Comparative Life-Cycle Assessment of Slurry vs Wet carbonation of BOF slag

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Abstract
Accelerated carbonation is a new CO₂ storage method under development as a solution for climate change caused by anthropogenic activities. In accelerated carbonation an alkaline source such as minerals or industrial residues react with carbon dioxide in a presence of slightly acidic solution to produce stable solid carbonates. There are varieties of accelerated carbonation routes which differ in process condition.

The aim of this study was to evaluate and compare the potential of a slurry route process and a wet route process for the carbonation of basic oxygen furnace (BOF) slag using the CO₂ emitted by a conventional natural gas power plant.

For this purpose a life cycle assessment (LCA) study was performed based on principles and guidelines provided by ISO 14040:2006 and routines and data provided by the SimaPro v8 software package. The material and energy requirements for each of the steps involved in the carbonation process, i.e. pre-treatment of raw material, CO₂ compression, transportation, carbonation step, after-treatment and landfill, were calculated and included in the LCA study.

The slurry and wet route resulted in net CO₂ reduction of 87.4% and 72.3% respectively. However, a positive contribution to other environmental issues was observed with the wet route leading to higher impact mainly due to high heating requirement. An exception was contribution of slurry route to abiotic resource depletion, which was higher for the slurry route due to high water requirement. A general conclusion was that the electricity consumption is the main cause of environmental issues. Sensitivity analyses showed that the environmental impacts are dependent on the transportation distance and electricity source, while no dependence was observed with respect to construction of the carbonation plant.
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### Symbols

<table>
<thead>
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<th>Symbol</th>
<th>Description</th>
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<tbody>
<tr>
<td>( \eta_{ls} )</td>
<td>Isentropic efficiency of compressor ([-])</td>
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<tr>
<td>( \eta_p )</td>
<td>Pump efficiency ([-])</td>
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<tr>
<td>( \mu )</td>
<td>Dynamic viscosity</td>
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<tr>
<td>( \rho )</td>
<td>Density ([\text{kg/m}^3])</td>
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<td>( \rho_c )</td>
<td>Density of cold (non carbonated) slurry ([\text{kg/m}^3])</td>
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<td>( \rho_{csl} )</td>
<td>Density of carbonated slurry ([\text{kg/m}^3])</td>
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<tr>
<td>( \rho_h )</td>
<td>Density of hot (carbonated) slurry ([\text{kg/m}^3])</td>
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<tr>
<td>( \rho_{H_2O} )</td>
<td>Density of water ([\text{kg/m}^3])</td>
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<td>( \rho_l )</td>
<td>Density of limestone ([\text{kg/m}^3])</td>
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<td>( \rho_s )</td>
<td>Density of slag ([\text{kg/m}^3])</td>
</tr>
<tr>
<td>( \rho_{sl} )</td>
<td>Density of slurry ([\text{kg/m}^3])</td>
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<tr>
<td>( \rho_{st} )</td>
<td>Density of steel ([\text{kg/m}^3])</td>
</tr>
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<td>( \omega )</td>
<td>Angular velocity</td>
</tr>
<tr>
<td>( A )</td>
<td>Heat transfer area of heat exchanger ([\text{m}^2])</td>
</tr>
<tr>
<td>( A_b )</td>
<td>Heat transfer area of the biggest heat exchanger ([\text{m}^2])</td>
</tr>
<tr>
<td>( A_s )</td>
<td>Heat transfer area of the smallest heat exchanger ([\text{m}^2])</td>
</tr>
<tr>
<td>( C )</td>
<td>Annual production of power plant at full capacity ([\text{MWh}_{el}/\text{year}])</td>
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<tr>
<td>( c_{Ca^{2+}} )</td>
<td>Molar concentration of (\text{Ca}^{2+}) ([\text{kmol/kmol}])</td>
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<tr>
<td>( c_{Mg^{2+}} )</td>
<td>Molar concentration of (\text{Mg}^{2+}) ([\text{kmol/kmol}])</td>
</tr>
<tr>
<td>( C_p )</td>
<td>Specific heat capacity at constant pressure ([\text{J/mol \cdot K}])</td>
</tr>
<tr>
<td>( C_R )</td>
<td>Compression ratio of each stage ([-])</td>
</tr>
<tr>
<td>( C_v )</td>
<td>Specific heat capacity at constant volume ([\text{J/mol \cdot K}])</td>
</tr>
<tr>
<td>( D )</td>
<td>Depth of heat exchanger ([\text{m}])</td>
</tr>
<tr>
<td>( D_b )</td>
<td>Depth of biggest heat exchanger ([\text{m}])</td>
</tr>
<tr>
<td>( d_0 )</td>
<td>Pre-grinding particle size ([\text{m}])</td>
</tr>
</tbody>
</table>
$d_1$ Theoretical sieve size through which 80 wt% of residues pass [m]

$d_c$ Diameter of the channels of heat exchanger [m]

$d_{\text{plate}}$ Thickness of spiral plate [m]

$D_s$ Depth of smallest heat exchanger [m]

$E_{\text{agg}}$ Energy for agitation in slurry route reactor [kJ/MWh$_{el}$]

$E_{\text{comp}}$ Compression energy requirement [kJ/MWh$_{el}$]

$E_g$ Energy requirement for grinding [kWh/t]

$E_i$ Standard Bond’s work index of residues [kJ/MWh$_{el}$]

$E_{\text{heat}}$ Heating requirement [MJ/tCO$_2$]

$E_{\text{hx}}$ Energy requirement for pumping slurries into heat exchanger

$E_{\text{mix}}$ Energy for mixing [MJ/tCO$_2$]

$E_{\text{stage},i}$ Compression energy at stage $i$, [MJ/tCO$_2$]

$E_{\text{cf}}$ Energy requirement of centrifugal filter [kJ/MWh$_{el}$]

$E_{\text{clarifier}}$ Energy requirement of clarification [kJ/MWh$_{el}$]

$E_p$ Pumping energy [kJ/MWh$_{el}$]

$G$ Average velocity gradient [s$^{-1}$]

$h$ Height of reactor/mixer or material on conveyor belt [m]

$H$ Height of heat exchanger [m]

$h_l$ Height of limestone on conveyor belt [m]

$h_s$ Height of slag on conveyor belt [m]

$k_s$ Ratio of specific heat capacities of CO$_2$ for each compression stage $i$ [-]

$l$ Length [m]

$L$ Length of spiral plate in heat exchanger [m]

$L/S$ Liquid to solid ratio [l/kg]

$l_t$ Length of conveyor belt occupied by 1 kg limestone [m]

$l_s$ Length of conveyor belt occupied by 1 kg slag [m]
$m$ Mass [kg]

$m_c$ Mass of cold (non carbonated) slurry in heat exchanger [kg]

$m_{CO_2}$ Molar mass of CO$_2$ [kg]

$M_{CO_2}$ Molar mass of CO$_2$ [kg/kmol]

$m_{CO_2}(FG)$ Mass of CO$_2$ in flue gas [kg]

$m_{cs}$ Mass of carbonated slag [kg]

$m_{cst}$ Mass of carbonated slurry [kg]

$m_{gw}$ Mass of glass wool [kg]

$m_h$ Mass of hot (carbonated) slurry in heat exchanger [kg]

$m_{H_2O}$ Mass of water [kg]

$M_{rot}$ Momentum of rotation

$m_s$ Mass of slag [kg]

$m_{st}$ Mass of unreacted slurry [kg]

$m_{st}$ Mass of steel [kg]

$N$ Number of compression stages [—]

$n_{CO_2}$ Number of moles of CO$_2$ to be captured [kmol]

$\Delta P$ Pressure drop in heat exchanger [kPa]

$\Delta P_c$ Pressure drop on cold side of heat exchanger [kPa]

$p_{cf}$ Portion of centrifugal filter [p/MWh$_{el}$]

$p_{cl}$ Portion of clarifier used [p/MWh$_{el}$]

$p_{CO_2}$ Pressure of CO$_2$ achieved after compression [MPa]

$P_{cut}$ Cut off pressure of CO$_2$ [MPa]

$P_{FG}$ Pressure of flue gas [Pa]

$\Delta P_h$ Pressure drop on hot side of heat exchanger [kPa]

$p_{hx}$ Portion of heat exchanger [p/MWh$_{el}$]

$P_{hx}$ Power requirement for pumping slurries into heat exchanger [kW]
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$P_{\text{in}}$</td>
<td>Pressure of CO$_2$ at inlet of compressor [MPa]</td>
</tr>
<tr>
<td>$P_{\text{initial}}$</td>
<td>Initial pressure of CO$_2$ [MPa]</td>
</tr>
<tr>
<td>$p_{\text{ltfetime}}$</td>
<td>Lifetime factor [p/MWh$_{\text{el}}$]</td>
</tr>
<tr>
<td>$p_{\text{mix}}$</td>
<td>Portion of mixer used [p/MWh$_{\text{el}}$]</td>
</tr>
<tr>
<td>$P_{\text{out}}$</td>
<td>Pressure of CO$_2$ at outlet of compressor [MPa]</td>
</tr>
<tr>
<td>$P_{\text{rot}}$</td>
<td>Rotation power [kW]</td>
</tr>
<tr>
<td>$p_{\text{rlrt}}$</td>
<td>Portion of slurry route carbonation reactor used [p/MWh$_{\text{el}}$]</td>
</tr>
<tr>
<td>$p_{\text{rw}}$</td>
<td>Portion of wet route carbonation reactor used [p/MWh$_{\text{el}}$]</td>
</tr>
<tr>
<td>$P_s$</td>
<td>Standard pressure [Pa]</td>
</tr>
<tr>
<td>$q$</td>
<td>Heat transferred [kJ]</td>
</tr>
<tr>
<td>$Q$</td>
<td>Heat transferred [kW]</td>
</tr>
<tr>
<td>$q_c$</td>
<td>Heat content of cold stream into heat exchanger [kJ]</td>
</tr>
<tr>
<td>$q_h$</td>
<td>Heat content of cold stream into heat exchanger [kJ]</td>
</tr>
<tr>
<td>$q_{\text{tot}}$</td>
<td>Total heat content of cold- and hot streams flowing into heat exchanger [kJ]</td>
</tr>
<tr>
<td>$r$</td>
<td>Radius [m]</td>
</tr>
<tr>
<td>$R$</td>
<td>Gas constant [J/mol · K]</td>
</tr>
<tr>
<td>$R_{\text{CO}_2}$</td>
<td>Carbonation potential [kg residues/kgCO$_2$]</td>
</tr>
<tr>
<td>$R_{\text{eff}}$</td>
<td>Slag requirement per unit mass of CO$_2$</td>
</tr>
<tr>
<td>$r_i$</td>
<td>Internal radius [m]</td>
</tr>
<tr>
<td>$R_x$</td>
<td>Reactivity of the slag [-]</td>
</tr>
<tr>
<td>$T$</td>
<td>Temperature [K]</td>
</tr>
<tr>
<td>$T_{\text{Cu}}$</td>
<td>Thickness of heating coil [m]</td>
</tr>
<tr>
<td>$t_{\text{gw}}$</td>
<td>Thickness of glass wool layer [m]</td>
</tr>
<tr>
<td>$\Delta T$</td>
<td>Temperature difference [K]</td>
</tr>
<tr>
<td>$T_{\text{FG}}$</td>
<td>Temperature of flue gas [K]</td>
</tr>
<tr>
<td>$T_{\text{in}}$</td>
<td>Inlet temperature to compressor [K]</td>
</tr>
</tbody>
</table>
\(T_m\) Logarithmical mean temperature [K]
\(t_{\text{MWh}}\) Time of producing 1 MWh electricity [s/MWh\(_{\text{el}}\)]
\(t_R\) Reaction time [h]
\(T_s\) Standard temperature [K]
\(t_{\text{st1}}\) Thickness of steel layer, first layer [m]
\(t_{\text{st2}}\) Thickness of steel layer, second layer [m]
u Average velocity of slurries [m/s]
\(V\) Volume [m\(^3\)]
\(V_{\text{clf}}\) Full capacity of clarifier [m\(^3\)]
\(v_{\text{CO}_2}\) Specific volume of CO\(_2\) [l/mol]
\(V_{\text{CO}_2}\) Volume of CO\(_2\) to be captured [m\(^3\)]
\(V_{\text{mfc}}\) Full capacity of mixer [m\(^3\)]
\(V_{\text{plate}}\) Volume of steel needed for spiral plate in heat exchanger [m\(^3\)]
\(V_{\text{shell}}\) Volume of steel needed for Shell of heat exchanger [m\(^3\)]
\(V_{\text{sl}}\) Volume of slurry [m\(^3\)]
\(V_{\text{st}}\) Volume of steel [m\(^3\)]
\(v_{\text{STP}}\) Normal volume at standard pressure and temperature [l/mol]
\(V_{\text{wrf}}\) Full capacity of wet route carbonation reactor [m\(^3\)]
w Width of material on conveyor belt [m]
\(W\) Width of heat exchanger [m]
w\(_l\) Width of limestone on conveyor belt [m]
w\(_s\) Width of slag on conveyor belt [m]
x\(_{\text{cs}}\) Mass fraction of carbonated slag [−]
x\(_{\text{H}_2}\text{O}\) Mass fraction of water [−]
x\(_s\) Mass fraction of slag [−]
\(Z_s\) Average compressibility factor for CO\(_2\) at stage \(i\) [−]
Abbreviations

AC – Acidification
AD – Abiotic resource depletion
ADF – Abiotic depletion (fossil fuel)
APC – Air pollution control
ÅAU – Åbo Akademi University
C – Compressor
CKD – Cement kiln dust
Csl – Carbonated slurry
CS – Carbonated slag
ET – Ecotoxicity
EU – Eutrophication
FA – Fly ash
FG – Flue gas
FWAE – Fresh water aqueous ecotoxicity
G – Grinding
GHG – Green house gas
GWP – Global warming potential
HT – Human toxicity
Hx – Heat exchanger
ISO – International Organization for Standardization
L – Landfill
M – Mixer
MAE – Marine aqueous ecotoxicity
NETL – National Energy Technology Laboratory
NGCC – Natural gas combined cycle
ODP – Ozone layer depletion potential
PC – Pulverized coal
PO – Photochemical oxidation
R – Carbonation
SS – Steelmaking slag
T – Transportation
TE – Terrestrial ecotoxicity
W – Wastewater treatment
1 Introduction
Since the recognition of climate change due to greenhouse gas (GHG) emissions, many approaches have been made aiming at finding solutions for this environmental issue. The main cause of global warming is the emission of CO$_2$ from anthropogenic activities, mainly energy consumption within industrial sectors. With the growing population the demand will increase which reflects in an increase of CO$_2$ emissions. A switch to renewable energy requires a transition in the already existing infrastructure. At the same time, the transition and development are not occurring at a pace that is fast enough to overwin the existing processes. Until a complete switch is achieved the emitted CO$_2$ from the existing industries must be taken care of. This results in the CO$_2$ capture and storage being the most feasible solution to the current situation.

Geological storage of CO$_2$ in soil or depleted oil reservoirs has been studied but is still at the research and development state. There are some drawbacks to this method such as the need for long term control of the storage site and lack of knowledge about the destiny of the stored CO$_2$ (Giannoulakis et al., 2014; Xiao et al., 2014). The process is energy intensive (Nduagu et al., 2012) which in turn leads to additional environmental impacts. In addition, there is a chance of gas leakage and the method is not yet well-developed, making it difficult to be accepted by public or investors (Giannoulakis et al., 2014). A new CO$_2$ storage method has gained a lot of attention, the so called accelerated carbonation. In accelerated carbonation alkaline magnesium- and calcium ions react with carbon dioxide in presence of an acid to form a precipitate containing solid carbonates.

1.1 Principles of accelerated carbonation
Accelerated carbonation mimics the natural weathering process, which is a neutralization process between alkaline silicates and weak carbonic acid. During natural weathering, atmospheric CO$_2$ hydrolyses to carbonic acid in rainwater. The acidic rainwater lowers the pH of the minerals forcing calcium and magnesium ions to leach out of the silicate matrix and migrate into the water. The alkaline ions are then transferred into the rivers where they precipitate as solid carbonates (Pan et al., 2012).

There are numerous potential alkaline sources that can be used for sequestering CO$_2$, which fall into two categories namely, natural minerals and industrial residues/wastes.

1.1.1 Natural minerals
Among natural minerals olivine ((Mg, Fe$^{2+}$)$_2$SiO$_4$), serpentine (Mg$_3$Si$_2$O$_5$(OH)$_4$) and wollastonite (CaSiO$_3$) are being investigated due to their abundance and ease of accessibility (Kirchofer et al., 2012; Pan et al., 2012). The overall reaction of accelerated mineral carbonation can be summarized in equation 1 (Xiao et al., 2014):

$$(Mg, Ca)_x Si_y O_{x+2y+2} H_{2z}(s) + x CO_2(g) \rightarrow x(Mg, Ca)CO_3(s) + y SiO_2(s) + z H_2O (l) \quad (1)$$

The problem with using natural mineral is their low conversion rate, the need for material extraction from ores and transport of raw material. To increase the reaction rate, pre-treatment of raw material, such as size reduction through grinding or heat treatment can be applied which in turn leads to increased energy usage (Pan et al., 2012; Kirchofer et al., 2012).
1.1.2 Industrial residues
Cement kiln dust (CKD), steelmaking slag (SS), fly ash (FA), air pollution control residues (APC), paper mill residues are some of industrial wastes that can be used as alkaline source. The advantage of using these industrial residues is their higher activity towards carbonation due to presence of alkaline-containing oxides and hydroxides, which eliminates the need for extracting the reactive ions from the silicate matrix. In addition, these residues are usually already reduced in size, eliminating or decreasing the need for size reduction. Extraction and transport of the material can also be eliminated since these residues are usually generated at the site of CO₂ emissions. Using waste to produce useful products increases its value and at the same time leads to environmental benefits. Instead of disposal, the waste is transformed into valuable (even if of low value) products that can replace other products along with eliminating all the activities involved in producing and disposing them (Pan et al., 2012; Kirchofer et al., 2012; Xiao et al., 2014; Zingaretti et al., 2013).

The chemistry and mechanisms of alkaline waste carbonation are slightly different from natural weathering. One disadvantage of using alkaline residues is the variety in their chemical composition as a result of different production paths and process conditions, making it difficult to understand and optimize the reaction pathways. Although the mechanisms are not well known, here is a summary of suggested reaction pathways obtained from literature. Similar to weathering process, a presence of liquid is required. The liquid forms a thin film around the alkaline waste particles and their pores. CO₂ from the bulk is diffused and dissolved in the process liquid and later hydrolyzed to carbonic acid (which is a rate limiting step). Next step consist of ionization of carbonic acid according to equation 2 and 3 (Pan et al., 2012):

\[
H_2CO_3 \leftrightarrow H^+ + HCO_3^-
\]  \hspace{1cm} (2)

\[
HCO_3^- \leftrightarrow CO_3^{2-} + H^+
\]  \hspace{1cm} (3)

This happens simultaneously as leaching of the calcium- and/or magnesium ions caused by the acidic environment. In some cases, as for steelmaking slag, the calcium- or magnesium oxide phases are first hydrated to alkaline hydroxides as shown in equation 4. The alkaline ions of hydroxides then diffuse to the reaction site where carbonation, resulting in nucleation and precipitation of carbonates (Fernández Bertos et al., 2004; Pan et al., 2012). The nucleation step is favored by slightly increased temperature. Carbonation forms a precipitate layer at the surface of the particles or the pores, leading to possible pore blockage. For further carbonation, the reactants have to pass through the precipitate coating into the interior of the particles, which causes diffusion limitations (Fernández Bertos et al., 2004). Equations 5 to 8 present the reaction pathways involved in industrial residues carbonation (Pan et al., 2012).

\[
\text{CaO(s) or MgO(s) + H}_2\text{O (l) } \rightarrow \text{Ca(OH)}_2(\text{aq}) \text{ or Mg(OH)}_2(\text{aq})
\]  \hspace{1cm} (4)

\[
\text{Ca(OH)}_2(\text{s}) + \text{CO}_2(\text{g}) \rightarrow \text{CaCO}_3(\text{s}) + \text{H}_2\text{O (l)}
\]  \hspace{1cm} (5)

\[
\text{Mg(OH)}_2(\text{s}) + \text{CO}_2(\text{g}) \rightarrow \text{MgCO}_3(\text{s}) + \text{H}_2\text{O (l)}
\]  \hspace{1cm} (6)

\[
\text{CaO(s) + CO}_2(\text{g}) \rightarrow \text{CaCO}_3(\text{s})
\]  \hspace{1cm} (7)
Kirchofer et al. (2012) performed a life-cycle assessment on carbonation of various alkaline feedstocks in order to compare their carbonation potential. Capturing 1000 tonnes of CO\textsubscript{2} per day (t-CO\textsubscript{2}/day) was set as functional unit while including extraction of raw material, pre-treatment-, transportation-, post processing requirements and potential usage or disposal of the product. The study was based on best experimental values obtained from literature. The results showed that the maximum net CO\textsubscript{2} reduction was achieved from carbonation of CKD corresponding to 858 t-CO\textsubscript{2} per day followed by olivine (747 t-CO\textsubscript{2}/day), SS (656 t-CO\textsubscript{2}/day), FA (552 t-CO\textsubscript{2}/day) and serpentine (346 t-CO\textsubscript{2}/day), proving that the net carbonation potential of industrial residues are higher than minerals (Kirchofer et al. 2012).

1.2 Improvements and limitations

Storing CO\textsubscript{2} in form of solid carbonates is a stable and safe solution, eliminating disadvantages of geological storage such as long-term monitoring and risk of CO\textsubscript{2} leakage. Another advantage is that carbonation has shown to improve leaching properties of the feed material and decrease its porosity, making the product suitable for use in e.g. construction (Huijgen et al., 2006; Morone et al. 2014). Among other benefits of alkaline carbonation is that the reaction is thermodynamically favorable, i.e. it is an exothermic reaction. The reaction heat can be recovered and used to lower the energy penalties (Pan et al., 2012; Giannoulakis et al., 2014).

Even though the alkaline carbonation process is thermodynamically favorable, the kinetics is slow. In accelerated carbonation the reaction kinetics is enhanced through feedstock pretreatment, optimized reaction conditions, increased CO\textsubscript{2} concentration and/or input of more reactive feedstock (Pan et al. 2012).

Optimizing reaction conditions is a challenging task especially regarding temperature and pH-value. Increasing the temperature enhances the carbonation reaction rate while having an opposite effect on dissolution of carbon dioxide into the reaction solution. The dissolution of carbonation active species is favored at low pH values while the formation of carbonates is favored at high pH values (Pan et al. 2012). Therefore, there are variety of processes being studied aiming at finding a carbonation route that can result in maximized yield while minimizing the material or energy requirements.

There are two main routes of accelerated carbonation, namely the direct and indirect carbonation which are distinguished by the steps involved in the carbonation process. Direct carbonation refers to a process where carbonation of the alkaline material occurs in a single step. Indirect carbonation consists of first extracting the carbonation active components from the solid matrix followed by a separate carbonation step. In addition to characterizing the carbonation processes as direct and indirect, they can also be categorized into gas/solid- or liquid/solid carbonation (Pan et al., 2012; Giannoulakis et al., 2014; Nduagu et al. 2012).

Direct carbonation is a simpler process making it easy to improve. It can be carried out in either a gas/solid- or liquid/solid reaction medium. The reaction rate is very slow in the gas-solid carbonation route but this route is more economical than the liquid/solid route due to
absence of the need for treatment of reaction solution. The solid/liquid route can in turn be categorized as slurry or wet route depending on the ratio between the solvent and the solid. Usually the liquid/solid ratio, $L/S$, lies between 5 to 0.2 l/kg. For $L/S > 1.5$ l/kg the process falls under slurry route category while processes with $L/S < 1.5$ are considered as wet route. Slurry route requires after-treatment of process liquid (Zingaretti et al., 2013; Pan et al., 2012).

Indirect carbonation, oppose to direct carbonation, is more complicated and for this route a solvent recovery unit is necessary which contributes to significant amount of energy requirements. There is however, the advantage of making use of the reaction energy released during carbonation which is higher than for direct case.

Besides optimizing the reaction conditions, other improvements involve feedstock pretreatment, increased $\text{CO}_2$ concentration and/or input of more reactive feedstock. Potential feedstock was discussed previously (Pan et al., 2012). Increasing $\text{CO}_2$ concentration requires an additional step of capturing and compressing $\text{CO}_2$ prior to carbonation reaction. Size reduction through grinding or thermal activation of the raw material is an example of feedstock pretreatment. These activities results in additional energy, material and transportation requirements which can lead to a higher net $\text{CO}_2$ emissions, making it an insufficient solution. Therefore, it is important to perform a life cycle assessment, including all the activities and units involved in production of carbonates from extraction of raw material till disposal of the product.

1.3 State of art

As mentioned earlier there is variety of carbonation processes under research and development. However, there are two well-known carbonation processes which are used as reference for many carbonation routes and basis for many LCA studies carried out on alkaline $\text{CO}_2$ sequestration. The processes are firstly developed by National Energy Technology Laboratory (NETL) and Åbo Akademi University (ÅAU), which are direct and indirect carbonation respectively. A short and general description of these processes is provided here followed by a summary of LCA studies performed on NETL and ÅAU.

1.3.1 NETL

The NETL process is a direct, liquid/solid carbonation process and can be applied to minerals such as olivine, serpentinite and wollastonite. Prior to carbonation, the minerals go through mechanical treatment such as grinding and crushing. Next step is the activation of the minerals either thermally (heating) or mechanically (another grinding step). The activated minerals are then mixed with water and pumped into the reactor. A solution of 0.64 M $\text{NaHCO}_3$ and 1 M $\text{NaCl}$ is added in order to increase the pH and enhance the precipitation. The compressed $\text{CO}_2$ is also fed to the reactor and carbonation occurs. The reaction medium is then sent to a gas separator where the un-reacted $\text{CO}_2$ is separated and sent back to the reactor. The process liquid is also separated from the solid carbonates downstream from the reactor and recycled (Giannoulakis et al., 2014; Nduagu et al., 2011).
1.3.2 ÅAU

This indirect, gas/solid process has shown to be suitable only for production of carbonates from serpentine. Similar to NETL, ÅAU consists of first mechanically pre-treating the mineral through grinding. This followed by extraction of carbonation active species, in this case Mg$^{2+}$ ions. The extraction is an endothermic reaction and is carried out by an ammonium sulfate solvent. After dissolving of the undesired products, two precipitation steps are performed. In the first precipitation step calcium and iron hydroxides are formed and separated for use, while in the second precipitation the desired magnesium hydroxide is produced (Giannoulakis et al., 2014; Khoo et al., 2011; Nduagu et al., 2011).

The produced magnesium hydroxide is then sent to a pressurized fluidized bed reactor at 50 bar and 550 °C, while the solvent is sent to be recovered. The compressed CO$_2$ is also fed into the reactor, allowing carbonation to occur. The un-reacted CO$_2$ is sent back to the compressor and later fed back to the reactor. It is known that the carbonation of magnesium hydroxides is more thermodynamically favorable than the reaction path in the NETL where the active ions react directly after breaking free from the silicate matrix. In addition, steam is produced which can pass a turbine to produce electricity. Recovering the reaction heat and the produced steam is useful for lowering the external heat requirements. However, the solvent recovery is a great electricity consumer making up a disadvantage (Giannoulakis et al., 2014; Khoo et al., 2011; Nduagu et al., 2011).

1.4 Literature review

A study performed by (Giannoulakis et al., 2014) which perhaps is the most detailed and complete LCA study on accelerated carbonation so far, compares the environmental impacts of applying the NETL and ÅAU process to a natural gas combined cycle (NGCC) and pulverized coal (PC) power plant with the purpose of capturing the CO$_2$ emitted per MWh electricity generated. According to Giannoulakis et al. (2014) while mineral carbonation results in 15 – 64 % reduction in GHG emission, an increase in the overall contributions to other environmental impact categories is observed. The authors also pointed out that this method is not applicable for a PC power plant as a total positive contribution to global warming potential was observed. Heat and energy requirement which in turn are related to mining, processing and fuel combustion, were shown to be the main contributors to other environmental impacts. Another comparative study by Nduagu et al. (2011) also showed that the ÅAU requires less energy for carbonating 1 tonne of CO$_2$ while resulting in higher net CO$_2$ reduction. However, this study was based on carbonation of serpentine only.

Xiao et al. (2014) performed an LCA to compare different direct and indirect routes of steelmaking slag carbonation. The direct routes were shown to provide higher carbonation conversion i.e. 56.6 – 93.5 %, compared to indirect routes with conversions of 25.5 – 59 %. However, direct routes occur at higher temperatures needing more energy for heating, which sets off their life cycle capture potential. The indirect routes require less energy but on the other hand there is a need for chemical input. Looking at the whole life cycle, lower GWP was achieved for indirect routes compared to direct routes, indicating that energy consumption due to heating is the main source of CO$_2$ emissions.
1.5 Problem formulation and aim (Goal definition)

In the previous life cycle assessment studies done on accelerated carbonation the main focus was to examine and compare the direct and indirect carbonation methods. As for the direct route, only slurry route has been studied. In this paper an LCA was performed on comparing wet and slurry carbonation of steelmaking slag. Four scenarios were examined, namely two basic and two improved scenarios. The basic scenarios refer to a slurry and wet route carbonation where all of the products are disposed at landfill. These scenarios are called Slurry1 and Wet1. In the improved scenarios, i.e. Slurry2 and Wet2, it is assumed that half of the products are landfilled while the other halves substitute limestone and natural aggregates respectively.

The starting point of an LCA is to decide the functional unit which is a fixed parameter and the basis for all calculations, process design and assumptions. As the development of alkaline carbonation is intended to capture the CO$_2$ emitted from a power plant, the functional unit is set to 1 MWh electricity produced from the power plant.

Next step consisted of choosing a power plant to which the carbonation processes could be implemented. Based on a literature survey, it was concluded that alkaline sequestration of CO$_2$ is more suitable for a natural gas based power plant than a coal fired power plant with regards to CO$_2$ emissions and environmental impacts. Another factor considered in the choice of power plant was the geographical location. Since the technical, social and economical factors affect the inventory and thereby the outcome of the LCA, the choices were narrowed down to global or European averages. In these processes an average value are obtained taking into account inventory data from several countries. This way the effect of the above mentioned factors on the outcome could be insignificant or eliminated. Since this study is a continuation of a study performed by a European institute (Zingaretti et al., 2013) it was considered relevant to choose a process based on European averages.

The criteria mentioned limited the choices to three natural gas power plants namely, a conventional 10 MW, a conventional 100 MW and a combined cycle 400 MW natural gas power plant. Finally, the decision was made based on the yearly CO$_2$ emission and the life cycle environmental impacts. The importance of yearly CO$_2$ emission is due to the fact that the access to steel slag is limited. An Italian steelmaking plant produces around 700 000 tons of steel slag per year. Based on carbonation reaction efficiencies, for both wet and slurry routes, the quantity of slag required for each of the above mentioned power plants were calculated and compared with the yearly production of a steelmaking plant. It was discovered that both of the conventional plants are suitable, however, the environmental impact of the bigger plant was of course higher, leading to the conclusion that the conventional natural gas power plant with 10 MW full capacity is the choice.
2 Principles and methodology of LCA

Life cycle assessment is a useful tool for predicting or evaluating environmental impacts of a product or a service or to compare the environmental impacts between different products or services. Performing an LCA in an early stage of a research can help determine whether the process or product under development is worth investing on. More importantly, an LCA helps to discover where in a product’s or service’s life chain improvements are required by pointing out which units/processes are the main causes of environmental impacts. As the name implies LCA takes into account all the processes involved in a product’s life, starting from extraction of raw material, processing of raw material and production, usage and finally disposal or recycling. All the intermediate processes such as transportation, maintenance and distribution are also considered in an LCA (Arvanitoyannis, 2008; Swedish Standards Institute, 2006; Caffery; Pré-Sustainability; Zbicinski, 2006).

ISO 14040 and 14044 set the framework of LCA, presenting guidelines and principles on LCA methodology. According to ISO 14040, LCA is composed of four steps, namely definition of goal and scope, inventory analysis, impact assessment and interpretation (Swedish Standards Institute, 2006). These steps interact through the whole assessment and should not be treated as independent steps. A brief description of these steps is presented here:

- **Definition of goal and scope**
  This is the most important stage of an LCA since it sets the constrains for the other three stages of the LCA. The goal is synonym to the purpose of the study and which audience is the LCA intended for (Arvanitoyannis, 2008). The scope of LCA includes building a detailed model of the life cycle, presenting the assumptions, process schemes and system boundaries, inputs and outputs (qualitatively) and the geological and technical coverage of the data. In addition, a functional unit must be determined which is a basis for the quantitative data collection and also a basis for presenting the impact assessments. The functional unit depends on which function that a product or a service fulfills and is especially important when a comparative LCA study is performed. (Arvanitoyannis, 2008; Swedish Standards Institute, 2006; Caffery; Pré-Sustainability).

- **Inventory analysis**
  In this step all the input- and output flows are gathered and quantified related to the functional unit. Inputs and outputs can mean raw material and energy requirements, co-products and waste production, emissions to air, water and soil (Arvanitoyannis, 2008; Swedish Standards Institute, 2006; Caffery et al.; Pré-Sustainability). In some cases data are accessed from databases while in other cases data must be collected internally. In general, a common issue faced while performing an inventory analysis is lack of data, which may lead to redefining the goal or scope of LCA (Caffery et al.). It is possible to predict the environmental issues through inventory analysis, however it is time consuming and requires years of experience.
**Impact assessment**

The impact assessment consists of two compulsory steps, i.e. classification and characterization, and two optional steps namely normalization and weighting. The data obtained during inventory analysis are divided into categories depending on the effects they have on the environment. This procedure is referred to as classification. To quantify the contribution of each substance to the relevant impact category, the amount of each substance is multiplied by an equivalence factor. The equivalence factor relates the impacts of each substance to the impact of a reference substance in the respective category. This step is called characterization. The normalization and weighting steps are performed to compare the impact categories among themselves (Caffery et al.; Pré-Sustainability; Zbicinski, 2006).

Environmental impact categories are sorted into two main categories: mid-point (problem-oriented) and end-point (damage-oriented) impacts. Examples of mid-point categories are acidification, climate change, human toxicity. The end-point categories are divided into human health, resource depletion and ecosystem quality (Pré-Sustainability; UNEP, 2011).

Depending on which environmental impacts are of interest in a study a selection of LCA method must be done at the stage of the scope definition. CML 2001, ReCiPe and Eco-indicator 99 are some of the commonly used LCA methods (Acero et al., 2014). In addition to what categories are considered, they differ in geological coverage, calculation methods, assumptions and units.

The impact categories considered in CML 2001 method, which is used in this study, are briefly described here. CML 2001 categories are all mid-point categories while ReCiPe considers both mid-point and en-point categories.

- **Abiotic depletion and abiotic depletion fossil fuel:** refers to extraction of non-biological raw material or fuels respectively. The calculation method is based on the extraction rate and accessibility of the resources and is measured in kg Sb-equivalent and MJ respectively (Oers et al. 2002).
- **Acidification:** is due to emission of sulfur- and nitrogen oxides which in contact with water hydrolysis to acids. Acidification is measured in kg SO\textsubscript{2}-equivalent.
- **Ecotoxicity:** is a common name for fresh water aqueous ecotoxicity, marine ecotoxicity and terrestrial ecotoxicity impact categories. The impacts are measured in terms of kg 1,4-dichlorobenzene (1,4-DB) equivalence and are mainly caused by emission of heavy metals (Acero et al., 2014).
- **Eutrophication:** is a consequence of enrichment of chemical nutrients in an ecosystem which leads to abnormal productivity. Emissions of ammonia, nitrates, NO\textsubscript{x} and phosphorus to soil, water and air are the main causes to eutrophication. The impacts are measured in kg PO\textsubscript{4}\textsuperscript{3-}equivalent. (Ministry for the Environment)
- Global warming potential: refers to emission of greenhouse gases and is measured in kg CO\textsubscript{2} equivalent.

- Human toxicity: is based on potential damage caused by emission of a unit substance considering toxicity and dose. Examples of substances classified under this category are arsenic, sodium dichromate and hydrogen fluoride. The emissions of these compounds are mainly caused by electricity production from fossil fuels. The unit of measurement is kg 1,4-DB equivalence (Acero et al., 2014).

- Ozone layer depletion: is caused by emission of CFC-compounds, i.e. compounds containing Cl, F and/or Br, which catalyse the degradation of the ozone layer reducing its ability to absorb UV-B lights which is harmful to humans and the ecosystem. The ozone layer depletion potential of a substance is measured relative to impact of 1 kg of CFC-11 i.e. trichlorofluoromethane (CCl\textsubscript{3}F) (Norihiro et al., 2012; Acero et al., 2014; Steinmann, 1980).

- Photochemical oxidation: or formation of ground level ozone is a phenomena caused by reaction between volatile organic compounds, NO\textsubscript{x} and UV-light. The reaction product is ozone which is a strong oxidation agent causing e.g. oxidation of proteins. The impacts are expressed as kg C\textsubscript{2}H\textsubscript{4} (ethylene) equivalence (Acero et al., 2014).

- **Interpretation**

  This step is integrated into other three stages and consists of analyzing, evaluating and comparing the results (Arvanitoyannis, 2008; Swedish Standards Institute, 2006; Caffery; Pré Sustainability).
3 Carbonation processes investigated in this work

This study is a comparative LCA of two carbonation processes using steel slag as alkaline source. The LCA is based on a study done by Zingaretti et al. (2014) who evaluated the material and energy requirements for different scenarios of wet and slurry routes. The aim of the study was to examine the effect of parameters such as type of feedstock, solid/liquid ratio and operating conditions on material and energy requirements. A description of the different routes, assumptions and calculations will be presented in this section as well as the provided data. This will proceed with the calculations necessary for implementing the processes in SimaPro. SimaPro is an LCA software which uses mathematical models to calculate the impacts of any processes. It includes different LCA methods and inventory obtained from different databases. In this study SimaPro v8.0.5 and Ecoinvent 3.1 were used.

3.1 Process description (Scope)

Figure 1 and Figure 2 show schematic descriptions of the slurry and the wet route carbonation, respectively. The basis of this study is to capture 90 percent of CO\(_2\) emitted by a power plant. Since the amount of CO\(_2\) is fixed, the capture and compression units are identical for both routes. It is assumed that the flue gas from the power plant goes through an MEA scrubbing capture unit, where 90 percent of CO\(_2\) content of the flue gas is separated. The flow going out of capture unit is assumed to consist of pure CO\(_2\), which is compressed and then fed into the carbonation reactor. The power plant and capture unit are placed outside the system boundaries.

In the slurry route (Figure 1), the size of the slag particles are first reduced through grinding and later fed into a mixer where it is mixed with water to form a slurry. The mixed slurry is then pumped into a heat exchanger, where it meets the hot carbonated slurry leaving the reactor. In the heat exchanger the cold slurry is heated to a temperature that is 30 K below the reaction temperature. For further heating, the slurry flows through a heater to reach the reaction temperature. The heated slurry and CO\(_2\) stream coming from the compressor are fed to the reactor to allow for carbonation to occur. The carbonated slag returns to the heat exchanger before entering the solid-liquid separation units. The separated water is transported to wastewater treatment while the solid product is transported to site of landfill.

The wet route (Figure 2) is a simpler process, consisting of fewer units. As for the slurry route, the wet route starts with size reduction of the slag. The ground slag enters the reactor, which is similar to a rotary kiln, and is heated to the reaction temperature. Water is then sprayed into the reactor to wet the solid, hence, the name wet route. The slag reacts with CO\(_2\) and forms carbonated products that are transported to landfill.
Figure 1. Schematic description of slurry route carbonation of BOF slag considered in this study.

Figure 2. Schematic description of wet route carbonation of BOF slag considered in this study.
3.1.1 Operating conditions and background data

As mentioned earlier, different scenarios of wet and slurry route carbonations were studied by Zingaretti et al. (2014), which differed in type of residues used and reaction conditions. For this study the scenarios with BOF slag as feedstock was chosen.

Table 1. Initial composition of BOF slag type chosen for this study

<table>
<thead>
<tr>
<th>Compound</th>
<th>Mass fraction [wt%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ca$_2$SiO$_4$</td>
<td>17.2</td>
</tr>
<tr>
<td>Ca(OH)$_2$</td>
<td>28.0</td>
</tr>
<tr>
<td>Mg(OH)$_2$</td>
<td>11.5</td>
</tr>
<tr>
<td>CaCO$_3$</td>
<td>8.4</td>
</tr>
<tr>
<td>Fe$_2$O$_3$</td>
<td>34.9</td>
</tr>
</tbody>
</table>

The chemical composition of the slag is presented in Table 1. The slag consists mainly of Ca-compounds in the form of Ca$_2$SiO$_4$ and Ca(OH)$_2$, and smaller amount of Mg(OH)$_2$. Only 45.3% of the slag consists of Fe$_2$O$_3$ and CaCO$_3$ which are considered not to participate in carbonation. According to the compositions of the slag the following reactions were assumed to occur during carbonation:

\[
\text{Ca}_2\text{SiO}_4 + 2\text{CO}_2 \rightarrow 2\text{CaCO}_3 + \text{SiO}_2 \quad (9)
\]

\[
\text{Ca(OH)}_2 + \text{CO}_2 \rightarrow \text{CaCO}_3 + \text{H}_2\text{O} \quad (10)
\]

\[
\text{Mg(OH)}_2 + 2\text{CO}_2 + \text{CaO} \rightarrow \text{MgCa(CO}_3)_2 + \text{H}_2\text{O} \quad (11)
\]

Table 2 lists the operation conditions. Particle size of the ground slag for both processes is 150 mm and partial pressure of CO$_2$ is 10 bar. The processes differ in L/S ratio, which is 5 for the slurry route and 0.3 for the wet route, and the reaction temperature. Carbonation occurs at a lower temperature in the wet route than the slurry route, i.e. 323 K and 373 K respectively.

Table 2. Operating conditions of the studied processes (Zingaretti et al., 2014)

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Slurry</th>
<th>Wet</th>
</tr>
</thead>
<tbody>
<tr>
<td>$d_0 \cdot 10^{-3} \text{[m]}$</td>
<td>1.19</td>
<td>1.19</td>
</tr>
<tr>
<td>$d_1 \cdot 10^{-3} \text{[m]}$</td>
<td>0.15</td>
<td>0.15</td>
</tr>
<tr>
<td>$L/S \text{[l/kg]}$</td>
<td>5</td>
<td>0.3</td>
</tr>
<tr>
<td>$p_{CO_2} \text{[MPa]}$</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>$T \text{[K]}$</td>
<td>373</td>
<td>323</td>
</tr>
<tr>
<td>$t_R \text{[h]}$</td>
<td>1</td>
<td>1</td>
</tr>
</tbody>
</table>
4 Methodology

Having defined the goal and scope, the next step consisted of data collection through material- and energy balances, i.e. the inventory. In this section calculations and assumptions necessary for performing the LCA study are presented. Some of the data were provided by Zingaretti et al. (2014) while the rest were calculated. Finally, the assumptions made for implementing and programming the processes in SimaPro were provided. The results of LCA are summarized in section 5.

4.1 Material balance (Inventory)

The first step is to determine the quantity of material needed to uptake the CO$_2$ flow, which is the basis of the calculations. The amount of slag needed for the carbonation reaction was calculated considering both the reactivity ($R_x$) and carbonation potential of the slag ($R_{CO_2}$). The reactivity at the given operational conditions was obtained from experimental values. The potential of the slag is expressed as the mass ratio between the mass of slag required and a unit mass of CO$_2$ and is estimated according to equation 12a (O’Connor et al., 2005; Zingaretti et al., 2013). In this equation only the carbonation of calcium and magnesium phases are considered while ignoring the reactivity of iron species due to its low concentration (see Table 1).

\[
R_{CO_2} = \frac{100}{M_{CO_2} \Sigma c_{Ca^{2+}} + c_{Mg^{2+}}}
\]  

(12a)

Taking the ratio of the carbonation potential and reactivity the amount of slag needed to capture a unit mass of CO$_2$ is obtained. Multiplying the ratio with the mass of CO$_2$ to be stored, the total slag requirement is obtained. (See equation 12b)

\[
R_{eff} = \frac{R_{CO_2}}{R_x}
\]  

(12b)

The obtained data and the results are summarized in Table 3. Also, the water requirement is presented in Table 3, which is obtained by simply multiplying the mass of slag with the liquid-solid ratios presented in Table 2. Based on reaction formulas 9 to 11 the mass of reaction products were also calculated.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Slurry</th>
<th>Wet</th>
</tr>
</thead>
<tbody>
<tr>
<td>$R_{CO_2}$ [kg/kg CO$_2$]</td>
<td>2.93</td>
<td>3.93</td>
</tr>
<tr>
<td>$R_x$</td>
<td>0.73</td>
<td>0.39</td>
</tr>
<tr>
<td>$R_{eff}$ [kg/kg CO$_2$]</td>
<td>4</td>
<td>10</td>
</tr>
<tr>
<td>$m_s$ [kg/MWh$_{el}$]</td>
<td>2191.3</td>
<td>5478.3</td>
</tr>
<tr>
<td>$m_{H_2,oin}$ [kg/MWh$_{el}$]</td>
<td>10956.6</td>
<td>1643.5</td>
</tr>
<tr>
<td>$m_{es}$ [kg/MWh$_{el}$]</td>
<td>2572.5</td>
<td>5878.9</td>
</tr>
<tr>
<td>$m_{H_2,out}$ [kg/MWh$_{el}$]</td>
<td>11123.1</td>
<td>0</td>
</tr>
<tr>
<td>$m_{CO_2}$ ($FG$) [kg/MWh$_{el}$]</td>
<td>608.7</td>
<td>608.7</td>
</tr>
<tr>
<td>$m_{CO_2}$ [kg/MWh$_{el}$]</td>
<td>547.8</td>
<td>547.8</td>
</tr>
</tbody>
</table>
4.2 Energy balances (Inventory)

Compression
To calculate the energy requirement for compressing CO$_2$, equation 13 was applied which is valid for compressing CO$_2$ within the range of 0.1 to its supercritical pressure of 7.38 MPa. It was assumed that the compression was carried out in a 5-stage compressor (Zingaretti et al. 2014; McCollum et al., 2006).

\[
E_{\text{stage},i} = \left( \frac{Z_i R T_i m_{CO_2\gamma_{i,s}}}{M_{CO_2\gamma_{i,s}}} \right) \left( k_s \right)^{\frac{1}{k_s-1}} \left( \frac{C_R}{k_s} \right)^{k_s-1} \left( \frac{k_s}{k_s-1} \right) - 1
\]

(13)

\[
k_s = \frac{C_p}{C_v}
\]

(14)

\[
C_R = \left( \frac{P_{\text{cut}}}{P_{\text{initial}}} \right) \frac{1}{R}
\]

(15)

Grinding
The energy requirement for grinding was calculated according to equation 16 with regard to the initial and final particle size (see Table 2) (Zingaretti et al. 2014). The value for Bond’s work index ($E_i$) is obtained from literature and is specific for different type of material. For slag this value corresponds to 15.72 (Perry et al., 1997).

\[
E_g = 10 \ E_i \left( \frac{1}{\sqrt{d_1}} - \frac{1}{\sqrt{d_0}} \right)
\]

(16)

Heating
For the slurry route the heating requirement is defined as the sum of the energy required to reach the reaction temperature, the heat recovered in the heat exchanger, the reaction heat and heat loss from carbonation reactor. In this case it was assumed that the reaction heat is sufficient to heat the cold slurry to the reaction temperature, eliminating the external heating requirement. Since it is not possible to recover the reaction heat from solid phase, for the wet route heating requirement was calculated mainly based on the energy needed to raise the temperature of the ingoing material to the reaction temperature. Equation 17 was used to calculate the heating requirements (Zingaretti et al., 2014).

\[
E_{\text{heat}} = \sum_i m_i C_{p,i} \Delta T / m_{CO_2}
\]

(17)

Liquid/solid separation
The separation was considered to be performed in two steps. As the carbonated slurry leaving the reactor consists of approximately 19 weight % solid, a clarifier was considered feasible as the first S/L separator. Assuming that the efficiency of the clarifier is 75 %, that is it removes 75 % of the water content in the inflowing slurry, the solid content increases to 48 wt% after the clarifier. This makes up the criteria for the next separation step. A centrifugal filter was found to be a suitable choice and it was assumed that it increases the solid content to 90 wt%.
To obtain the energy requirement of the clarifier equation 18 was used assuming a scrapping velocity of 60 s$^{-1}$ (Zingaretti et al. 2014).

$$ E_{\text{clarifier}} = \frac{\nu \mu g^2}{m_{\text{CO}_2}} $$

(18)

The energy consumption for the centrifugal filter was obtained from Perry’s Chemical handbook, assuming that a conical-screen centrifugal filter is used. The energy consumption of this type of machines ranges from 0.3 – 1 kWh/ton dry solid (Perry et al., 1984). Due to high solid content of the ingoing and outgoing flows the highest value in this range was chosen i.e. 1 kWh/ton solid separated. The total energy consumption was obtained accordingly:

$$ E_{\text{cf}} = 1 \frac{kWh}{\text{ton dry solid}} \cdot 3.6 \frac{kWh}{kWh} \cdot m_{cs} $$

(19)

**Mixing**

The energy requirement for mixing was calculated with equation 18, assuming an impeller velocity of 500 s$^{-1}$.

**Pumping**

According to Zingaretti et al. (2014), the energy requirement for pumping was calculated based on the flow rate of the slurry, pressure in the carbonation reactor and friction through pipelines. Assuming an isentropic pump efficiency of 80% the pumping energy requirement was calculated and is presented in Table 4.

**Rotation**

The energy requirement for the wet route reactor was calculated based on the power required for rotation and the total mass including the mass of reactants and the reactor. The power of rotation was calculated according to equation 20:

$$ P_{rot} = M_{rot} \omega $$

(20)
Summary
The energy requirements for each step of the carbonation routes are summarized in Table 4 and Table 5 and are related to Figure 1 and Figure 2.

Table 4. Energy requirements for slurry route

<table>
<thead>
<tr>
<th>Energy requirement</th>
<th>Value [kJ/MWh_el]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$E_{agg}$</td>
<td>22254.8</td>
</tr>
<tr>
<td>$E_{cf}$</td>
<td>10289.9</td>
</tr>
<tr>
<td>$E_{clarifier}$</td>
<td>95.4</td>
</tr>
<tr>
<td>$E_{comp}$</td>
<td>158894.0</td>
</tr>
<tr>
<td>$E_g$</td>
<td>63145.4</td>
</tr>
<tr>
<td>$E_{heat}$</td>
<td>0</td>
</tr>
<tr>
<td>$E_{mix}$</td>
<td>2225.5</td>
</tr>
<tr>
<td>$E_p$</td>
<td>18354.3</td>
</tr>
</tbody>
</table>

Table 5. Energy requirements for wet route

<table>
<thead>
<tr>
<th>Energy requirement</th>
<th>Value [kJ/MWh_el]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$E_{rot}$</td>
<td>31952.3</td>
</tr>
<tr>
<td>$E_{comp}$</td>
<td>158894.0</td>
</tr>
<tr>
<td>$E_g$</td>
<td>126291</td>
</tr>
<tr>
<td>$E_{heat}$</td>
<td>448064.6</td>
</tr>
</tbody>
</table>
4.3 Construction of carbonation plant (Inventory)

In order to obtain information on construction of the carbonation plant, first a scheme was drawn marking out the units/steps involved in the carbonation plant. This is already presented in Figure 1 and Figure 2. The next step was to decide what is to be included in the infrastructure. In this study the construction of equipments was the main concern. Land occupation and transportation roads are also considered as part of the infrastructure. However, as it was assumed that the carbonation plant will be built close to or inside a power plant, it would mean that the area, in which the carbonation plant is to be built, has already been transformed to an industrial area. Hence, the environmental impacts associated with land occupation are not specifically related to the building of the carbonation plant. The same applies for transportation, assuming that the roads necessary for transportation already exists.

For the construction of the equipments, it was necessary to determine the dimensions, type of equipments and type of construction material. Dimensions of the reactors, solid-liquid separator and the mixer were provided by Zingaretti et al. (2014), which are presented in Table 6 and Table 7. As for the rest of the units some were found existing in SimaPro which through some modification could be used in this study, while the remaining units were dimensioned based on literature.

### Table 6. Dimensions of carbonation reactors

<table>
<thead>
<tr>
<th>Dimensions [m]</th>
<th>Slurry</th>
<th>Wet</th>
</tr>
</thead>
<tbody>
<tr>
<td>$h$</td>
<td>3</td>
<td>8</td>
</tr>
<tr>
<td>$r_l$</td>
<td>1.5</td>
<td>1.47</td>
</tr>
<tr>
<td>$t_{st,1}$ ($1^{st}$ layer)</td>
<td>0.005</td>
<td>0.03</td>
</tr>
<tr>
<td>$t_{gw}$</td>
<td>0.01</td>
<td>-</td>
</tr>
<tr>
<td>$t_{st,2}$ ($2^{nd}$ layer)</td>
<td>0.002</td>
<td>-</td>
</tr>
</tbody>
</table>

### Table 7. Dimensions of mixer and clarifier

<table>
<thead>
<tr>
<th>Dimensions [m]</th>
<th>Mixer</th>
<th>Clarifier</th>
</tr>
</thead>
<tbody>
<tr>
<td>$h$</td>
<td>3</td>
<td>3</td>
</tr>
<tr>
<td>$r_l$</td>
<td>1.5</td>
<td>2.5</td>
</tr>
</tbody>
</table>

Due to corrosion, a stainless steel alloy called steel 304L, containing 18% chromium and 8% nickel and having density of 8030 kg/m$^3$ (AK Steel, 2007) was chosen as construction material for slurry route reactor. Since the same type of material flows through the mixer, heat exchanger and the wet route reactor, the same material was chosen for constructing these units.

As for the other units the following were found in SimaPro: a compressor and a pump. The heat exchanger, clarifier and centrifugal filter were to be dimensioned. The assumptions and calculations related to construction of the plant will be presented shortly. A general assumption made for simplifying the calculations was to consider that the material usage for construction of the units consists of the main component of the unit and the material usage for other parts are negligible. For example, for the mixer, only the cylinder is considered in
calculating the amount of construction material required, while neglecting the need for usage of screws, support, agitator, etc.

One important consideration regarding construction is to calculate what portion of the equipment is used per functional unit. This was done based on two main factors, lifetime and capacity of the equipment. The lifetime factor is a constant and is identical for all equipment. The environmental impacts caused due to production of equipment are a one-time contribution and should be distributed over the lifetime of the carbonation plant. Assuming that the equipment has the same lifetime as the power plant, i.e. 25 years, the lifetime factor was calculated accordingly:

\[ p_{\text{lifetime}} = \frac{1}{c \cdot \text{MWh}_{\text{el}} / \text{year} \cdot 25 \text{ years}} = \frac{1}{50000 \text{MWh}_{\text{el}} / \text{year} \cdot 25 \text{ years}} = 8 \cdot 10^{-7} \cdot \frac{p}{\text{MWh}_{\text{el}}} \]  

(21)

The other factor differs for each unit and was calculated individually; mainly based on taking the ratio between the volume of the medium flowing through the equipment and the volumetric capacity of the equipment. This factor will be referred to as volumetric portion. The total portion was then defined as a product of the lifetime factor and the volumetric portion. In the case of the pump using the volumetric portion was irrelevant and only lifetime factor was applied. In the following pages the calculations and results related to construction of the carbonation plant are presented.

**Mixer**

It was assumed that the mixer is shaped as a circular cylinder and the wall thickness to be 0.005 m. In order to calculate the mass of steel used for the mixer, it was assumed that the mixer consists of two cylinders, with the smaller one (cylinder 1) having the dimensions presented in Table 7. The mass of steel needed were calculated according to equations 22 – 25. The results of these calculations are summarized in Table 8.

\[ V_1 = \pi r_1^2 h_1 \]  

(22)

\[ V_2 = \pi r_2^2 h_2 = \pi (r_1 + 2 \cdot t_{st1})^2 (h_1 + 2 \cdot t_{st1}) \]  

(23)

\[ V_{st} = V_2 - V_1 \]  

(24)

\[ m_{st} = V_{st} \cdot \rho_{st} \]  

(25)

**Table 8. Data for mixing**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>(V_1) [m³]</td>
<td>21.20</td>
</tr>
<tr>
<td>(V_2) [m³]</td>
<td>21.38</td>
</tr>
<tr>
<td>(V_{st}) [m³]</td>
<td>0.1772</td>
</tr>
<tr>
<td>(m_{st}) [kg]</td>
<td>1422.8</td>
</tr>
<tr>
<td>(V_{mf}) [m³]</td>
<td>19.8</td>
</tr>
<tr>
<td>(V_{st}) [m³]</td>
<td>9.75</td>
</tr>
<tr>
<td>(p_{mix}) [p/MWh]</td>
<td>3.94 (\cdot 10^{-7})</td>
</tr>
</tbody>
</table>
Finally, the portion of the mixer used per functional unit was calculated. Firstly, full capacity of the mixer was obtained based on the assumption that the highest allowed liquid level is 20 cm below the top. Taking the ratio between the volume of the slurry and full capacity volume of the mixer, the portion of mixer occupied by the slurry was calculated. This value was then multiplied by the lifetime factor. The calculations are presented in equations 26 – 31.

\[ m_{sl} = m_{H_2O} + m_s \]  \hspace{1cm} (26)

\[ x_{H_2O} = \frac{m_{H_2O}}{m_{sl}} \]  \hspace{1cm} (27)

\[ x_s = \frac{m_{slag}}{m_{sl}} \]  \hspace{1cm} (28)

\[ \rho_{sl} = x_{H_2O} \rho_{H_2O} + x_s \rho_s \]  \hspace{1cm} (29)

\[ V_{mfc} = \pi r_1^2 (h - 0.2) \]  \hspace{1cm} (30)

\[ p_{mix} = \frac{v_{sl}}{v_{mfc}} \cdot p_{lifetime} = \frac{\rho_{sl}}{v_{mfc}} \cdot \rho_{slag} \cdot p_{lifetime} \]  \hspace{1cm} (31)

Density of slag corresponding to 3094 kg/m$^3$, was obtained from (Zingaretti et al., 2014).

**Reactor, slurry**

The same assumptions as for the mixer were made for the slurry reactor. Hence, the calculations were carried out in the same manner. The only difference was to calculate the reactor portion based on the carbonated slurry instead of the un-reacted slurry. It was assumed that the reactor consists of two steel layers and a layer of glass wool in between for insulation purposes (Zingaretti et al., 2014). The thicknesses of these layers are presented in Table 6. The calculation results are summarized in Table 9 where $V_{st1}$ and $V_{st2}$ are the volume of the first and second steel layer respectively.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$V_{st1}$ [m$^3$]</td>
<td>0.1772</td>
</tr>
<tr>
<td>$\rho_{gw}$ [kg/m$^3$]</td>
<td>40.5</td>
</tr>
<tr>
<td>$V_{gw}$ [m$^3$]</td>
<td>0.3572</td>
</tr>
<tr>
<td>$V_{st2}$ [m$^3$]</td>
<td>0.0719</td>
</tr>
<tr>
<td>$m_{gw}$ [kg]</td>
<td>17.41</td>
</tr>
<tr>
<td>$m_{st}$ [kg]</td>
<td>2404.2</td>
</tr>
<tr>
<td>$\rho_{cst}$ [kg/m$^3$]</td>
<td>3178</td>
</tr>
<tr>
<td>$p_{cst}$ [p/MWh]</td>
<td>$4.06 \cdot 10^{-7}$</td>
</tr>
</tbody>
</table>

**Reactor, wet**

The carbonation reactor used for the wet route is similar to a rotary kiln and is assumed to be heated by a heating coil wire wrapped around the reactor. In addition to the heating coil, the
reactor is insulated with a layer of glass wool. Using the equation for volume of a circular cylinder and assuming that the top of the reactor is open, the amount of steel needed could be determined. To simplify the calculation for dimensioning of the heating coil, it was assumed that instead of a wire, it consist of a thin solid coat embracing the reactor. A heating coil made of copper was assumed. The thickness of the heating coat and glass wool layer was set to 0.0016 (SimaPro v8.0.5) and 0.01 m respectively.

The capacity of the wet route reactor was assumed to be approximately 10 % of its inner volume. This is based on the fact that a slag layer thickness of 7 cm is required to achieve a good carbonation performance (Zingaretti et al., 2014). The portion of the reactor taken up by the slag was then calculated based on this assumption and the lifetime factor. The results for wet reactor are presented in Table 10.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$m_{st}$ [m$^3$]</td>
<td>19684.6</td>
</tr>
<tr>
<td>$m_{Cu}$ [kg/m$^3$]</td>
<td>1081.5</td>
</tr>
<tr>
<td>$m_{gw}$ [kg]</td>
<td>17.41</td>
</tr>
<tr>
<td>$V_{wrfc}$ [m$^3$]</td>
<td>5.43</td>
</tr>
<tr>
<td>$p_{rw}$ [p/MWh]</td>
<td>$5.03 \cdot 10^{-7}$</td>
</tr>
</tbody>
</table>

### Heat exchanger

Through some research it was found that a spiral heat exchanger is a suitable choice for slurry phase medium. To design a spiral heat exchanger there are many parameters that should be considered, such as overall heat transfer coefficient, heat capacity, viscosity, heat conductivity etc. These parameters were not available and would lead to very complicated calculations and uncertainties would be difficult to estimate. Instead, a list of commercialized heat exchangers (AlfaLaval) was found and through some simple calculation a suitable heat exchanger was chosen, which was then taken as the basis for calculations of construction material requirements.

Assuming that the cold and hot streams have approximately the same heat capacities, the amount of heat flowing from the hot stream to the cold stream is calculated accordingly:

\[
q_{tot} = q_h + q_c
\]

\[
q = q_h - \left( \frac{q_{tot}}{2} \right)
\]

\[
Q = \frac{q}{t_{MWhe}}
\]

\[
t_{MW} = \frac{1}{C \cdot 3600 \text{ seconds/hour} \cdot 24 \text{ hours/day} \cdot 300 \text{ days/year}}
\]
The heat content of the cold and the hot streams were obtained from equation 17, corresponding to 242.7 and 291.9 MJ respectively. Equation 34 gives the transferred heat to be 474.7 kW. Based on this value the ALSHE STW 600 heat exchanger was chosen with the dimensions presented in Table 11.

Table 11. Dimensions of heat exchanger (AlfaLaval, Zhejiang)

<table>
<thead>
<tr>
<th>Dimension [m]</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>W</td>
<td>1.290</td>
</tr>
<tr>
<td>D</td>
<td>1.070</td>
</tr>
<tr>
<td>H</td>
<td>1.025</td>
</tr>
<tr>
<td>d_c</td>
<td>0.025</td>
</tr>
<tr>
<td>d_plate</td>
<td>0.002</td>
</tr>
</tbody>
</table>

A range of heat transfer areas is provided by AlfaLaval for a range of different heat exchangers. As the width and height of these heat exchangers are identical, but the depths are different it was concluded that the parameter on which the heat exchange area depends on is the depth. Having the heating area of the biggest and smallest heat exchanger, the heat transfer area for the chosen heat exchanger was calculated through linear interpolation.

Having the heat exchange area, the length of the metal plate which made up the spiral could be calculated according to equation 36 (Bidabadi et al., 2013; Picón-Núñez et al., 2007). To simplify the calculation, it was assumed that the spiral consists of a rectangular plate folded in half and then rolled up into a spiral. This forms two rolled up channels, one for the cold stream and one for the hot stream.

\[ L = \frac{A}{2d} \]  

The factor “2” in the denominator of equations 36 is considered here because the total heat exchange area is the sum of the surface area on both sides of the plate. The thickness of the heat exchanger shell plate was assumed to be the same as the thickness of the spiral plate. From these dimensions the volume of the steel needed for the plate and the shell could be calculated. The results are presented in Table 12.

Table 12. Data for heat exchanger

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>A [m²]</td>
<td>11.3</td>
</tr>
<tr>
<td>Q [kW]</td>
<td>474.7</td>
</tr>
<tr>
<td>L [m]</td>
<td>5.3</td>
</tr>
<tr>
<td>m_p[kg]</td>
<td>90.7</td>
</tr>
<tr>
<td>m_shell [kg]</td>
<td>111.9</td>
</tr>
<tr>
<td>m_S(Tot)[kg]</td>
<td>202.7</td>
</tr>
<tr>
<td>p_hx [p/MWh_al]</td>
<td>8 \cdot 10^{-7}</td>
</tr>
</tbody>
</table>
As for calculating the portion of heat exchanger, it was assumed that heat exchange is performed as a continuous process i.e. 1 unit of heat exchanger is used. In this case only the lifetime factor is considered.

**Separation unit**

The dimensions of clarifier are obtained from Table 7. The construction material of the clarifier consists of concrete. The thickness of the clarifier wall was assumed to be 0.4 m (Nosive Strukture). The amount of concrete needed was calculated according to equation 37.

\[ V_{\text{concrete}} = \pi \cdot [(r_i + t_{\text{conce}})^2(h + t_{\text{conce}}) - r_i^2h] \]  

(37)

The centrifugal filter was assumed to be in shape of a cylinder having a diameter of 0.8 m, a height of 1 meter (Kirchofer et al. 2012) and wall thickness of 5 mm and constructed of the same material as the slurry reactor. The calculations were carried out in the same manner as for the slurry reactor and the mixer. The following results presented in Table 13 were obtained:

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>(V_{\text{concrete}}, [\text{m}^3])</td>
<td>30.92</td>
</tr>
<tr>
<td>(m_{\text{ct}}, [\text{m}^3])</td>
<td>71.05</td>
</tr>
<tr>
<td>(V_{\text{ctf}}, [\text{m}^3])</td>
<td>54.98</td>
</tr>
<tr>
<td>(p_{\text{ct}}, [\text{p/MWh}_\text{el}])</td>
<td>(1.89 \cdot 10^{-7})</td>
</tr>
<tr>
<td>(p_{\text{cf}}, [\text{p/MWh}_\text{el}])</td>
<td>(8 \cdot 10^{-7})</td>
</tr>
</tbody>
</table>
4.4 SimaPro

Having obtained the information about material and energy requirements, the next step is to build the processes in SimaPro. Each of the units was entered in SimaPro separately and independent of each other. Also, productions of the equipment were entered in SimaPro as separate processes and later as input into each process unit. Finally, to simulate the whole carbonation process, an assembly of the units was gathered to evaluate the life cycle of each carbonation route. In the proceeding text, the assumptions, modifications and calculations performed before simulating the processes in SimaPro are presented.

Compression

A process for compressing was found in SimaPro, which consisted of input of a compressor, maintenance of the compressor and electricity consumption. The output was expressed in terms of 1 m$^3$ of air to be compressed to 10 bar. The process was adjusted to match the compression step considered in this study by changing the energy required for compressing 1 m$^3$ of CO$_2$. Therefore, it was required to convert the mass flow of the CO$_2$ to volume flow and then obtain the energy per volume of CO$_2$.

Assuming CO$_2$ behaves as an ideal gas and that the flue gas is at atmospheric pressure and 363 K, the volume of CO$_2$ is obtained as follows:

\[ \nu_{STP} = \frac{RT_s}{P_s} = 22.4 \text{ l/mol} \]  
(38)

\[ \frac{\nu_{CO_2}}{\nu_{STP}} = \frac{RT_{FG}}{P_{FG}} \frac{R}{P_s} \frac{T_s}{T_s} = \frac{T_{FG}}{T_s} \]  
(39)

\[ \nu_{CO_2} = \nu_{STP} \frac{T_{FG}}{T_s} \]  
(40)

\[ \nu_{CO_2} = 22.41 \text{ l/mol} \cdot \frac{363 \text{ K}}{273 \text{ K}} = 29.78 \text{ l/mol} \]  
(41)

\[ \nu_{CO_2} = \frac{\nu_{CO_2}}{n_{CO_2}} \]  
(42)

\[ V_{CO_2} = \frac{m_{CO_2} \nu_{CO_2}}{M_{CO_2}} \]  
(43)

Grinding

It was found that SimaPro contains a process for crushing 1 kg limestone, which with some simple adjustment could be used for simulating the grinding step. This process included the infrastructure, feed of limestone and water, energy requirement and emission of particulate to air. The infrastructure considered was heavy machinery and conveyor belt. The particulate emissions and use of heavy machinery were kept constant while making the following adjustments:

- Changing energy consumption based on own calculations
- Removing input of limestone
- Recalculating the length of the conveyor belt needed to transport 1 kg of slag:
The input of conveyor belt is expressed as the length of the conveyor belt occupied by 1 kg of limestone. Assuming the width of the belt is constant as well as the height of the material piled up on the belt, the occupied length could be obtained based on density of the slag and limestone. The following calculations explain the theory behind it.

\[ V = l \cdot w \cdot h \]  
(44)

\[ \rho = \frac{m}{V} \]  
(45)

Substituting equation 45 in 44 and solving for length gives:

\[ l = \frac{m}{h \cdot w \cdot \rho} \]  
(46)

\[ l_l = \frac{m_l}{h_l \cdot w \cdot \rho_l} \]  
(47)

\[ l_s = \frac{m_s}{h_s \cdot w \cdot \rho_s} \]  
(48)

Taking the ratio between length taken up by slag and the limestone gives:

\[ l_s = \frac{l_l \cdot \rho_l}{\rho_s} \]  
(49)

**Mixing**

The mixing was programmed in SimaPro consisting of the following inputs:
- Total liquid requirement for slurry route carbonation in terms of volume [m³].
- Mixer: in terms of portion [p].
- Electricity: consumption in terms of energy [kJ].
- Conveyor belt: for transporting the slag from grinding site to mixing. The same calculations as the grinding are applicable here.

**Pumping**

For simulating the pumping in SimaPro, two main inputs were considered: input of a pump and energy requirement. SimaPro already contains a process with 1 unit of 40 kW pump as output. Pumping was simply programmed by putting the total pumping energy requirement and production of the pump as inputs.

**Heating**

As mentioned earlier, for the slurry route it was assumed that no external heating is required. This led to the simplification of only considering the material requirement for construction of the heat exchanger. However, it was necessary to consider the pressure drop within the heat exchanger and the associate energy requirements to overcome this pressure drop (Bidabadi et al., 2013).
\[ \Delta P = \frac{1.45 L u^2 \rho_d}{1.75 \cdot 10^3} \]  
(50)

\[ u = \frac{v_{sl}}{t_{MWH} A_c} \]  
(51)

\[ P_{hx} = \frac{1}{\eta_p} \left( \frac{m_h}{\rho_h} \Delta P_h + \frac{m_c}{\rho_c} \Delta P_c \right) \]  
(52)

\[ E_{hx} = P_{hx} \cdot t_{MWH} = 23.75 \text{ kJ} \]  
(53)

It should be mentioned that the volume considered here is the average volume of the hot and cold stream and it is assumed that both streams have the same velocity. This assumption was made due to insignificant difference of volumetric flow between the two streams.

**Carbonation**

As for the slurry route only the electricity required for agitation and the portion of the reactor used was fed into the process. In the case of the wet route, in addition to energy requirement for rotation and heating and the input of the reactor, the water requirement was also added to the list of inputs. Heating was assumed to be provided by electricity delivering the heat with 95% efficiency. Hence, the electricity requirement for heating needed to be recalculated. The new heating requirement was calculated to 471.65 MJ.

**Solid/liquid separation**

For both the clarifier and centrifugal filter the inputs consisted of input of the equipments and energy consumption for separation. To transport the thickened mixture of slag and water a conveyor belt and excavator were added to both separation steps with the length calculated through the equations presented for grinding site. The input of the excavator is expressed in terms of volume of the material to load. The data for outputs from the separation are summarized in Table 14.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Separation 1</th>
<th>Separation 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>( m_{csl} ) [kg]</td>
<td>5353.2</td>
<td>2858.3</td>
</tr>
<tr>
<td>( x_{H_2O} ) [kg ( H_2O )/kg csl]</td>
<td>51.9</td>
<td>10.0</td>
</tr>
<tr>
<td>( x_{cs} ) [kg cs/kg csl]</td>
<td>48.1</td>
<td>90.0</td>
</tr>
<tr>
<td>( \rho_{csl} ) [kg csl/m(^3) csl]</td>
<td>1056.6</td>
<td>2960.2</td>
</tr>
<tr>
<td>( V_{H_2O} ) (separated) [m(^3)]</td>
<td>8.3</td>
<td>2.5</td>
</tr>
</tbody>
</table>

**Wastewater treatment**

The separated water in the slurry route, corresponding to 10.84 m\(^3\), was assumed to be sent to a wastewater treatment. An existing process in SimaPro representing treatment of waste water from pig iron production was assumed to be compatible with treating of waste water from BOF carbonation, since they have the same origin. The only adjustment made was to remove the input of construction of the treatment plant since it is considered that a treatment plant already exists and is not to be built solely intended for the carbonation process.
Transportation
Transportation of steel slag to the carbonation plant and the carbonated product to the landfill site were considered. A transportation distance of 20 km was considered to be reasonable for each transportation step. Hence, transportation was assumed to be by trucks (Kirchofer et al., 2012) and only fuel consumption and maintenance requirements were considered as inputs. This process was considered to be identical for both the slurry and the wet routes, with the mass transported being the only difference. Transportation is expressed in ton kilometric defined as the product of the mass of the material, in this case mass of slag, with the transportation distance. The results are as follows:
- Transport to grinding, slurry: 43.8 tkm
- Transport to landfill, slurry: 57.2 tkm
- Transport to grinding, wet: 109.6 tkm
- Transport to landfill, wet: 117.6 tkm

Landfill
The carbonated products were assumed to be landfilled in all scenarios. In the basic scenarios it was assumed that all of the carbonated products were landfilled. The BOF residues used as reactant were entered as avoided product. SimaPro subtracts the contribution of the avoided product from the total contribution. The reason for entering the BOF flow into the carbonation plant as avoided product is that these residues are considered as waste/byproducts produced by the steelmaking plant. Hence, instead of disposing this waste, it is used to produce a useful product meaning that the environmental impacts of disposal are eliminated.

\[ m_{\text{slag}}(\text{landfilled}) = m_{\text{cs}} - m_{s} \]  \hspace{1cm} (54)

The term \( m_{s} \) refers to avoided product or in other words input of un-reacted slag. For the scenario Slurry1 the total slag being landfilled is:

\[ m_{\text{slag}}(\text{landfilled}) = 2572.5 \text{ kg} - 2191.3 \text{ kg} = 381.2 \text{ kg} \]  \hspace{1cm} (55)

While for the scenario Wet1 the total amount corresponds to:

\[ m_{\text{slag}}(\text{landfilled}) = 5878.9 \text{ kg} - 5478.3 \text{ kg} = 400.7 \text{ kg} \]  \hspace{1cm} (56)

These calculations show that the difference in the total amount of slag being landfilled is quite small even though the amounts of slag required for the carbonation processes differ much.

In the improved cases, it was assumed that only half of the product is being landfilled while the other half substitutes limestone and natural aggregates in the Slurry2 and Wet2 scenarios respectively. In these cases, in addition to input of un-reacted slag as avoided product, the production of limestone (washed and crushed) and aggregates were also inserted as avoided product. That is:

\[ m_{\text{slag}}(\text{landfilled}) = \frac{m_{\text{ag}}}{2} - m_{s} \]  \hspace{1cm} (57)
The results are presented in Table 15.

<table>
<thead>
<tr>
<th>Avoided products</th>
<th>Slurry2</th>
<th>Wet2</th>
</tr>
</thead>
<tbody>
<tr>
<td>$m_{\text{slag(landfilled)}}$ [kg]</td>
<td>-905.1</td>
<td>-2538.8</td>
</tr>
<tr>
<td>$m_{\text{substitute}}$ [kg]</td>
<td>1286.2</td>
<td>2939.5</td>
</tr>
</tbody>
</table>
5 Results and discussion

5.1 Energy and material requirements
The energy calculated in sections 4.1-4.3 are summarized and presented in Table 16 and Table 17, on descending order, for the slurry and wet routes respectively.

Table 16. Energy requirements of different units in the slurry route

<table>
<thead>
<tr>
<th>Slurry</th>
<th>Energy [kJ]</th>
<th>Percentage</th>
</tr>
</thead>
<tbody>
<tr>
<td>Compression</td>
<td>1.59E+05</td>
<td>57.4</td>
</tr>
<tr>
<td>Grinding</td>
<td>6.31E+04</td>
<td>22.8</td>
</tr>
<tr>
<td>Reaction</td>
<td>2.23E+04</td>
<td>8.04</td>
</tr>
<tr>
<td>Pumping</td>
<td>1.84E+04</td>
<td>6.63</td>
</tr>
<tr>
<td>Separation2</td>
<td>1.03E+04</td>
<td>3.72</td>
</tr>
<tr>
<td>Mixing</td>
<td>2.23E+03</td>
<td>0.80</td>
</tr>
<tr>
<td>Wastewater</td>
<td>1.43E+03</td>
<td>0.52</td>
</tr>
<tr>
<td>Separation1</td>
<td>9.54E+01</td>
<td>3.45E-02</td>
</tr>
<tr>
<td>Heat exchange</td>
<td>2.38E+01</td>
<td>8.58E-03</td>
</tr>
<tr>
<td>Sum</td>
<td>2.77E+05</td>
<td>100</td>
</tr>
</tbody>
</table>

Table 17. Energy requirements of different units of wet route

<table>
<thead>
<tr>
<th>Wet</th>
<th>Energy [kJ]</th>
<th>Percentage</th>
</tr>
</thead>
<tbody>
<tr>
<td>Carbonation (heating)</td>
<td>4.72E+05</td>
<td>59.8</td>
</tr>
<tr>
<td>Compression</td>
<td>1.59E+05</td>
<td>20.1</td>
</tr>
<tr>
<td>Grinding</td>
<td>1.26E+05</td>
<td>16.0</td>
</tr>
<tr>
<td>Carbonation (rotation)</td>
<td>3.20E+04</td>
<td>40.5</td>
</tr>
<tr>
<td>Sum</td>
<td>7.89E+05</td>
<td>100</td>
</tr>
</tbody>
</table>

The tables show that the wet route requires 2.8 times higher energy input than the slurry route despite the fewer number of units. This is mainly due to high energy requirement for heating the slag in the rotary kiln reactor. On the other hand, the compression is the main energy consumer in the slurry route. Eliminating the energy requirement for compression, since this unit is identical for all cases, it can be seen that the main energy consumers are the heating requirements and grinding of the slag in the wet route cases, while grinding, agitation in carbonation reactor, pumping and centrifugal filtration are the main consumers in slurry route carbonation. It should be mentioned that the energy requirement for centrifugal filtration is based on the assumption that the highest value from the range of the energy consumptions of centrifugal filters was chosen. However, if the lowest value within the range was assumed this step would still fall between pumping and mixing in Table 16.

The material requirements were also calculated based on the equations and assumptions described in sections 4.1-4.3. The results are presented in Table 18 and Table 19 for the slurry and the wet route, respectively.
Table 18. Material requirement for carbonation and construction in the slurry route

<table>
<thead>
<tr>
<th>Material</th>
<th>Amount</th>
</tr>
</thead>
<tbody>
<tr>
<td>Steel [kg/MWh]</td>
<td>1.87E-03</td>
</tr>
<tr>
<td>Water [l/MWh]</td>
<td>1.14E+04</td>
</tr>
<tr>
<td>Glass wool [kg/MWh]</td>
<td>7.07E-06</td>
</tr>
<tr>
<td>Concrete [m3/MWh]</td>
<td>5.83E-06</td>
</tr>
<tr>
<td>Slag [kg/MWh]</td>
<td>2.19E+03</td>
</tr>
</tbody>
</table>

Table 19. Material requirement for carbonation and construction in the wet route

<table>
<thead>
<tr>
<th>Material</th>
<th>Amount</th>
</tr>
</thead>
<tbody>
<tr>
<td>Steel [kg/MWh]</td>
<td>9.90E-03</td>
</tr>
<tr>
<td>Water [l/MWh]</td>
<td>2.67E+03</td>
</tr>
<tr>
<td>Glass wool [kg/MWh]</td>
<td>1.69E-05</td>
</tr>
<tr>
<td>Copper [kg/MWh]</td>
<td>5.4E-03</td>
</tr>
<tr>
<td>Slag [kg/MWh]</td>
<td>5.48E+03</td>
</tr>
</tbody>
</table>

As can be seen the wet route carbonation requires higher slag, steel and glass wool, on the other hand, the water requirement is 4.3 times higher for slurry route.

5.2 LCA results

5.2.1 Global warming potential and CO₂ emissions

The additional global warming potential of the different carbonation scenarios are presented in Figure 3 and Figure 4. The only difference between the basic scenarios (scenario 1) and their respective improved scenarios (scenario 2) is the landfilling step. As can be seen landfill of slag does not have any effect on GWP. Substituting limestone with carbonated product from the slurry route leads to saving of the GWP that would have been caused by producing that amount of limestone. The same applies for the wet route carbonation and substituting aggregates with the carbonation product.

Substitution leads to 3.35 GWP and 14.2 GWP reduction for the slurry and the wet route respectively. The difference in GWP reduction between slurry and wet carbonation is partially due to higher amount of slag processed in the wet route, hence higher amount of aggregates are substituted. For this reason the impacts of producing 1 kg of aggregates and limestone were compared. It was observed that aggregate production leads to higher impact in GWP category. This means that higher environmental benefits are achieved from saving 1 kg of aggregates than saving 1 kg of limestone.

Considering the slurry routes the main contributor to the GWP is compression of CO₂, followed by mixing, grinding and transportation. Carbonation and pumping have minor contribution while the contributions of the rest of the steps are insignificant. It would be expected that the contribution of the units to the GWP would follow the order of energy
consumption as shown in Table 16. This is true for compression and grinding. However, looking closely into the mixing process, it was shown that the high water requirement is the main reason and the contribution of electricity for mixing the slurry is negligible in comparison. This is probably due to high energy consumption in the processes upstream of the water production. As for transportation, fuel combustion is the main contributor to GWP.

In the case of the wet route, the order of GWP contributor is related to the energy requirements of the units. As the energy requirement increases the GWP also increases. Figure 4 shows that the main contributor is the carbonation step (due to high heating requirement) followed by compression, grinding and transportation. Electricity use is the main reason for GWP of all these steps except for transportation where fuel combustion is the contributor. The wet carbonation scenarios are shown to have doubled the impact on global warming than the slurry routes which, as explained earlier, is due to the difference in energy requirements.

![GWP of Slurry Carbonation](image)

*Figure 3. Contribution of slurry route to global warming potential*
Including the sequestered CO$_2$, the total avoided CO$_2$ emission and GWP were obtained and presented in Table 20. The efficiencies presented in Table 20 are obtained by taking the ratio between the total amount of CO$_2$ avoided and the inflow of CO$_2$ to the carbonation processes. According to the values presented, BOF slag carbonation can lead to an overall negative contribution to GWP, meaning CO$_2$ emissions are actually avoided. However, there is a difference in amount of CO$_2$ being avoided by the different routes due to the difference in energy consumptions, as discussed earlier. The slurry cases show a higher capture potential than the wet cases, seen from the bigger negative values of the slurry cases. Also, the efficiencies of the slurry scenarios are higher than the wet scenarios. The basic scenarios differ in 83 kg more CO$_2$ avoided/MWh$_{el}$ through slurry carbonation, while for the improved cases the difference is 73 kg CO$_2$/MWh$_{el}$. The difference is smaller between the improved scenarios due to the higher improvement achieved through substitution of aggregates.

Table 20. CO$_2$ reduction potential

<table>
<thead>
<tr>
<th>Case</th>
<th>CO$<em>2$ input [kg/MWh$</em>{el}$]</th>
<th>Total GWP [kg CO$<em>2$ eq/MWh$</em>{el}$]</th>
<th>Avoided CO$<em>2$ [kg/MWh$</em>{el}$]</th>
<th>Efficiency [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Slurry1</td>
<td>547.82</td>
<td>-472.91</td>
<td>-478.78</td>
<td>87.40</td>
</tr>
<tr>
<td>Slurry2</td>
<td>547.82</td>
<td>-476.26</td>
<td>-481.73</td>
<td>87.94</td>
</tr>
<tr>
<td>Wet1</td>
<td>547.82</td>
<td>-383.52</td>
<td>-396.08</td>
<td>72.30</td>
</tr>
<tr>
<td>Wet2</td>
<td>547.82</td>
<td>-397.69</td>
<td>-408.51</td>
<td>74.57</td>
</tr>
</tbody>
</table>
5.2.2 Impact assessment

5.2.2.1 Abiotic depletion
The contribution of the two basic carbonation routes, i.e. Slurry1 and Wet1, are presented in Figure 5. Abiotic depletion is connected to extraction of raw material such as water, metal and minerals.

In the slurry case, the highest impact is caused by mixing, due to the water requirement for carbonation reaction. The same applies for the wet route where water consumption in the carbonation step is the main contributor followed by material required for reactor construction.

The second contributor in both cases is caused by transportation. It is observed that the contributor of transportation in the wet route is more than double as high as the slurry route. The difference is explained by the difference in masses being transported; hence, higher material for maintenance of the trucks is needed. The same applies for grinding which is the third and forth contributor in wet and slurry route respectively.

Overall, the slurry route leads to higher contribution in abiotic depletion impact category, which as explained before is due to the high water consumption.

Figure 5. Contribution of slurry and wet route to abiotic depletion
5.2.2.2 *Abiotic depletion (fossil fuel)*

This category is directly connected to extraction of fossil fuels. This is confirmed by comparing the energy requirements presented in Table 16 and Table 17, with the graphs obtained in Figure 6. According to Table 16 and Figure 6 the highest energy consumers in the slurry route in descending order are: compression, grinding, mixing, transportation, carbonation and pumping. The energy requirement for de-ionised water production adds to the contribution of mixing, explaining the deviation between Figure 6 and Table 16. For the wet route the order of energy consumers from high to low is as follows: carbonation, compression, grinding and finally transportation. Overall, the main contributor in the wet route is electricity input for heating of slag.

The difference between the slurry and the wet route regarding grinding and transportation is, as mentioned before, due to the higher amount of slag that is processed in the wet route carbonation. It is clear that overall the wet route has much higher impact than the slurry route; the carbonation step alone can make up for the total contribution from the slurry route. The impact of the wet route is 2.2 times higher than the slurry route which is consistent with the values obtained in Table 17.

![Abiotic Depletion Fossil Fuel (ADF)](image_url)

*Figure 6. Impact on abiotic depletion in terms of fossil fuel*
5.2.2.3 Acidification and photochemical oxidation

Emissions of NO\textsubscript{x} and SO\textsubscript{x} to the air are the cause of acidification and are results of combustion of fossil fuels such as natural gas, oil and hard coal to gain energy. Hence, the graphs in Figure 7 should follow the same pattern as the graphs representing contribution to ADF shown in Figure 6, which is the case here. This is also observed from Table 21. These data are obtained from inventory analysis, where main emissions to air, water and soil and their main emitters were determined.

One difference is observed between the graphs in Figure 6 and Figure 7 for the slurry case. Mixing is shown to result in a lower contribution than the compressor in Figure 6, however, Figure 7 shows that mixing leads to almost the same contribution as compression. This difference is due to emission of chloride from water production to water, which increases the acidification potential of the mixing step.

![Figure 7. Effect on acidification](image)

<table>
<thead>
<tr>
<th>Substance</th>
<th>Total emission (Slurry1)</th>
<th>Emitters (Slurry1)</th>
<th>Total emission (Wet1)</th>
<th>Emitters (Wet1)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CO [g]</td>
<td>56.93</td>
<td>C, M, G</td>
<td>101.63</td>
<td>R, C, G</td>
</tr>
<tr>
<td>NO\textsubscript{x} [g]</td>
<td>171.29</td>
<td>C, T, M, G</td>
<td>349.39</td>
<td>R, T, C, G</td>
</tr>
<tr>
<td>NMVOC [g]</td>
<td>27.22</td>
<td>C, T, M, G</td>
<td>60.77</td>
<td>R, T, C</td>
</tr>
<tr>
<td>SO\textsubscript{x} [g]</td>
<td>210.92</td>
<td>C, M, G</td>
<td>399.28</td>
<td>R, C, G</td>
</tr>
</tbody>
</table>
The same substances, i.e. SO\textsubscript{x} and NO\textsubscript{x}, which lead to acidification, also cause photochemical oxidation. In addition to these substances NMVOC and CO also contribute to formation of ground level ozone. These are as mentioned previously related to emissions from combustion of fossil fuels. Hence, same observations as ADF are valid. The values presented in Table 21 on emission of the above mention compounds confirm these observations (See Figure 8 and Table 21).

**Figure 8. Contribution to photochemical oxidation**

5.2.2.4 **Ozone layer depletion**
As for the slurry route the mixing is the main contributor to ozone layer depletion, which is connected to upstream processes of hydrochloric acid production for de-ionising water through chlor-alkali electrolysis. An additional contribution comes from pipeline transportation of natural gas for electricity production. This is connected to the part of country mix electricity produced from natural gas. Therefore, with exception of mixing, the order of contributors follows the energy consumption for both the slurry and wet cases. There is of course a contribution from input of de-ionised water in wet carbonation, as for the mixing. However, it is not significant in comparison to contribution from high energy input. Landfill does not have any impact on ozone layer depletion as is observed from Slurry1 and Wet1 cases (See Figure 9).
5.2.2.5 Human and ecotoxicity

Figure 10 presents and compares the impact of the slurry- and wet route on human toxicity. The emission of chromium containing compound from processes upstream of metal alloys production, steel for construction in this case, is the primary contribution to this category. This is followed by the spoil from hard coal mining which is related to electricity production. For the slurry route the de-inosied water consumption is the main contributor to the overall impact on human toxicity. There is a noticeable contribution from landfill due to leaching of heavy metals from the carbonated slag. The contributions of landfill for Slurry1 and Wet1 are almost the same since the overall mass of product being landfill are approximately the same. Contribution of landfill is also observed from inventory analysis.

Table 22 summarizes the main emissions to water and shows that emissions of metals are mainly caused from landfill of slag. It should be mentioned that emissions due to leaching are based on data for landfill of un-reacted BOF slag. According to literature carbonation improves leaching behavior of steelmaking slag hence, in the future studies the emissions due to landfill should be adjusted with experimental values obtained on leaching behavior of CS.
Table 22. Main emissions to water along with their main contributors

<table>
<thead>
<tr>
<th>Substance</th>
<th>Total (Slurry1)</th>
<th>Source (Slurry1)</th>
<th>Total (Wet1)</th>
<th>Source (Wet1)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aluminium [kg]</td>
<td>1.55</td>
<td>L</td>
<td>1.61</td>
<td>L</td>
</tr>
<tr>
<td>Calcium [kg]</td>
<td>13.41</td>
<td>L</td>
<td>14.19</td>
<td>L</td>
</tr>
<tr>
<td>Chloride [kg]</td>
<td>2.98</td>
<td>M</td>
<td>1.08</td>
<td>R, C</td>
</tr>
<tr>
<td>Chromium VI [g]</td>
<td>380.92</td>
<td>L</td>
<td>400.46</td>
<td>L</td>
</tr>
<tr>
<td>Magnesium [kg]</td>
<td>1.10</td>
<td>L, M</td>
<td>1.21</td>
<td>L, R</td>
</tr>
<tr>
<td>Silicon [kg]</td>
<td>32.11</td>
<td>L, M</td>
<td>33.81</td>
<td>L, R</td>
</tr>
<tr>
<td>Sodium [g]</td>
<td>969.88</td>
<td>M, C</td>
<td>463.10</td>
<td>R, C</td>
</tr>
<tr>
<td>Sulfate [kg]</td>
<td>1.07</td>
<td>M, C, G</td>
<td>1.56</td>
<td>R, C, G</td>
</tr>
</tbody>
</table>

Figure 10. Impact on human toxicity

The impacts on fresh water aqueous ecotoxicity, marine aqueous ecotoxicity and terrestrial ecotoxicity were added together to simplify the evaluation. The results are presented in Figure 11. It should be mentioned that the impacts were significantly higher in marine aqueous ecotoxicity in comparison to the other two categories. However, looking through the processes contributing to these categories resulted in similar observations. In summary, mining of hard coal for electricity production is the main contributor. Mining of hard coal leads to acid mine drainage which, in addition to acidification, contaminates ground water with heavy metals. This means processes with high energy requirement lead to high impact in
both cases. Landfill of carbonated product leads to a small contribution due to leaching of the heavy metals presented in the raw material (BOF slag) (See Table 22).

![Ecotoxicity (ET)](chart.png)

**Figure 11. Impact on ecotoxicity**

### 5.2.2.6 Eutrophication

Excess of phosphorus and nitrogen compounds causes overgrowth of plants. Anthropogenic activities resulting in emission of these substances include usage of fertilizer or combustion of fossil fuels. In this case combustion of fossil fuel with purpose of energy production is the main cause. Hence, the contribution of steps follow the same order as presented in Table 16 and Table 17. In addition, mining of coal for electricity production is one of the primary reasons for effects on eutrophication as presented in Figure 12.
5.2.3 Improved scenarios

Since the improved scenarios differ from the basic cases in only landfill, the results obtained from landfill are presented separately. Table 23 and Table 24 present the results for the slurry and the wet cases, respectively. In each table the contribution of landfill from the basic and improved scenarios are compared. In addition, the resultant percentage change due to the improved scenarios relative to the life-cycle impact of the basic scenarios is presented.

Table 23. Impact assessment of landfill step in Slurry1 and Slurry2

<table>
<thead>
<tr>
<th>Impact category</th>
<th>Landfill (Slurry2)</th>
<th>Landfill (Slurry1)</th>
<th>Change [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>AD [kg Sb eq]</td>
<td>-4.89E-06</td>
<td>0</td>
<td>-6.23</td>
</tr>
<tr>
<td>ADF [MJ]</td>
<td>-42.56</td>
<td>0</td>
<td>-3.98</td>
</tr>
<tr>
<td>ODP [kg CFC-11 eq]</td>
<td>-1.72E-07</td>
<td>0</td>
<td>-1.58</td>
</tr>
<tr>
<td>HT [kg 1,4-DB eq]</td>
<td>-3.84</td>
<td>1.30</td>
<td>-25.08</td>
</tr>
<tr>
<td>FWAE [1,4-DB eq]</td>
<td>-25.4</td>
<td>10.55</td>
<td>-112.17</td>
</tr>
<tr>
<td>MAE [1,4-DB eq]</td>
<td>-5.81E+03</td>
<td>1309.58</td>
<td>-12.31</td>
</tr>
<tr>
<td>TE [1,4-DB eq]</td>
<td>-1.43E-03</td>
<td>8.64E-20</td>
<td>-1.73</td>
</tr>
<tr>
<td>PO [kg C2H4 eq]</td>
<td>-1.51E-03</td>
<td>0</td>
<td>-10.02</td>
</tr>
<tr>
<td>AC [kg SO2 eq]</td>
<td>-0.05</td>
<td>0</td>
<td>-13.93</td>
</tr>
<tr>
<td>EU [kg PO4^3- eq]</td>
<td>-0.01</td>
<td>0</td>
<td>-16.82</td>
</tr>
</tbody>
</table>
Table 24. Comparing impact assessment of landfill in Wet1 and Wet2

<table>
<thead>
<tr>
<th>Impact category</th>
<th>Landfill (Wet2)</th>
<th>Landfill (Wet1)</th>
<th>Change [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>AD [kg Sb eq]</td>
<td>-2.52E-06</td>
<td>0</td>
<td>-4.06</td>
</tr>
<tr>
<td>ADF [MJ]</td>
<td>-177.20</td>
<td>0</td>
<td>-7.48</td>
</tr>
<tr>
<td>ODP [kg CFC-11 eq]</td>
<td>-3.10E-07</td>
<td>0</td>
<td>-1.75</td>
</tr>
<tr>
<td>HT [kg 1,4-DB eq]</td>
<td>-10.76</td>
<td>1.37</td>
<td>-51.42</td>
</tr>
<tr>
<td>FWAE [1,4-DB eq]</td>
<td>-71.33</td>
<td>11.09</td>
<td>-257.78</td>
</tr>
<tr>
<td>MAE [1,4-DB eq]</td>
<td>-1.90E+04</td>
<td>1376.69</td>
<td>-26.96</td>
</tr>
<tr>
<td>TE [1,4-DB eq]</td>
<td>-6.95E-03</td>
<td>9.08E-20</td>
<td>-3.90</td>
</tr>
<tr>
<td>PO [kg C₂H₄ eq]</td>
<td>-6.18E-03</td>
<td>0</td>
<td>-20.66</td>
</tr>
<tr>
<td>AC [kg SO₂ eq]</td>
<td>-0.12</td>
<td>0</td>
<td>-18.66</td>
</tr>
<tr>
<td>EU [kg PO₄³⁻eq]</td>
<td>-0.02</td>
<td>0</td>
<td>-13.17</td>
</tr>
</tbody>
</table>

As was seen from analyzing the impact assessment of Slurry1 and Wet1 scenarios, landfill of raw slag and carbonated slag does not have any effect on any of the impact categories except HT, FWAE, MAE and TE. This was shown to be mainly due to leaching of metals into ground water and acid drainage caused by mining of hard coal for energy production. Substitution of limestone and natural aggregates with the carbonated products result in a slight decrease in categories AD, ADF, ODP, PO, AC and EU. This reduction is thanks to eliminating energy requirement involved in the lifecycle of limestone and aggregate production. The reduction in the remaining categories, in addition to benefits gained from substitution, is mostly obtained from avoiding landfill of raw BOF slag. The benefits are higher for the wet route than slurry route partially due to landfill of bigger quantity of slag being avoided in the wet route, and partially due to production of aggregates having higher contribution to environmental impacts than production of limestone. Even though there is a decrease in the contributions, the decrease is not enough to cancel out the total lifecycle impacts in any of the categories except FWAE.

5.3 Sensitivity analysis

5.3.1 Lifetime of the carbonation plant

It was assumed that the lifetime of the equipment necessary for building the carbonation plant were 25 years. Due to uncertainty about this assumption and the calculations performed for designing the equipments a sensitivity analysis was performed based on the lifetime of the equipment. The lifetime analyzed consist of 25-5 years with 5 years of difference. The effect of lifetime on global warming potential is presented in Figure 13. It is clear that the lifetime has no significant effect on the contribution of the carbonation process to GWP. This was however, expected since the results in the previous section showed that the electricity has the highest impact in all impact categories while effect of construction was insignificant.
However, comparing the effect on the other categories showed different results. Figure 14 and Figure 15 present the percentage change in contribution to impact categories relative to the base year (25 year) for Slurry1 and Wet1, respectively. Both figures show that as the material requirement for construction of equipment increases (due to the shorter lifetime), the contribution to the categories AD, HT and ET increases. This is due to increased extraction of raw material leading to more impacts on AD, while the emission of chromium compounds from processing of the material for production of steel leads to increased impact on HT and ET. Other impact categories do not show any dependence on the lifetime.
Figure 14. Graphs presenting relation between lifetime and impact assessment of slurry route

Figure 15. Graphs presenting relation between lifetime and impact assessment of wet route
Table 25 and Table 26 compare the quantified contribution of the slurry and the wet route with respect to the above mentioned impact categories. The wet route, as observed earlier, has higher impact in all categories except in AD regardless of different lifetimes. The slurry route has higher impact on AD than the wet route for all 5 cases of different lifetimes, which proves that the impact from input of water overcomes the impact of construction. This is in consistent with the results presented previously.

Table 25. Comparing dependence of impact categories on lifetime (slurry)

<table>
<thead>
<tr>
<th>Impact category</th>
<th>25 year</th>
<th>20 year</th>
<th>15 year</th>
<th>10 year</th>
<th>5 year</th>
</tr>
</thead>
<tbody>
<tr>
<td>AD [kg Sb eq]</td>
<td>7.84E-05</td>
<td>8.02E-05</td>
<td>8.32E-05</td>
<td>8.92E-05</td>
<td>1.07E-04</td>
</tr>
<tr>
<td>HT [kg 1,4-DB eq]</td>
<td>1.53E+01</td>
<td>1.58E+01</td>
<td>1.67E+01</td>
<td>1.84E+01</td>
<td>2.35E+01</td>
</tr>
<tr>
<td>ET [kg 1,4-DB eq]</td>
<td>4.72E+04</td>
<td>4.75E+04</td>
<td>4.80E+04</td>
<td>4.91E+04</td>
<td>5.23E+04</td>
</tr>
</tbody>
</table>

Table 26. Comparing dependence of impact categories on lifetime (wet)

<table>
<thead>
<tr>
<th>Impact category</th>
<th>25 year</th>
<th>20 year</th>
<th>15 year</th>
<th>10 year</th>
<th>5 year</th>
</tr>
</thead>
<tbody>
<tr>
<td>AD [kg Sb eq]</td>
<td>6.21E-05</td>
<td>6.34E-05</td>
<td>6.55E-05</td>
<td>6.97E-05</td>
<td>8.24E-05</td>
</tr>
<tr>
<td>HT [kg 1,4-DB eq]</td>
<td>2.09E+01</td>
<td>2.13E+01</td>
<td>2.19E+01</td>
<td>2.31E+01</td>
<td>2.67E+01</td>
</tr>
<tr>
<td>ET [kg 1,4-DB eq]</td>
<td>7.06E+04</td>
<td>7.08E+04</td>
<td>7.10E+04</td>
<td>7.16E+04</td>
<td>7.33E+04</td>
</tr>
</tbody>
</table>

5.3.2 Transportation distance

In the previous section it was observed that transportation also has a noticeable contribution to the environmental impact. Therefore, a sensitivity analysis was performed changing the transportation distance in order to examine its importance. Distances of 10, 20, 40, 60, 80 and 100 were analyzed. It should be mentioned that these distances were applied for each transportation step, transport to grinding and transport to landfill. That is the total distance is twice as the ones given above.

![Figure 16. Sensitivity analysis performed based on different transportation distance](image-url)
The results presented in Figure 16 show that the GWP of the carbonation process is dependent on the transportation distance. However, the wet route is more sensitive to changes in distance due to higher mass being transported. There is a bigger difference between the two wet scenarios than between the two slurry scenarios, which as mentioned earlier is partially due to higher environmental benefits of substituting gravel than limestone and partially due to higher mass of slag being processed. The impacts of Slurry1 and Wet1 on other categories are presented in Figure 17 and Figure 18 respectively.

![Sensitivity Analysis (Transport Distance) Slurry1](image)

**Figure 17.** Effect of transportation distance on contribution to other impact categories (Slurry1)

Examining the data for the slurry route (Figure 17) shows that all impact categories are dependent on transportation distance. However, the impact categories ADF, AD, EU and AC are more sensitive to the change in transportation distance than the other four categories. This observation is in consistency with the observation obtained for the basic cases which showed that transportation gave noticeable and slightly high contribution to AD, ADF, AC and EU while its contribution to the other categories was insignificant. The data also show that the dependence is especially high when the distance is longer than 60 km.
The same observations are valid for wet route shown in Figure 18. However; AD seems to be the most sensitive towards the changes in transportation distance. When the distance is changed from 20 to 200 km the contribution increases with 2.4 times. Overall the changes in both the slurry and wet scenarios are due to increase in fuel consumption and thereby increased emissions as the distance increases. The wet route results of course in higher contribution in all cases due to higher mass being transported.
5.3.3 Difference source of electricity
Knowing that the energy requirement is the primary cause of environmental impacts, it is useful to see whether using electricity produced from different sources have any effect on the CO$_2$ capturing potential of the carbonation processes. The dependency of the GWP and energy source is presented in Figure 19.

![Figure 19. Results of sensitivity analysis comparing effect of electricity production source on net CO$_2$ reduction](image)

It is obvious that electricity produced from fossil fuels, hydropower (pumped storage) and biogas leads to emission of a higher amount of CO$_2$ leading to positive contribution to global warming potential. On the other hand the non fossil based power production actually lead to a higher negative contribution of around 530 kg CO$_2$ eq/MWh$_{el}$ which correspond to approximately 96 % of ingoing CO$_2$ being avoided. It is clear that the wet routes are more sensitive to source of energy than the slurry route which is reasonable considering that the wet route requires higher electricity input.
6 Conclusions

A comparative LCA study was performed on direct carbonation of BOF slag through slurry and wet routes. The basis for the LCA was defined as 1 MWh electricity produced by a conventional natural gas power plant. In addition, a sensitivity analysis was carried out to investigate the effect of construction, transportation distance and power source on the outcome.

The results showed that both routes are capable of storing CO\textsubscript{2} and resulting in a negative net contribution to global warming potential. However, the slurry route showed a higher reduction potential. Even though a negative GWP was observed, both routes gave a positive contribution to all the other environmental impact categories. In general, the wet route had approximately two times higher impact than the slurry route. One exception was the impact category of abiotic depletion, where the slurry route appears to have the highest impact.

The contributions to the mid-point categories were caused mainly due to energy requirements. Hence, in the case of the slurry route mixing, compression, grinding and transportation were the main contributors in descending order. On the other hand, for the wet route the order was as follows: carbonation, compression, grinding and transportation.

By performing a sensitivity analysis, it was concluded that construction of the carbonation plant does not have a significant effect on GWP, while a dependency on transportation distance and electricity source was observed. As the distance increased the GWP also increased. If electricity was fossil fuel based, the GWP would increase but still result in a total negative contribution. Since the contribution in the other impact categories were connected to electricity requirements, same trends were observed.
7 References


