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Environmental assessment of CO₂ mineralisation for sustainable construction materials



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ABSTRACT

Mineral carbonation is a carbon utilisation technology in which an alkaline material reacts with carbon dioxide forming stable carbonates that can have different further uses, for instance as construction material. The alkaline material can be a residue from industrial activities (e.g. metallurgic slags) while CO_2 can be recovered from industrial flue gasses. Mineral carbonation presents several potential environmental advantages: (i) industrial residues valorisation, (ii) CO_2 sequestration and (iii) substitution of conventional concrete based on Portland cement (PC).

However, both the carbonation and the CO_2 recovery processes require energy. To understand the trade-off between the environmental benefits and drawbacks of CO_2 recovery and mineral carbonation, this study presents a life cycle assessment (LCA) of carbonated construction blocks from mineral carbonation of stainless steel slags. The carbonated blocks are compared to traditional PC-based concrete blocks with similar properties.

The results of the LCA analysis show that the carbonated blocks present lower environmental impacts in most of the analysed impact categories. The key finding is that the carbonated blocks present a negative carbon footprint. Nonetheless, the energy required represents the main environmental hotspot. An increase in the energy efficiency of the mineral carbonation process and a CO_2 valorisation network are among the suggestions to further lower the environmental impacts of carbonated blocks production.

Finally, the LCA results can promote the development of policy recommendations to support the implementation of mineral carbonation technology. Further research should enable the use of mineral carbonation on a broader range and large volume of alkaline residues.

1. Introduction

The IPCC special report on the "impacts of global warming of $1.5 \,^{\circ}$ C" suggests that to limit global warming within $1.5 \,^{\circ}$ C, the global net anthropogenic CO₂ emission needs to decline 45% from 2010 level by 2030 (IPCC, 2018). Carbon capture and utilisation (CCU) techniques have gained attention worldwide, because they can turn CO₂ emissions into a valuable resource for new products, such as fuels and chemicals (Balucan et al., 2013; Giannoulakis et al., 2014). Therefore, CCU technologies can contribute to climate change mitigation and, at the same time, increase the economic profitability of carbon capture processes (Cuéllar-Franca and Azapagic, 2015; Styring et al., 2011). However, the current markets for these products alone cannot provide a sufficient demand, compared to the amount of CO₂ emitted globally (Cuéllar-Franca and Azapagic, 2015). Another possibility to utilised the captured CO₂ is mineral carbonation, a process which produces carbonated materials as a long-term storage for CO₂ (Di Maria et al., 2018; Kelly et al., 2011; Mo et al., 2017; Salman et al., 2013). Mineral carbonation involves the capture of CO₂ in a mineral form by its reaction with alkaline materials, to form solid carbonate products that can be used, for instance, in construction activities (Santos et al., 2013). The alkaline materials for mineral carbonation can be (i) naturally occurring rocks and minerals, or (ii) residues from industrial processes, such as steel or cement manufacturing (Hampel, 1968). The use of alkaline industrial residues for mineral carbonation is especially promising since their estimated global carbonation potential may increase to $3.3 \text{ GtCO}_2/\text{y}^{-1}$ in 2100, which could represent between the 5% and 12% of the 2100 global CO₂ emissions (Renforth, 2019). Additionally, alkaline industrial residues can be carbonated in situ in the same plant where they are produced, avoiding waste transports to external

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Fig. 1. The mineral carbonation concept using alkaline industrial residue: CO_2 -containing flue gas is generated by industrial processes, as for instance steel or power plants. The CO_2 from the flue gas is then recovered through a CO_2 -recovery process, and then utilised in the mineral carbonation plant, where alkaline industrial residues are carbonised to produce carbonated blocks that can be used by the construction industry.

treatment plants (IPCC, 2005).

Following the framework indicated by the IPCC special report on carbon dioxide and capture storage (IPCC, 2005), mineral carbonation using alkaline industrial residues can be schematised as illustrated in Fig. 1. The alkaline industrial residues used for mineral carbonation are residues containing calcium and magnesium-rich oxides and silicates.

Although mineral carbonation is seen today as one of the most promising technology offering a cost-effective climate change mitigation, several barriers are still hindering its deployment at an industrial scale, such as technical feasibility, economic viability, and the lack of legislative framework for carbon recovery (Olajire, 2013; Salman et al., 2016; Santos et al., 2013). Additionally, a sustainable environmental profile is another fundamental prerequisite for a successful large-scale deployment of new technologies. Mineral carbonation is an energy-intensive process, since the kinetics of the reaction must be accelerated by a combination of increased temperature and higher CO_2 concentration. Thanks to the enhanced reaction conditions, the carbonation rate can be 40 times higher than the rate of natural carbonation (Sanjuán et al., 2003). However, the increased reaction kinetics are at the expense of additional energy needs, which may partially offset the environmental benefits.

In this regard, several studies have been published on the environmental performances of mineral carbonation, but only a few have focused specifically on the mineral carbonation using industrial alkaline residues (some of these studies are listed in Table 1 and will be discussed later). All the previous LCA studies are based on data from literature or lab-scale developments. However, when upscaling of a process at an industrial level, the process parameters will be tailored to the utilised alkaline raw material, optimising its resources consumption (energy, water, materials) compared to a lab-scale development (Shibasaki et al., 2006). Therefore, data from real industrial applications are massively needed. Following this need, the current study presents a life-cycle-assessment (LCA) of a mineral carbonation process performed at a real industrial demonstrator, built and operated in Belgium by the company "Orbix". The process uses stainless steel slag (SSS) as alkaline media for the process. SSS has been chosen thanks to its high carbonation potential (Baciocchi et al., 2010; Capobianco et al., 2014; Salman et al., 2014; Santos et al., 2013) and to its high availability, with 0.5 Mt processed each year in Belgium (Quaghebeur et al., 2015).

LCA is a methodology that allows to estimate the environmental impacts attributable to a product or a process along its life cycle (Pennington et al., 2004; Rebitzer et al., 2004). The previous LCA studies listed in Table 1 have identified the energy used for mineral

carbonation as one the factor causing the main environmental impact of the process. Another major impact is caused by the CO₂ recovery, which is an energy intensive process that captures CO₂ from industrial waste gas (Di Maria et al., 2018; Khoo et al., 2011b; Salman et al., 2016). Although mineral carbonation does not require a 100% purity of the CO₂ gas stream, a high purity increases the kinetics of the reaction and, consequently, it lowers the time needed to complete the process. Additionally, waste gas from industrial processes can contain impurities such as SO_x or Cl₂ that will compete with the CO₂ in binding the free lime or the calcium silicates, lowering the carbonation efficiency. Therefore, although possible, studies have demonstrated that working with lower grade CO₂ can compromise the economics of the carbonation process.

All analysed LCA studies assumed that CO2 is recovered through chemical absorption by methylethanolamine (MEA), which is today the most common technique for CO₂ capturing and recovering from industrial flue gas. However, for CO2 recovery there is no a "one-size-fitsall technology", and several techniques are today available to ensure compatibility with the specific industry producing the CO₂-containing flue gas (Cuéllar-Franca and Azapagic, 2015). Each of these techniques presents technical and environmental advantages and disadvantages that should be considered when evaluating the most appropriate CO_2 source for mineral carbonation. For this reason, the current study compares the environmental performances of three different techniques for CO₂ recovery: (i) cryogenic process; (ii) membrane separation; (iii) chemical absorption through MEA. Although many other possibilities for CO2 recovery are available, the above-mentioned techniques are increasingly drawing the interest of the Belgian industry. The LCA for the considered CO₂ recovery process is based on data gathered from previous literature. The inclusion of the CO₂ recovery process into the system boundaries provides a more complete and realistic evaluation of an eventual mineral carbonation facility integrated in an industrial CO₂ valorisation network.

Finally, the LCA results of the carbonated blocks production are compared to the LCA results of the traditional PC-based concrete available in the market, to find the trade-off between environmental impacts caused by the CO_2 capturing and the carbonation process, and the benefits of the CO_2 storage and the substitution of raw materialbased product in the market. Т

International Journal of	Greenhouse Gas	Control 93	(2020)	102882
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2. Materials and methods

2.1. Mineral carbonation

When CO_2 reacts with the metal oxides (MO) contained in the alkaline material, it forms the corresponding carbonate, following the chemical reaction:

$$(M)O + CO_2 = (M)CO_3$$
 (1)

where M is the divalent metal, such as calcium or magnesium. The mineral carbonation process analysed in this study refers to an industrial demonstrator producing carbonated blocks using SSS as alkaline material. The carbonated blocks are called Carbstone[®], a patented technology developed to produce high strength building materials by treating slags with CO₂ at elevated pressure and temperature, without the addition of binders (Quaghebeur et al., 2015). Fig. 2 shows a schematic overview of the Carbstone[®] production process. It consists of three main steps: (i) pre-treatment of the slag, (ii) shaping of the building blocks and (iii) CO₂ curing of the building blocks in an autoclave.

Two variants of the carbonation process have been developed at the industrial demonstrator: a low-pressure process for the production of common building blocks (compressive strength up to 50 MPa) and a high-pressure process for the production of high-strength materials (compressive strength > 50 MPa). In this study, the low-pressure process was selected, as this process may be the first one commercialised in Belgium. The low-pressure process is based on a recipe in which the raw material mix consists of a mixture of 50% dried fine-grained stainless steel slags and 50% fine sand. The SSS used in the process is produced by the company Orbix, located in Genk, Belgium. The chemical composition of the SSS is reported in Table 2 (further details on the characterisation of the slag (including micro-elements such as Cr, Mo, Ba and others) can be found in Quaghebeur et al. (2015).

2.2. Life cycle assessment

According to the ISO 14040:2006 (ISO, 2006), a LCA is carried out in four different phases: definition of goal and scope, life cycle inventory (LCI), life cycle impact assessment (LCIA), results interpretation and conclusions.

2.2.1. Goal and scope and functional unit

The goal of the LCA presented in this study is to provide an environmental analysis of carbonated blocks produced through carbonation of SSS. Three different scenarios are compared for the production of the carbonated blocks. Each scenario uses a different CO_2 recovery process as a source of CO_2 for the mineral carbonation. The carbonated blocks are further compared to the PC-based concrete blocks presenting similar properties. The results of the environmental analysis will help to identify the environmental benefits and weaknesses of the production of SSS-based carbonated blocks. In addition, the comparison between carbonated SSS-based blocks and PC-based concrete will help to understand the potential environmental effect of the mineral carbonation technology in the construction sector when replacing traditional PC-based concrete based products.

The functional unit is a fundamental parameter in LCA, as it defines the reference to which all calculations are performed during the study. It also identifies the function that serves as base for comparison of two products. The analysed blocks are used for walls construction. In this application the key technical function is the compressive strength. Therefore, the functional unit of the study refers to the capacity of the carbonated blocks and the PC-based concrete blocks to provide a specific compressive strength. The compressive strength can be defined as the capacity of materials to withstand a load without fractures, and it is measured in MPa (N/mm²). As a reference for the proposed functional unit, the LCA compares the production of 1 m² of carbonated blocks

 Table 1

 Overview of literature LCA studies on mineral carbonation using alkaline industrial residues

		o				
Study	Goal	FU	Alkaline material	(CO ₂ capture method and data source)	(Data source for mineral carbonation)	Results
Bodénan et al. (2014)	To assess the feasibility and the environmental performances of ex situ mineral carbonation to mining waste	Production of 1 MWh of electricity with a coal power plant	Mafic/ultramafic mining waste, pyrometallurgical slag	MEA	Experimental results at lab scale	The LCA results varied greatly depending on the carbonation process studied. A commercial application for the carbonation product can reduce the global environmental impact
Chang et al. (2011)	To evaluate the environmental performances of carbonation for several steel slags	1 kg of carbonated slags	Steel slags	Non-considered	Experimental results at lab scale	Energy consumption represents the highest impact during mineral carbonation, in particular due to the heating process
Di Maria et al. (2018)	LCA of carbonation compared to traditional concrete	Compressive strength provided by 1 m^2 of the compared materials	Industrial by-product (stainless steel slag)	MEA (ecoinvent)	Experimental results at lab scale	Carbonated blocks show a lower impact compared to traditional concrete. CO ₂ production and electricity consumption in carbonation are the environmental hotspot of carbonation process.
Mattila et al. (2014)	Comparative LCA of two processes for precipitated-calcium-carbonate production: Carbonation of lime vs carbonation of steel slag	Production of 1 kg of final product	Steel slags	CO ₂ taken directly from flue gas stream	Literature sources and process simulations	Carbonation of steel slags in precipitated calcium carbonate production has several advantages, possible post-treatment steps of the products and residues should be included.
Kirchofer et al. (2013)	Comparison of the carbonation performances of different alkaline materials from industrial residues	Storage CO ₂ potential of different alkaline materials	Cement kiln dust, fly ash, steel slag	Not specified	Literature	The CO ₂ mitigation potential of carbonation depends on the carbonation potential of each alkaline material and its local availability.



Fig. 2. Schematic drawing of the Carbstone® production process on a lab scale (Quaghebeur et al., 2015).

Table 2

Average composition of the SSS used in the carbonation process (Quaghebeur et al., 2015).

Main elements	wt%
CaO MgO Fe ₂ O ₃ MnO SiO ₂	$45 \pm 0.5 \\ 9.3 \pm 2.3 \\ 1.5 \pm 0.9 \\ 1.4 \pm 0.09 \\ 31 \pm 5.3 \\ 3.0 \pm 0.1 \\ 1.4 \pm$
2 0	

with 1 m^2 of PC-based concrete blocks, with both materials able to provide an equal compressive strength of 40 MPa. The compared surfaces are made of 50 blocks, each measuring 20 cm (length), 10 cm (width) and 4 cm (thickness), for a total volume of 800 cm³ (0.008 m³).

2.2.2. System boundaries and life cycle inventory

Carbonated blocks. The flow diagram for the production of the carbonated blocks is shown in Fig. 3, that also indicates the boundaries of the analysed system for the LCA study. The SSS (0–60 mm) is used as alkaline material. No transport of the SSS to the mineral carbonation plant is considered. This assumption will not significantly affect the

results since, as demonstrated by various authors, the effect of transports of alkaline residues is rather small on the overall LCI of the carbonation process (Khoo et al., 2011a,b; Slotte, 2017).

According to the data collected at the industrial demonstrator, the carbonated blocks present a compressive strength of 43 MPa, 2.5% total inorganic carbon and a density of 2300 kg/m^3 . To produce the required volume defined by the functional unit (0.008 m^3 , with a weight of 92 kg) 44.2 kg of SSS are carbonated. The different steps for the carbonated blocks production and the data inventory are summarised in Table 3 and Fig. 4.

Before entering the carbonation process, the SSS undergoes a pretreatment for metal recovery. In the pre-treatment, some metals are recuperated from the SSS using a wet mechanical treatment. The metals are reused as a raw material for the production of stainless steel. However, data on metals recovery were not disclosed, and therefore not included in the scope of this analysis. Although the lack of data on metals recovery can be considered as a limitation of the study, the available data on energy consumption and CO_2 requirements from an industrial demonstrator can provide relevant indications on the overall environmental impacts of the mineral carbonation process.

After the pre-treatment, the SSS enters the carbonation plant. First, the fine, wet slag is dried before entering the mixing process. The sand used is the same kind of sand used as fine aggregate in PC concrete. The



Fig. 3. Analysed system and boundaries of the LCA for the carbonated blocks.

Table 3

Life cycle inventory for the carbonated blocks and the PC-based concrete blocks.

Functional unit Volume	0.08 m ³
Carbonated blocks	
Density	2300 kg/m ³
Weight (as reference of the functional unit)	92 kg
Drying	
SSS (kg)	44.2
Thermal energy	5.8 kWh
Mixing	
Electricity	2.5 kWh
Water	9.8 kg
Sand	44.2 kg
Curing	
Electricity	0.2 kWh
CO ₂	8.5 kg
Water (released)	7.9 kg
00	
CO ₂ recovery	
S1 – Cryogenic	E 0 1-34/h
	5.3 KWN
52 – memorane	0.21-34/h
Electricity	0.3 KWII
S3 – chemical absorption	2 4 h-147h
Electricity	3.4 KWII
PC-based concrete	
Density	2360 kg/m ³
Weight (as reference of the functional unit)	94.4 kg
PC-based concrete production	J4.4 Kg
CEM I	14 4 ko
Gravel	48 kg
Sand	24.8 kg
Water	7 2 kg
THE CI	,.2 ng

mixture SSS–water–sand is then shaped via compacting (pressing). The shaped mix, which has the desired dimensions but only minimal strength, is then placed in a curing reactor where it is put into contact with 8.5 kg CO₂ at elevated pressure (5 bar), in a CO₂ (> 90%) atmosphere. No CO₂ is released in the atmosphere during the curing. The stream of CO₂ used in the curing reactor is considered to be generated by three different CO₂-recovery processes: cryogenic separation (scenario 1), membrane separation (scenario 2) and chemical absorption via MEA (scenario 3). A more detailed description of the inventory for the three processes is reported in the next section of this paper. The reaction of CO₂ with the Ca and Mg bearing minerals in the SSS forms the carbonates that yield the required compressive strength. The carbonated blocks resulting from the curing process are immediately ready to be commercialised.

 CO_2 recovery. As mentioned above, the study presents three scenarios, where CO_2 is obtained from three different sources: flue gases from steel plants or waste incineration plants (CO_2 concentrations of 10–20% (Mikunda et al., 2015), fermenters producing biogas (25–55% CO_2 (Zhang et al., 2013)), and ammonia production waste gas (the initial concentration of CO_2 is not declared by the CO_2 producers from ammonia waste streams). The three gas streams undergo a CO_2 recovery process that concentrates the CO_2 for carbonation. Each process could potentially recover CO_2 from different gas streams, as for instance chemical absorption can be applied both to steel plant flue gas and ammonia production waste gas. Hence, for the purpose of simplicity, the three considered scenarios refer to the classification presented by Cuéllar-Franca and Azapagic (2012), which report the most commonly used gas stream for each CO_2 recovery processes:

- Scenario 1: cryogenic process (in the case of steel flue gas)
- Scenario 2: membrane separation (in the case of biogas)
- Scenario 3: chemical absorption through monoethanolamine (MEA) (in the case of ammonia production waste gas)



Fig. 4. Life cycle inventory for the carbonated blocks and the PC-based concrete. The inconsistency in the mass balance of carbonated blocks production (total input: 106.7 kg, total output: 99.9 kg) derives from the challenge in accounting the exact amount of water that is released during curing. Part of this water is created by the carbonation of hydroxides and hydrates in the SSS and it can be recycled.

Carbonated blocks

It is assumed that the three CO_2 recovery processes produce a pure CO_2 gas stream. Although a purity of 100% of the CO_2 gas stream is not required by the carbonation process, a lower CO_2 concentration generally prologues the carbonation process.

S1 – Cryogenic separation of CO₂ from flue gas. S1 focuses on the recovery of CO₂ from steel production and waste incineration via cryogenic separation. Cryogenic separation is a relatively novel technology that removes CO₂ from flue gas in a liquid or solid form. The CO2 obtained from the cryogenic separation can be readily reused for the carbonation process (Tuinier et al., 2010). In a cryogenic separation system, the CO₂ is separated from other gasses by condensing it at extremely low temperature. The amount of CO₂ recovered ranges between 90% and 95% of all CO₂ initially contained in the flue gas (Khoo and Tan, 2006). Therefore, the main advantages of the cryogenic CO_2 separation are that no chemical absorbents are required and that the process can be operated at atmospheric pressure (Clodic et al., 2005). On the other hand, the high energy demand required to keep the process at low temperature may affect the environmental performance of the cryogenic process. As reported by Khoo and Tan (2006), cryogenic CO₂ separation requires 630 kWh per ton of CO₂ recovered as liquid form.

S2 - Membrane separation of CO2 from biogas. In S2, a membrane separation process is used to separate the CO₂ from the biogas produced by an anaerobic digestor for organic waste. Membrane separation is a physical process that allows CO₂ to pass through a membrane while excluding all other gasses. Polymeric gas separation membranes are the most common membranes used for CO2 recovery, with an energy demand around 75 kWh per ton of CO2 recovered, which is determined mostly by the pressure ratio needed across the membrane, and a removal rate around 82-88% of CO2 from the biogas (Khoo and Tan, 2006). Additionally, as shown in Fig. 3 for scenario 2, the membrane separation of biogas produces a double output: the CO₂ stream on one side, and purified biogas (mostly concentrated CH₄) on the other side. Therefore, when accounting for the environmental impact of CO₂ recovery through membrane separation, the impact must be allocated between the two products (CO2 and purified biogas). According to the ISO 14044 (2006), allocation in LCA is defined as the partitioning of the inputs/outputs of a multifunctional process between the product under study and the other products of the process. The use of market price as allocation criteria is often found in practice. The allocation based on prices is commonly used because of its simplicity and its ability to summarise complex attributes of the different products produced during a multifunctional process (Ardente and Cellura, 2012). For the current study, the environmental impacts arising from the membrane separation process are allocated between CO₂ and purified biogas according to their market prices. According to Mikunda et al. (2015), the price of purified biogas in 2015 is 168 USD/t, while the market price of liquid CO2 is 115 USD/t. Therefore, 41% of the impacts from the membrane separation process are assigned to the CO₂ production, while the 59% are assigned to the purified biogas production.

S3 – chemical absorption of CO₂ via MEA. S3 analyses the CO₂ recovery through chemical absorption during ammonia production. Chemical absorption by solvents is the most widely used process for CO₂ removal, while MEA is the most commonly used solvent in this process (Cuéllar-Franca and Azapagic, 2015). The MEA is completely regenerated during the process, but high heat consumption is required for solvent regeneration. Data for the CO₂ chemical absorption considered in this study refer to the dataset described by Althaus et al. (2007), which also represents the reference process for CO₂ chemical absorption in the Ecoinvent 3.3 database. This dataset represents the extraction and purification of CO₂ from an ammonia production flue gas. The energy demand is estimated to be 400 kWh per ton of CO₂ recovered.

PC-based concrete. The concrete mix design represents the ratio between cement, sand and coarse aggregates in concrete, and it plays a critical role in defining concrete performances such as durability,

compressive strength and workability. The concrete mix design for a required compressive strength can be calculated using the BRMCA methods, developed by the British Ready Mixed Concrete Association (Newman and Choo, 2003). Based on the BRMCA calculation for the required compressive strength of 40 MPa, the PC-based concrete has a density of 2360 kg/m³, and the design mix is 360 kg/m³ of cement CEM I, 620 kg/m³ of sand, 1200 kg/m³ of coarse aggregates and 180 kg/m³ of water. All data for the inventory of PC-based concrete production are also summarised in Table 3 and in Fig. 4.

2.2.3. Life cycle impact assessment

The life cycle impact assessment (LCIA) uses specific characterisation models to translate the inventory data into environmental impact categories. Characterisation models usually refer to two main approaches: the "problem-oriented", or midpoint, and the "damage-oriented", or endpoint. The midpoint analysis assesses the contribution of each material/energy flow to several different environmental categories, while the endpoint analysis identifies environmental issues potentially damaging the three main aspects of the natural environment, namely human health, ecosystem and natural resources.

As previously discussed by many authors (see for instance Benetto et al. (2004) or Kägi et al. (2015)), the midpoint analysis provides results that are reliable, since they are based on scientifically sound methods. However, midpoint results may not answer questions such as "is product A environmentally better than product B?", as some midpoint categories may show different tendencies than others. The decision-makers, which are considered among the final users of the LCA results, may consequently base their decision on an arbitrary weighting method, dependent on their subjective interests and experiences. In order to avoid decisions based on subjective considerations, the midpoint analysis can be aggregated to the endpoint level. The endpoint method proposes a weighting procedure that aggregates the results from the midpoint categories to endpoint damages. Although the results from the endpoint analysis are easier to be interpreted and compared, they contain additional uncertainty due to the aggregation and weighting process. Therefore, the combination of midpoint and endpoint results can assist in better interpretation and transparency: the midpoints allows to detect the highest contributing processes and to find possible room for improvements; the endpoint provides a general picture of the environmental costs and benefits for each scenario, making the results easy to be compared with results from other scenarios (Di Maria et al., 2018). For the reasons discussed above, the current study presents the LCIA results in both midpoint and endpoint.

For the midpoint analysis, the International Reference Life Cycle Data System (ILCD) v.1.09 framework is used. The ILCD framework, developed by the European Commission – Joint Research Centre (EC-JRC), selects the recommended methods for each midpoint impact category starting from a pre-selection of existing methods. For instance, for the category climate change, the recommended calculation method is the IPCC model with 100-year time-horizon, while for human toxicity the ILCD recommends the USEtox model 2008. A more detailed explanation of all the ILCD recommended methods for midpoint analysis can be found in JRC (2011).

On the other hand, no recommendations are currently provided within the ILCD framework for endpoint analysis, because of the unavailability of sufficiently mature models for most of the endpoint categories. Therefore, the endpoint analysis performed in the current study refers to the another widely accepted environmental assessment methodology, called Recipe, which includes three endpoint environmental damages: (i) human health damage, (ii) loss of species in ecosystems, and (iii) resources depletion (Goedkoop et al., 2012).

3. Results

The results of the midpoint analysis are shown in Fig. 5. The comparison between the three scenarios for carbonated blocks and the PC-



Fig. 5. Midpoint results (AE: acidification; GWP100: climate change midpoint, excl biogenic carbon; GWP100bio: climate change midpoint, incl biogenic carbon; CTUe: ecotoxicity freshwater; P: eutrophication freshwater; N: eutrophication marine; AEeutr: eutrophication terrestrial; CTUhcan: human toxicity, cancer effects; CTUh: human toxicity, non-cancer effects; U235: ionising radiation, human health; LU: land use; ODP: ozone depletion; PM2.5: particulate matter/respiratory inorganics; POF: photochemical ozone formation, human health; RDW: resource depletion water; RDF: resource depletion, mineral, fossils and renewables).

based concrete (columns form left to right: S1, S2, S3, PC-based concrete) is shown for each midpoint category. Since midpoint categories are expressed in different units, the results are normalised to the scenario having the highest impact (expressed as 100% in the graph). The traditional PC-based concrete has the highest impacts in nine out of the sixteen analysed midpoint categories (acidification, climate change biogenic and non-biogenic, ecotoxicity freshwater, eutrophication freshwater, human toxicity cancer and non-cancer effect, land use, resource depletion minerals). For instance, considering the category climate change including biogenic carbon (carbon that is already within the carbon cycle), the carbonated blocks present an emission reduction of 68% for S1, 77% for S2 and 71% for S3, compared to the PC-based concrete emissions. On top of that, the negative part of the graph in both climate change categories (biogenic and non-biogenic) represents the amount of CO2 uptaken by the carbonated blocks during the carbonation process (8.5 kg). The results show that the final CO₂-equivalent balance (the difference between the CO₂-equivalent emitted minus the CO₂-equivalent uptake) is negative for all carbonated blocks scenarios (-1.09 kg and -1.01 kg for S1; -1.96 kg and -1.90 kg for S2;-1.36 kg and -1.28 kg for S3). Therefore, mineral carbonation is a negative-carbon-footprint technology, since the amount of CO₂ uptake and stored during the process is higher than the amount of CO₂ emitted, considering the whole life cycle. The carbonated blocks also present significant impact reductions in the categories eutrophication freshwater (-64% for S1, -77% for S2, -69% for S3), human toxicity noncancer effect (-51% for S1, -73% for S2, -60% for S3) and resource depletion of minerals (-60% for S1, -72% for S2, -65% for S3). On the other hand, the carbonated blocks technology presents higher impacts compared to PC-based concrete in the categories eutrophication marine and terrestrial, ionising radiation, ozone depletion, particulate matter, photochemical ozone formation and resource depletion water. Looking at the single process contribution, the highest quota in almost all categories is given by the production and use of diesel in the drying process (equal for all carbonated blocks scenarios). The only difference between the three carbonated blocks scenarios is the electricity consumption to recover the CO_2 from the different sources. Since cryogenic CO_2 recovery is the technology requiring the highest amount of electricity, S1 is the scenario with the highest impacts. The electricity for CO_2 recovery has a most relevant contribution (> 50% on the total impact of the carbonated blocks) for S1 in the categories ionising radiation (63%) and resource depletion water (55%). Finally, as expected, for all analysed categories, cement production causes the highest contribution to the final impact of PC-based concrete.

The midpoint analysis provides a detailed description of the cause-effect relationship between emissions and their direct effect on the environment. However, the results in Fig. 5 do not help to conclude which is the material with the lowest environmental impact, as the analysed cases show different trends in different categories. Therefore, an aggregation to the endpoint can help to identify which of the analysed materials presents the best environmental profile. Fig. 6 shows the results of the endpoint analysis. PC-based concrete shows the highest impacts in the damages human health and ecosystems, while S1 has the highest impact on the damage resources. The scenario S2 has the lowest contribution for all three damages, while S3 has a higher impact compared to PC concrete only in the damage resources. Looking at the contributions of every single process, the endpoint results confirm the outcomes of the midpoint analysis. For the damages human health and ecosystems, the avoided impacts from the CO₂ uptake are higher than the impacts caused by the production of the carbonated blocks, for all three scenarios. At the same time, cement production contributes to 95% of the total impact of PC-based concrete. For the damage resources, the electricity and diesel consumption during the carbonated blocks production process have the highest contribution for all carbonation scenarios.



Fig. 6. Endpoint results.

3.1. Interpretation and limitations

The LCIA results discussed above highlight the environmental advantages and disadvantages of the production of the carbonated blocks compared to the production of traditional PC-based concrete. The midpoint analysis showed how carbonation technology presents a significant reduction in some categories such as climate change, where the impact is reduced by 70% compare to the PC-based concrete. Moreover, the results show that the amount of CO₂ uptake and stored within the carbonated blocks is higher than the CO₂ emitted during the whole mineral carbonation process. This negative-carbon balance is also confirmed by the endpoint analysis, where human health and ecosystem present a negative impact for all carbonation scenarios. Among the three considered carbonation scenarios, the S2 always presents the lowest impacts, due to the lower energy required by the membrane CO₂ recovery process.

On the other hand, in some other midpoint categories and the endpoint damage resources, all the carbonation scenarios present higher impacts compared to the traditional PC-based concrete. Looking at the single processes, the highest contribution for the carbonated blocks production is given by the fossil fuels used during the drying process. Therefore, a reduction of the moisture content in the wet slag, or an increased optimisation of the drying process could sensibly reduce the impacts of the carbonated blocks. This may lead to further improve the environmental profile of the carbonated blocks, especially for those categories where the carbonated blocks present a higher impact compared to PC-based concrete. The electricity consumption for CO_2 recovery significantly affects the final impact of the carbonated blocks (> 50%), especially in S1, for ionising radiation and water depletion in the midpoint, and resources in the endpoint.

It must also be noticed that carbonation of SSS (and in general any technology based on the recovery of metallurgic slag to produce new construction materials) is a constrained technology, meaning that the production capacity (or supply) is inelastic, as it cannot be adjusted to meet an increase in demand for the product. Due to the difference between the price of the stainless steel and the price of the SSS, the production is driven by the demand for stainless steel rather than the demand for SSS. Consequently, the metallurgic process will focus on the properties of stainless steel, rather than on the quality and volumes of the SSS. Therefore, variable compositions of the SSS may affect the technical and environmental performance of the carbonated blocks.

Finally, it is also worth to highlight the limitations of the presented LCA results. A first limitation is represented by the assumptions made to solve the allocation issue. First, an allocation problem arises when deciding what share of the environmental burdens of the stainless steel production generating the SSS should be allocated to the carbonated blocks. Following the Waste Framework Directive 2008/98/EC and the recommendations put forth in the ISO 14041, an allocation coefficient should indeed be applied only if the waste can be considered as a by-product, while no allocation is advised if the waste is considered as an unintended residue. So far, SSS does not have the status of a by-product, because of the uncertainty on its further use as secondary resources.

Consequently, SSS is today legally considered as waste material. Therefore, for the LCA presented in this paper, the allocation procedure has been avoided, and no impacts are attributed to the SSS. However, in the future, the carbonation technology is expected to develop, and further applications of an allocation coefficient for SSS may be needed, although, given the price ratio between stainless steel and slag, the effect may be limited.

Another limitation of the LCA study is represented by the intrinsic limitation of LCA when it comes to industrial decision-making and evaluation of industrial symbiosis applications. As clarified by Marvuglia et al. (2013) and Vázquez-Rowe et al. (2014), the traditional LCA approach (known as attributional approach) provides a sound environmental analysis and understanding of the main environmental impacts within the concerned production system. On the other hand, it omits the analysis of potential indirect effects engendered in the markets. Therefore, attributional LCA results provide an excellent environmental analysis at a product level, which enables a reliable comparison between alternative products. However, an expansion of the analysed system may be required to draw significant conclusions on the environmental consequences of product substitution.

4. Conclusions and perspectives

Aiming at the reduction of carbon dioxide (CO₂) emissions, carbon capture utilisation (CCU) technologies are raising the attention of the industries and the scientific community. Among the CCU technologies, mineral carbonation acts by chemically binding CO₂ in alkaline materials to produce a stable carbonated block that can be used by the construction industry. The CO₂ can be recovered from flue gases derived from carbon-intensive industrial processes, while the alkaline material can be a solid residue from other industries. Therefore, mineral carbonation can have several potential environmental benefits: (i) the valorisation of residues from other industries, (ii) the recovery of CO₂ from industrial flue gases, and (iii) substitution of traditional PC-based construction blocks with negative or low-carbon products.

On the other hand, the energy required by the carbonation and the CO_2 recovery processes may offset the beneficial effects. To understand the overall environmental performance of mineral carbonation, this study presents a life cycle assessment (LCA) of carbonated blocks made from stainless steel slag (SSS). The data for the carbonation process is collected from a real industrial demonstrator situated in Belgium. The study analyses three different scenarios, based on literature data for three techniques for CO_2 recovery: (i) cryogenic process; (ii) membrane separation; (iii) chemical absorption through MEA. Finally, the study compares the environmental performance of the carbonated blocks with the ones of traditional Portland cement (PC) based concrete with equivalent technical performance.

The results of the midpoint analysis show that the carbonated blocks have lower environmental impacts in nine out of sixteen impact categories, compared to PC-based concrete. For instance, all three carbonation scenarios present a significant reduction in global warming potential. A key finding is that the results show that the quantity of CO_2 uptake by the carbonation process is higher than the quantity of CO_2 emitted by the process. Therefore, the mineral carbonation process to produce carbonated blocks from stainless steel slag residue can be considered as a potentially negative carbon-footprint technology. The results of the endpoint analysis confirm that for the damages to human health and ecosystems, the carbonated blocks have a lower impact compared to the traditional PC-based concrete, and an overall negative impact.

On the other hand, although the industrial demonstrator showed a significant increase in the energy efficiency of the process compared to previous LCA studies based on lab-scale data, the energy consumption represents the main environmental hotspot. As a consequence of the energy requirements, the carbonation process has higher impacts in seven midpoint categories and the endpoint damage resources. Therefore, the future industrial development of mineral carbonation should further increase the energy efficiency of the process and find alternative sources for electricity and heat requirements, such as recovery of waste heat of the exothermal carbonation process. Another environmental hotspot highlighted by the LCA study is the electricity consumption for the CO_2 recovery process. This result confirms the outcomes of previous LCA studies. The comparison between the CO_2 recovery technologies showed that membrane separation is the technology with the lowest environmental impacts based on available data.

Nonetheless, when integrated into the carbonation process, all analysed technologies present environmental advantages. In this context, to support the introduction of the technology and development of industrial synergies, the development of CO_2 valorisation networks should be facilitated. Legislative frameworks and infrastructure development should be supported by the public sector, to enable CO_2 stream sharing between neighbour industries. Also a symbiosis network/platform for material streams exchange can serve as a base to the further development of a CO_2 valorisation network.

The construction sector is considered a critical sector for sustainability, and LCA represents an opportunity in this context. For instance, the implementation of green certificates based on LCA reports can be an essential asset to increase green public procurements for mineral carbonation. Green public procurement is a powerful tool to drive the whole sector towards sustainable production, as the public sector plays a vital and pioneering role in the market. Moreover, mineral carbonation represents an economic opportunity for the construction sector, since it can replace conventional PC-based concrete, which requires the extraction of primary raw materials, and it produces a significant amount of CO_2 emissions which, in Europe, can incur significant financial costs in the EU emission trading system. At the same time, quarrying activities are under increasing environmental and societal pressure or are subject to spatial constraints.

On the other hand, mineral carbonation of SSS is a resource-constrained technology, depending on the production of stainless steel. Therefore the quality and the quantity of SSS available for carbonation are dependent on the demands for the stainless steel. Consequently, future development of SSS carbonation technology will not likely change the amount of SSS available. In Belgium, for instance, a SSS production of 500 kt/y has been calculated, while recent data estimates a cement PC production in Belgium of 6500 kt/y. It is clear that the carbonated blocks from SSS can represent only a small share of the total construction materials market in Belgium. Therefore, ongoing research is currently undertaken to use other alkaline residues, such as concrete demolition waste, incineration and biomass ashes. This can strongly expand the volumes of secondary raw materials available for the mineral carbonation process.

Conflict of interest

ORBIX and VITO collaborated in the development of the Carbstone technology and have acquired joint intellectual property. KU Leuven took part in the study as an independent party, therefore KU Leuven declares no conflict of interest.

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