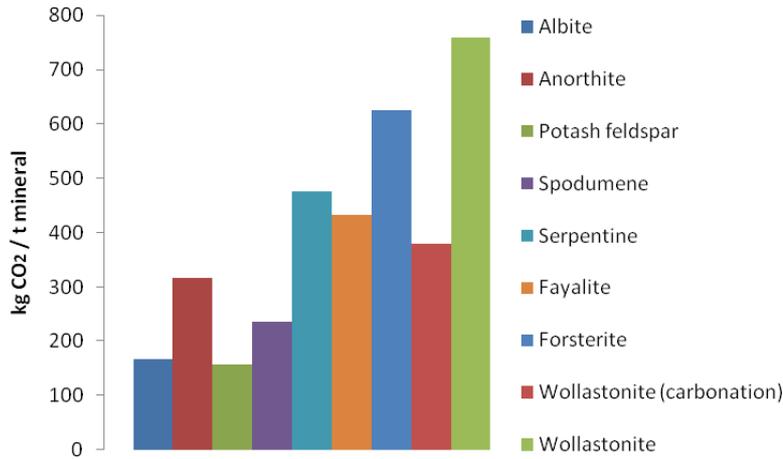


Figure 3.1. Theoretical neutralization capacities of various minerals.

Nevertheless, it is to be remembered that the neutralization capacities approximated here are based on stoichiometric reactions and are thus the theoretical maximums for the carbon dioxide neutralized. The actual neutralization capacities may significantly vary from theoretical estimations if the reactions are in reality running very imperfectly.

3.3 Neutralization by-products

Along with neutralizing carbon dioxide, the neutralization reactions also form various valuable substances as by-products. The most significant by-products are aluminum compounds and silicon oxide. Other possible by-products include lithium and magnesium carbonates, as well as certain other valuable metals, such as tantalum, niobium, and rare earths. After the neutralization has taken place, some products of less value are left behind, bicarbonate solution and used rock material being the most notable ones.

Nonetheless, the exploitation of the by-products is regulated by the current legislation that may set restrictions to their usage. The solubility of the by-products may also present a problem if the products are drifted into the environment, since some of them, for example, aluminum, are toxic. Therefore, the environmental influence of the process must be studied in depth.

3.3.1 Aluminum

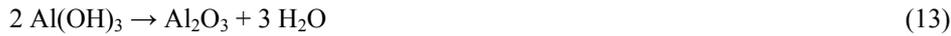
The neutralization reactions in Equations 2–5 form an aluminum compound called kaolinite. In acidic conditions kaolinite weathers further according to Equation 11:



Aluminum appears in this solute form when the pH of the surrounding solution is less than 4.5-5. However, it typically precipitates as kaolinite or aluminum hydroxide, $\text{Al}(\text{OH})_3$, also known as gibbsite. Gibbsite is formed from solute aluminum ions when the pH of the solution is greater than 5. The gibbsite-forming reaction is presented in Equation 12. (Huttunen, 2009)



The mixture of kaolinite and gibbsite is known as bauxite, which is the main source of aluminum. Bauxite can be converted into aluminum oxide, typically by leaching. The oxidization reaction is presented in Equation 13.



Aluminum oxide as such is a valuable by-product. It can also be converted into aluminum, usually via electrolytic reduction. (Totten & MacKenzie, 2003)

3.3.2 Silicon oxide

The neutralization reactions presented in Equations 2–5 can form silicic acid, H_4SiO_4 . Silicic acid is also formed when kaolinite weathers onwards, as can be seen from Equation 11. Silicic acid can be transformed into silicon oxide and water according to Equation 14. (Huttunen, 2009)



Silicon oxide or silica has seven polymorphs. One of the most common polymorph are known as quartz (Keramidas & Barbayiannis, 2005), and it is mainly used as foundry sand and construction gravel as well as in manufacturing concrete. To a smaller extent, quartz is also used in making glass, and in the ceramic industry. (Keramidas & Barbayiannis, 2005; Heaney, 2008)

3.3.3 Lithium

In addition to aluminum and silicon compounds, the neutralization reaction of spodumene presented in Equation 5 forms lithium. Lithium, eventually extracted as lithium carbonate (Nurmia M., 2011), is a valuable substance used in batteries, lubrication greases, and in ceramics, for example. (Sittig, 2008)

The estimation of lithium carbonate production according to the stoichiometric reaction equation presented in Equation 5 gives approximately 19 kg of lithium per one ton of spodumene. The corresponding lithium carbonate yield is thus 199 kg per one ton of mineral.

3.3.4 Other by-products

Carbonation with magnesium minerals forms magnesium carbonate. Magnesium compounds have several applications, and magnesium has also been classified as a critical mineral by the European Union. (GTK, 2010).

Other possible valuable by-products, depending on the composition of the neutralizing minerals, include for example tantalum, niobium, and rare-earth elements. (Nurmia M., 2011) Nevertheless, the amounts of these metals are so small that it is not cost-effective to process them individually, but rather in co-operation with other mining activities that already produce these metals. (Vuorimaa-Laukkanen, 2011)

All neutralization reactions except carbonation reactions also form bicarbonate solution that should not cause acidification, and can thus be released into natural waters. (Pat. FI 121216) Nonetheless, the water protection legislation must be taken into account when planning large-scale neutralization operations.

The used rock material, which contains minerals that have not reacted during the process, is also harmless and it can be used for many purposes. Applications include, for example, using it as material for construction or roadbeds. (Cuycha, 2011)

3.4 Generated heat

The neutralization reactions are exothermal, which means that heat is generated during the reactions. The energy released in the neutralization increases the temperature of the system. This must be taken into account when designing the process. The amount of heat released is highly mineral-specific, and therefore the values vary greatly.

The temperature change also leads to a change in volume. The volume change in neutralization is also related to the changes in chemical structure, but observing the volume change of the water involved in the process often gives an adequate approximation of its scale. (Vuorimaa-Laukkanen, 2011) Since the reactions are exothermal and increase the temperature of the system, the volume also slightly increases. This has to be considered, especially if the reactions are to take place in a reactor. The increase in volume is, however, relatively small.

3.5 Neutralization reaction speed

Weathering processes in nature are extremely slow. In order to be able to benefit from the weathering reactions in carbon dioxide neutralization, the reaction speed must somehow be accelerated. Major factors affecting the reaction speed are temperature, grain size of the mineral, and the acidity of the solution. (Huttunen, 2009) Furthermore, the mineral used to neutralize carbon dioxide affects the neutralization speed, since the minerals have different neutralization stabilities. As mentioned in Chapter 2.4, the weathering of anorthite and olivine is fairly fast, but potash feldspar in turn is relatively slow to weather. The dissolution speed of aluminum silicate minerals and thus also the neutralization speed of carbon dioxide is highly temperature dependent. The

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temperature dependence can be described by the Arrhenius equation presented in Equation 15:

$$k = A \exp(-E_a / RT), \quad (15)$$

where k is the reaction speed constant, A an exponential constant, E_a activation energy, R molar gas constant, which is 8,314 J/molK, and T is temperature. (Huttunen, 2009)

When it comes to carbonation reactions, it has been found that in high temperatures and pressures the carbonation of calcium silicates is more effective than that of magnesium silicates. For carbonation of calcium silicate wollastonite, there is an optimum temperature, approximately 200 °C. In general, raising the temperature accelerates the reaction, but at the optimal temperature of 200 °C, the effect of the activity of bicarbonate ion starts to be more dominant for the reaction speed than the effect of temperature. (Huttunen, 2009) For magnesium silicate carbonation, the optimal temperature has been found to range between 200 °C and 400 °C, depending on the partial pressure of carbon dioxide. (Kohlmann & Zevenhoven, 2002)

As mentioned above, the neutralization reactions are exothermal and thus release heat. Therefore, relatively low temperatures are thermodynamically favorable for the reactions, since extremely high temperatures would cause the reverse reaction to occur. Temperatures at which the reverse reaction starts to be more favorable are approximately 300 °C for magnesium carbonation reactions, and 900 °C for calcium carbonation. (IPCC, 2005)

The temperature rise has to be taken into account when determining the initial temperature of the process, since the actual temperature will be somewhat higher than the initial. It is also worth noting that even if the rise in temperature accelerates the reaction, it decreases the amount of carbon dioxide dissolved.

The grain size of the mineral has an effect on its specific surface area: when the grain size decreases, the surface area increases. The increase in the specific surface area increases the contact area between the mineral and the weathering factors, which include carbon dioxide and moisture. Smaller grain size also eases mass transfer, and thus means higher weathering speed. (Huttunen, 2009) Therefore, it is profitable to grind the mineral to decrease the grain size. However, grinding the mineral also increases the costs of the process, which is why there is an optimum grain size for the mineral.

It has been noted that the dissolution speed of aluminum silicate minerals is the highest when the solution is acidic. When the pH of the solution is between 5 and 8, the dissolution speed is notably lower, but increases again when the pH of the solution is more than 8. In neutralization processes the acidity of the solution is a result of carbon dioxide dissolution. In their studies, Navarre-Stichler and Thyle have found that the dissolution speeds of plagioclases are proportional to the partial pressure of carbon dioxide raised to the power of 0.45. According to Lagache, the dissolution speeds correlate with the partial pressure raised to the power of 0.3. According to Navarre-

Stichler and Thyle, however, protons do not cause the dissolution of the minerals, because the solutions in their experiments were neutral or even slightly alkaline. This means that the partial pressure of carbon dioxide only affects by raising the solubility of carbon dioxide, and thus increasing the acidity of the solution. (Huttunen, 2009)

Another observation is that in normal soil conditions regarding the pH, the temperature, and the pressure, the activity of carbonate ions affects the weathering speed of anorthite. Berg and Banwart have ran experiments and found that when the pH of the solution is more than 5, the carbon dioxide clearly accelerates the dissolution speed of aluminum, but in pH conditions under 5 the dissolution speed in the surroundings containing carbon dioxide is equivalent to that in the surroundings with nitrogen. They also noted that the dissolution of silicon is slower than that of aluminum. (Huttunen, 2009)

The rise in the partial pressure of carbon dioxide has been found to accelerate carbonation reactions as well, most likely as a result of the decrease in pH. The effect of the increase in partial pressure seems to be strongly temperature dependent: in temperatures between 100 °C and 150 °C, the increase in partial pressure notably accelerates the reaction until the partial pressure of 10 bar is achieved. The corresponding pressure limits for temperatures of 200 °C and 225 °C are 20–40 bar and 40 bar, respectively. (Huttunen, 2009)

In his experiments Huttunen has also studied the possible effects of magnetic mixing and ultrasonic treatment on the aluminum silicate mineral dissolution speed. The powers of the ultrasonic devices used in the experiments were 400 W and 650 W, frequencies being 40 kHz and 35 kHz, respectively. Both mixing and ultrasound appeared to improve the dissolution, though the improvement caused by mixing is most likely a result of the decreased grain size of the mineral. (Huttunen, 2009)

The effect of the mixing has also been studied in wollastonite carbonation. Mixing seems to improve the reaction speed more in low than in high speeds, and the mixing speed of more than 500 rpm does not accelerate the reaction any further. (Huttunen, 2009)

Activating the mineral by heat treatment also improves its solubility. Using heat treatment, however, would remarkably raise the costs of neutralization process, and thus the heat treatment may not be a very reasonable alternative for increasing the reaction speed. (IPCC, 2005)

4 NEUTRALIZATION PROCESS

The basic idea of the neutralization method is to dissolve carbon dioxide from the flue or other gas into water, and then neutralize the solution with rock minerals. Plain water can be used for dissolution; there is no need to add any chemicals into it. After the dissolution of carbon dioxide, the solution can be neutralized by passing it through rock minerals, and thus enforcing the neutralizing chemical reactions to occur.

4.1 Demands for the flue gas

Carbon dioxide solubility in water depends on the partial pressure of the carbon dioxide and on the prevailing temperature. Therefore, increasing the partial pressure of carbon dioxide decreases the consumption of process water as well as accelerates the neutralization process by increasing the acidity of the solution to be neutralized.

In order to maintain the amount of the required process water at a reasonable level, the partial pressure of carbon dioxide has to be high enough, 0.4 bar at the minimum. This level can be achieved by pressurizing the flue gas, or by having a sufficiently high concentration of carbon dioxide in the flue gas. The high carbon dioxide concentration can be attained, for example, by using the oxygen-fuel combustion method. While pressurizing the gas, the power input required to operate the compressor can be at least partially produced by exploiting the heat of the hot flue gases coming into the process, by letting them expand in a turbine that is coupled to the pressurizing compressor. (Pat. FI 121216)

4.2 Embodiments

In his patent application, Nurmia has introduced his invention with the help of three Embodiments, which are presented in Figures 1, 2, and 3 in Appendix 1. Embodiment 1 depicts an application where a part of the used water stream is recycled back into the process. In Embodiment 2, only a part of the gas stream is neutralized, and the dissolution and neutralization processes are combined. In Embodiment 3, the gas stream being neutralized is almost pure carbon dioxide. (Pat. FI 121216)

Nurmia emphasizes that the three Embodiments are only a few examples of different ways to apply his invention. Numerous new applications of the method can be developed, improving its feasibility in various uses.

4.3 Placement of the process

The neutralization process can be placed either at the site where carbon dioxide is formed, for example at a power plant, or at the quarry where the required minerals are mined. If the process is set to take place at the carbon dioxide forming site, minerals have to be transported from the quarry to the neutralization site. The masses of the minerals to be transported are several times greater than those of the carbon dioxide they are able to neutralize. Furthermore, the neutralizing minerals appear in rocks with minerals without neutralization capacity, which makes the transported masses even more abundant.

The need for transporting the minerals can be avoided by placing the process at the mine. Carbon dioxide can be transported to the mine, for example, by shipping it or via pipelines. This alternative will most likely be substantially more economical than transporting the minerals.

The neutralization process could possibly be carried out at an opencast quarry where the crushed rock material containing neutralizing minerals is covered with water to maintain the required pressure, and to prevent the evaporation of carbon dioxide. By realizing the process underground, many environmental disadvantages of opencast quarry, such as dust formation, could also be avoided. (Pat. FI 121216; Wennerström, 1994)

A further possibility for placing the process is to carry it out in a tanker, where the cargo holds have been filled with neutralizing minerals. This application facilitates the exploitation of by-products, since the tanker could transport them for utilization or further processing. Moreover, the bicarbonate solution formed in neutralization could be released into a wide area during the journey of the tanker, weakening the possible harmful effects it might have in nature. (Pat. GB 1109300.2)

4.4 Pilot projects

Cuycha Innovation Oy is already starting to build the first pilot plant applying the CCN method in the Republic of South Africa in co-operation with a local corporation, CircleLink Holdings. (CircleLink Holdings, 2011; Nurmia M., 2011) A similar plant is also planned to be built in Botswana. Cuycha Innovation Oy has been asked to consult in the design of similar plants in China and Dubai as well. (Nurmia I., 2011)

The Nordic Mining ASA has also been studying the neutralization method. Two extensive feasibility studies concerning carbon dioxide neutralization with, for example, anorthite have already been concluded. One of the studies has been made in Finland, another in Norway by Gassnova and IFE. The method has been found to work well, and it has even already produced aluminum. Keliber Oy, which is an affiliated company of the Nordic Mining ASA possessing the lithium deposits of Central Ostrobothnia, has also applied the neutralization method to lithium carbonate production. (Nurmia I., 2011)

5 NEUTRALIZATION COSTS

One of the major costs in the entire CCN method is the mining of the neutralizing minerals. Using by-products of other mining activities, such as waste rocks, may outstandingly decrease the mining costs. In addition to mining, the minerals must either be taken to the power plant producing carbon dioxide, or carbon dioxide has to be taken to the mine, as mentioned in the previous chapter. Since the masses of the neutralizing rock material are multifold in comparison to the amounts of carbon dioxide they are able to neutralize, transporting carbon dioxide to the mine, either by shipping or via pipelines, will most likely be a more economical solution. Besides bringing savings in transportation costs, the neutralization process carried out at the mine also enables implementing the process underground, in which case the process water is naturally pressurized to the required level, and the pressurization costs can thus be avoided.

The demands made for the flue gas incur costs as well, since in order to attain sufficient partial pressure of carbon dioxide, the carbon dioxide concentration in the gas has to be high enough. This can be reached, for example, by oxy-fuel combustion. Another option for maintaining the partial pressure of carbon dioxide sufficiently high is to pressurize the gas with a compressor. The power input required by the compressor incurs costs, but they can be at least partially covered by reclaiming the energy of hot flue gases in a turbine coupled to the compressor.

In spite of all the aforementioned costs, the CCN method has a huge economical advantage compared to other CCS methods because it produces valuable by-products that can be sold to cover the expenditures at least partially. The most important by-products are aluminum compounds and silica. The use of spodumene as a neutralizer produces lithium carbonate that can also be sold for a good price. Magnesium carbonate is the most important by-product in carbonation reactions. According to Ilkka Nurmi, the alumina market price in May 2011 was 420 e per ton, and the price of lithium carbonate 3 550 e per ton. The price of quartz sand was 105 e per ton (Pat. GB 1109300.2). However, the current waste legislation may affect the exploitation possibilities of the by-products.

When estimating the carbon dioxide neutralization costs, it is of course important to remember also the savings achieved through reduced carbon dioxide emissions. Therefore, the profitability of neutralizing carbon dioxide is strongly dependent on the development of emission limits, which in turn are dependent on the trade of emissions.

6 EXAMPLES

This chapter provides rough mass flow and cost estimations when neutralizing one ton of carbon dioxide with two different rocks. First, neutralization with typical Finnish granite is studied. As a special case, the neutralization with Leviäkangas spodumene is presented as well, since neutralization with spodumene produces lithium, and may thus be highly profitable. The estimates are based on theoretical stoichiometric reactions presented in Chapter 3.2.

For cost estimates, the price of the rock material is thought to be approximately 1 e per ton. The prices of alumina, silica, and lithium carbonate are estimated to be 420, 90, and 3000 e per ton, as mentioned in Chapter 5.

6.1 Typical Finnish granite

Finnish granites have been widely examined, for example, by the Geological Survey of Finland. Based on the mineral compositions given in various reports (Kesola, 1998; Matisto 1969; Haapala, 1977), Table 6.1 lists a possible composition of typical granite in Finland. The plagioclase is assumed to contain 50% both albite and anorthite; minor minerals under the name “others” are assumed to have no neutralization capacity, even though in reality they likely contain other neutralizing minerals as well, including muscovite and hornblende.

Table 6.1. A typical composition of Finnish granite.

	w- %
Quartz	30
Plagioclase	30
Potash feldspar	25
Others	15

The mass flows in theoretical neutralization of one ton of carbon dioxide are represented in Figure 6.1. The neutralization requires 8,33 tons of rock material while simultaneously producing 1160 kg of alumina and 3020 kg of silica.

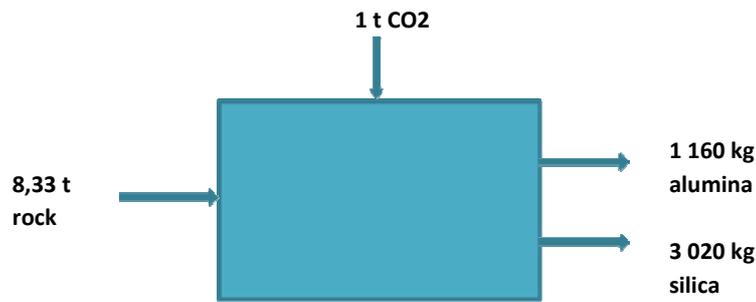


Figure 6.1. Theoretical mass flows in carbon dioxide neutralization with typical Finnish granite.

According to the estimated amounts of rock, the price of the neutralizing rock material for one ton of carbon dioxide would be approximately 8 e. Since the value of the by-products is in this case approximately 760 e, selling the by-products would clearly overrun the expenses of the neutralizing rock material.

6.2 Leviäkangas spodumene

The composition of Leviäkangas spodumene is presented in Table 6.2. (Ahtola et al., 2010) Theoretical mass flows needed for neutralizing one ton of carbon dioxide emissions are presented in Figure 6.2. Approximately 7.35 tons of spodumene is needed, and the neutralization produces 1160 kg of aluminum oxide, 3460 kg of silica, and 150 kg of lithium carbonate.

Table 6.2. Composition of Leviäkangas spodumene. (According to Ahtola et al., 2010)

	w-%
Albite	41
Quartz	28
Potash felspar	15
Spodumene	10
Muscovite	6

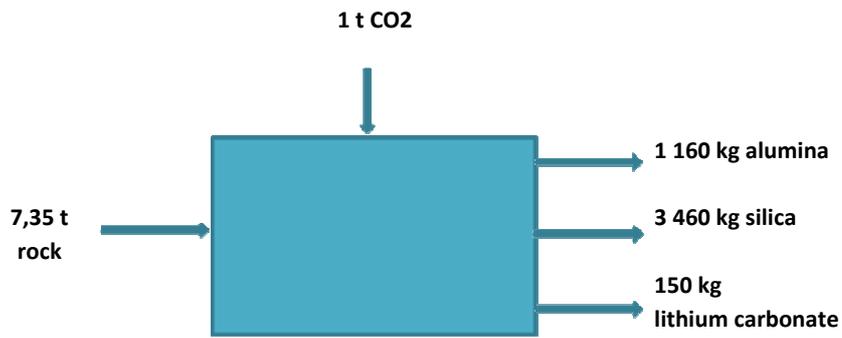


Figure 6.2. Theoretical mass flows in carbon dioxide neutralization with *Leviäkangas spodumene*.

In this case the cost of the neutralizing rock material would be approximately 7 e. Selling the by-products would bring the proceeds of approximately 1 280 e. Thus, the by-product sale would again completely cover the costs of the required neutralizing rock, giving remarkable profits as well.

7 SUMMARY AND CONCLUSION

The Carbon Capture and Neutralization (CCN) method invented by Matti Nurmi provides a new, innovative solution for Carbon Capture and Storage (CCS). Unlike in previous CCS methods, the captured carbon dioxide is not stored but transformed into a harmless form, thus totally avoiding the need for storage. The transformation, based on natural rock weathering processes, is done with minerals found in common rocks, feldspars being particularly useful. The neutralization produces bicarbonate solution and valuable by-products, such as aluminum compounds and silica.

In the CCN method, the captured carbon dioxide is first dissolved into water and the solution is then neutralized by passing it through mineral-containing material. In order to minimize the water consumption in the process, the partial pressure of carbon dioxide in the gas to be refined should be relatively high, at least 0.4 bar. This level can be attained, for example, by pressurizing the gas, or via oxygen combustion. There are numerous possible applications on the method, providing several alternatives for cost minimization as well.

The neutralization process can be placed either at the carbon dioxide-producing site, such as a power plant, or at the site where the neutralizing minerals are mined. The transportation of carbon dioxide and placing the process at the mine will most likely be more cost-efficient, since the mass flows of carbon dioxide are remarkably smaller than those of the neutralizing minerals. The alternatives for carbon dioxide transportation are shipping and pipelines. The choice between these alternatives is strongly case-specific. In the beginning, the neutralization process will most likely be realized in a reactor; at a later phase the possibility of passing the carbon dioxide water straight to mineral-containing rock foundation could also be considered. In this case, mining the minerals would become needless, but the by-products could not be exploited. Due to the low expenditures and large scale, this straightforward approach might still turn out to be the most affordable solution to realize the CCN method. Another suggested alternative is to place the entire process in a tanker.

The neutralization reaction speed is affected by several factors including temperature, grain size, and the acidity of the solution. Neutralization speed has already to some extent been studied, but further laboratory studies would be necessary for more information.

Theoretical neutralization capacities of different types of minerals were presented in this report, and feldspar-like weathering of wollastonite was found to seem the most effective way to neutralize carbon dioxide. Neutralization through carbonation reactions seems to be relatively effective as well, but the only valuable by-product produced

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through carbonation is silicon oxide. This would decrease the profits from the by-product sale. Among feldspars, anorthite seems to be the best neutralizer. The estimations were, nonetheless, based on theoretical stoichiometric reactions, which give only the minimums of the neutralizing rock materials needed, since the reactions may in reality run relatively imperfectly. Therefore, the actual mass flows in neutralization have to be determined, for example, by laboratory studies before the feasibility of the method can be defined accurately.

Chapter 6 presented carbon dioxide neutralization with two particular rocks, typical Finnish granite and Leviäkangas spodumene. The amount of rock theoretically needed to neutralize one carbon dioxide ton could range from 5 to 10 tons. Therefore, the price of the neutralizing rock material should not be too high, especially when comparing it to emission allowance prices, or to the proceeds from selling the by-products.

The survey and mapping of the rock resources suitable for neutralization, preferably in the vicinity of the power plants and other plants producing carbon dioxide to minimize the need of transportation, is essential for realizing the CCN. The best expertise for this type of studies can probably be found in the Geological Survey of Finland.

Furthermore, the possible environmental risks related to neutralization have to be examined in depth as well. For instance, the reversed reactions may release the carbon dioxide from bicarbonates back into the atmosphere. In addition, the solubility of the by-products and possible secondary reactions may present problems. As was the case with design and sizing information, the assessment of the environmental impacts is impossible without experimental research. Therefore, the use of a sufficiently large test reactor is necessary.

The competitiveness of the CCN method in comparison to other CCS methods is fundamental when rating its feasibility. The major costs of the neutralization consist of the mining of neutralizing minerals, transporting either carbon dioxide or minerals and realizing the process, including for example investment and pressurizing costs. The advantage of the CCN method compared to other CCS-methods is, however, the valuable by-products formed in the neutralization, since they can be sold to cover the neutralization costs at least partially. Careful cost estimation is essential for evaluating the feasibility of the method and should thus be made in detail.

All things considered, the CCN method seems to be an interesting and feasible way to carry out the CCS process. Before the method can be applied at an industrial scale, further research is needed to determine the design and dimensioning of the process, as well as environmental impacts of it. Both laboratory and pilot scale test reactors would be recommended. A survey of the rock resources suitable for neutralization is also necessary. The first pilot plants are already about to be built abroad, and if these projects are finished, they will provide plenty of valuable information about the CCN, but also probably increase the prices of the licenses of the patent. Therefore, swift actions for the research and appliance of the method are essential for gaining both the environmental and economical benefits of the invention.

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APPENDIX 1: FLOW CHARTS

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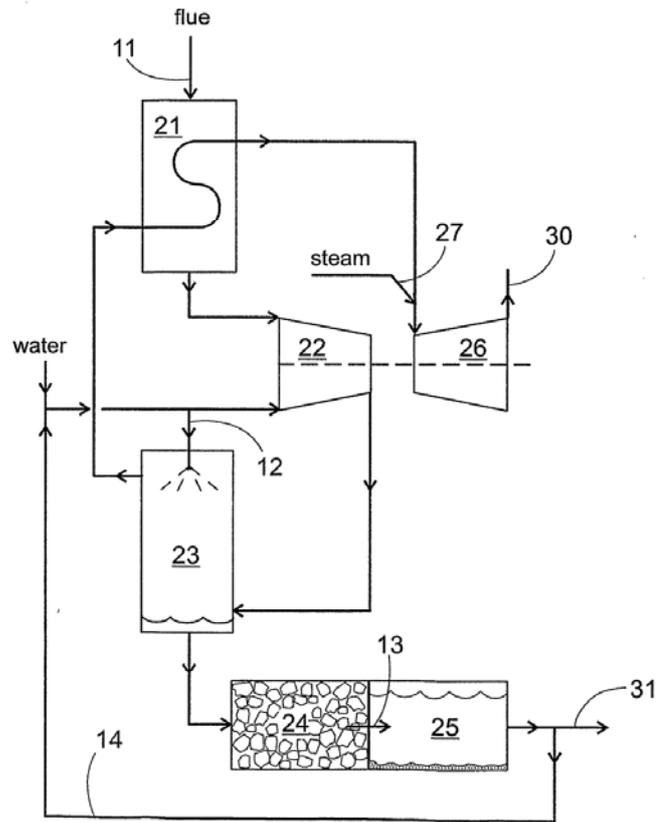


Fig. 1

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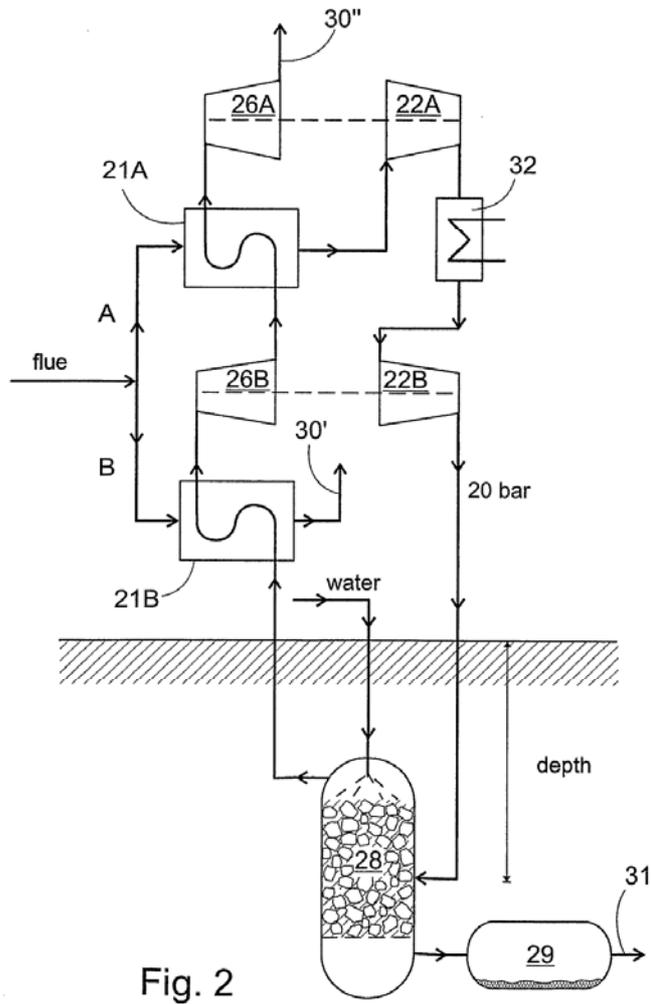


Fig. 2

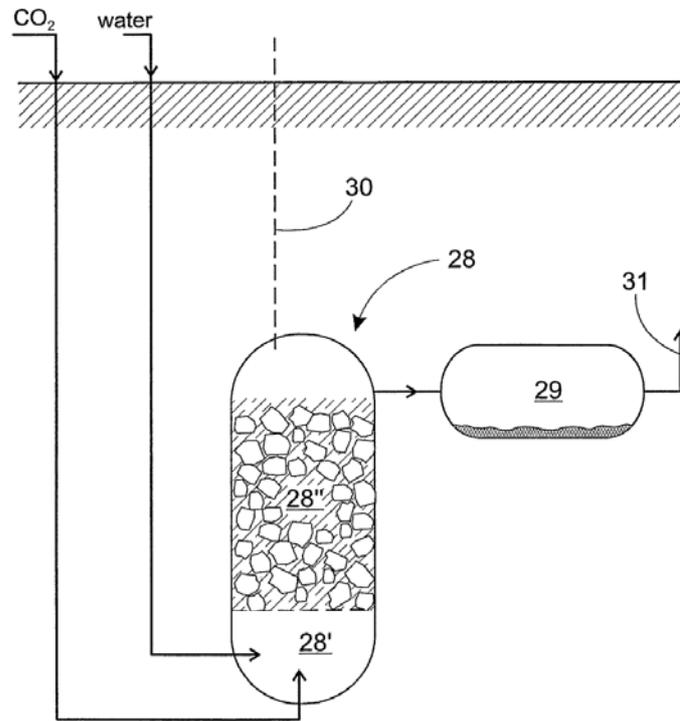


Fig. 3

APPENDIX 2: DICTIONARY

Aluminum hydroxide	Alumiinihydroksidi	$\text{Al}(\text{OH})_3$
Aluminum oxide, alumina	Alumiinioksidi, alumina	Al_2O_3
Andesine	Andesiini	$\text{An}_{30-50}\text{Al}_{50-70}$
Anidine	Anidiini	KAlSi_3O_8
Anorthite	Anortiitti	$\text{CaAl}_2\text{Si}_2\text{O}_8$
Albite	Albiitti	$\text{NaAl}_2\text{Si}_2\text{O}_8$
Augite	Augiitti	$(\text{Ca},\text{Mg},\text{Fe})_2\text{Si}_2\text{O}_6 /$ $(\text{Ca},\text{Na})(\text{Mg},\text{Fe},\text{Al},\text{Ti})(\text{Si},\text{Al})_2\text{O}_6$
Biotite	Biotiitti	$\text{K}(\text{Mg},\text{Fe})_3(\text{Al},\text{Fe})\text{Si}_3\text{O}_{10}(\text{OH},\text{F})_2$
Bytownite	Bytowniitti	$\text{An}_{70-90}\text{Al}_{10-30}$
Christobalite	Kristobaliitti	SiO_2
Dolomite	Dolomiitti	$\text{CaMg}(\text{CO}_3)_2$
Fayalite	Fayaliitti	Fe_2SiO_4
Feldspar	Maasälpä	
Forsterite	Forsteriitti	Mg_2SiO_4
Gibbsite	Gibbsiitti	$\text{Al}(\text{OH})_3$
Granite	Graniitti	
Hornblende	Sarvivälke	$\text{Ca}_2(\text{Mg},\text{Fe})_4\text{Al}(\text{Si}_7\text{Al})\text{O}_{22}(\text{OH},\text{F})_2$
Kaolinite	Kaoliniitti	$\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$
Labradorite	Labradoriitti	$\text{An}_{50-70}\text{Al}_{30-50}$
Limestone	Kalkkikivi	CaCO_3
Mica	Kiille	
Microcline	Mikrokliini	KAlSi_3O_8
Muscovite	Muskoviitti	$\text{KA}_2(\text{AlSi}_3)\text{O}_{10}(\text{OH},\text{F})_2$
Niobium	Niobium	Nb
Oligoclase	Oligoklaasi	$\text{An}_{10-30}\text{Al}_{70-90}$
Olivine	Oliiviini	$(\text{Mg},\text{Fe})_2\text{SiO}_4$
Orthoclase	Ortoklaasi	KAlSi_3O_8
Phlogopite	Flogopiitti	$\text{KMg}_3\text{AlSi}_3\text{O}_{10}(\text{OH})_2$
Plagioclase	Plagioklaasi	
Potash feldspar/ Potassium feldspar	Kalimaasälpä	KAlSi_3O_8
Quartz	Kvartsi	SiO_2
Serpentine	Serpentiini	$\text{Mg}_3\text{Si}_2\text{O}_5(\text{OH})_4$
Silicic acid	Piihappo	H_4SiO_4
Silicon	Pii	Si
Silicon oxide, silica	Pioksidi, silika	SiO_2
Spodumene	Spodumeeni	$\text{LiAl}(\text{SiO}_3)_2$
Tantalum	Tantaali	Ta

Wollastonite

Wollastoniitti

CaSiO_3