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Environmental assessment of aqueous alkaline absorption of carbon dioxide and its use to produce a construction material

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ABSTRACT

The use of carbon dioxide emissions as a raw material, e.g. for the chemical industry and for the production of synthetic fuels, is regarded as an alternative to its storage, as the potential benefits are dual: emissions are removed at the same time as greenhouse gases from the substitution of conventional goods are avoided. However, there is an outstanding lack of information on the overall carbon dioxide balance for these new approaches. This paper performs the greenhouse gases balance and resource efficiency assessment, in terms of water and energy use, of the production of a new construction material intended to have a negative or neutral carbon footprint, which composition is mainly magnesium carbonate trihydrate, known as nesquehonite. Desalination brines are considered the main source of Mg for the aqueous precipitation, which requires and alkaline agent. The overall carbon dioxide emissions balance of the process strongly depends on the amount of alkali consumed and the efficiency of its use. Additionally, the substitution of commercial materials with relatively high environmental footprint by the obtainable magnesium carbonates would yield a net reduction of emissions, embodied energy and water footprint. The capture process would have a net negative carbon footprint if the associated emissions linked to the alkalinity source were in the order of 22–27 kg of CO_2 per kmol of alkalinity. However, no commercial alkali source reaches such a low associated emissions, so alkaline wastes can be used.

1. Introduction

The average concentration of carbon dioxide, CO_2, in the atmosphere recently reached 402.24 ppm (ESRL, 2014). This accumulation, generated primarily through the combustion of fossil fuels, is regarded as the main cause of climate change. A decarbonised economy would halt this effect, but in the short-term carbon dioxide capture is a more realistic approach. The conventional process of carbon capture requires its separation from a gas stream with an amine absorption process, transportation and storage of the CO_2 mainly in liquid form with final disposal in exhausted oil fields or geological reservoirs. The technological feasibility has already been proven and demonstration facilities are already in operation (IEA, 2007). The process, known as Carbon Capture and Storage (CCS), can be implemented at large scale and the technology employed is mature and well understood. However, cost, energy demand and the massive investment required for infrastructure are still challenges to overcome (ETI, 2015).

Part of the research effort on CO_2 capture focuses on increasing the usability of the molecule as industrial chemical feedstock carbon dioxide can be used in the manufacture of synthetic fuels and chemicals, or directly applied as a clean refrigerant, carbonation agent, displacement fluid in enhanced oil recovery, clean acid for chemical neutralisation, raw material in urea manufacturing, as a solvent, etc. (Styring and Jansen, 2011; Amoureux et al., 2014). Very few processes achieve, or are designed to yield, a product from captured CO_2 with a net negative carbon footprint. A good example is curing of concrete mixtures by carbonation of concrete samples with additions of γ-dicalcium silicate, that can achieve carbon negative or neutral formulations (Higuchi et al., 2014). However, the total use of CO_2 as a chemical feedstock in the world is estimated to be less than 0.6% of the total emission (Aresta et al., 2013). Large scale *utilisation* of CO_2 is therefore still far from being a realistic mitigation option.

In this paper we investigate the environmental profile of carbonate precipitation using an aqueous CO_2 capture route in alkaline

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Nomenclature

a	moles of alkali consumed per mol of captured CO_2
AW	alkaline waste
CED	cumulative energy demand
C_{Mg}	concentration of Mg in the brine, g/L or kg per m^3
E_D	greenhouse gases emissions associated to the desalination of 1 m^3 of water in kg CO_2e per m^3 of water
E_F	greenhouse gases emissions from fossil fuel combustion for heat generation, in kg CO_2 per MJ
E_K	greenhouse gases emissions associated to the alkali, kg CO_2e per tonne of alkali
E_S	greenhouse gases emissions associated to the substituted material, in kg of CO_2 per tonne
GHG	greenhouse gases
HM	hydromagnesite
IPCC	Intergovernmental Panel for Climate Change
MED	multieffect distillation
M_K	molecular weight of alkali, kg/kmol
MSF	multistage flash distillation
NQ	nesquehonite, $MgCO_3{\cdot}3H_2O$
RO	reverse osmosis
S	substitution ratio (mass of used solid carbonate per mass of substituted material)
TDS	total dissolved solids
W	water used in the final washing of the carbonate, in m^3 per tonne of dry solid
W_K	water footprint of the alkali, in m^3 per kg of alkali
W_s	water footprint of the substituted material, in m^3 per kg of material
X_D	conversion of seawater to desalinated water (vol/vol)
X_{NQ}	rate of carbonate recovered assumed to be nesquehonite
Y	CO_2 absorption efficiency (mol/mol)
Z	Overall energy consumption in the activation of 1 kg of carbonate, GJ per tonne or MJ per kg
Z_K	embodied energy of the used alkali, MJ per kg
Z_s	embodied energy of the substituted material, MJ per kg
Z_W	embodied energy of desalinated water, MJ per m^3
ΔG^0	Gibbs free energy at 25 °C and 1 atm

media. The first approach for CO_2 capture using alkaline reagents was the mineralisation of Mg-rich minerals, e.g., olivine, Mg_2SiO_4, abundant enough in nature for long-term capture, but not as a solution with low associated impact, low embodied energy and/or cost-effectiveness (Haywood et al., 2001; Mazzotti et al., 2005). For example, quarrying and crushing Mg silicate rocks, dissolution in HCl and subsequent reaction of the liberated magnesium with CO_2, and using alkali as a catalyst consumes 23 MJ per kg of captured CO_2 and has an associated emission value of 1.3 kg of CO_2 per kg of captured CO_2 (Haywood et al., 2001).

The use of magnesium and calcium from seawater or desalination brines simplifies the wet mineralisation process, eliminating the dissolution step required for Mg or Ca silicates, the main debit factor in the economics of carbon capture by mineralisation. The process is simpler, requiring much lower temperatures and may produce a carbon-negative material suitable for construction. This is the aim of the Calera process (Pekdemir, 2015; Calera, 2012). The intrinsic nature of aqueous CO_2 is acidic, and needs to be hydrolysed in order to obtain carbonate or bicarbonate, so alkaline raw materials are always required to enhance the solubility of captured CO_2 and improve the kinetics of its absorption. The Calera process produces the required alkali, sodium hydroxide, through the ABLE process (Alkalinity Based on Low Energy), a low cost form of electrolysis (Zaelke et al., 2010). Some researchers have explored selection of low associated impact alkalinity, which is usually taken from highly alkaline wastes (by definition, with zero impact), e.g. steel slags or fly ash (Mattila et al., 2012; Eloneva et al., 2012; Calera, 2012). However, one essential point is the proximity of bulk quantities of the reactants. Carbon dioxide in flue gas and brines from desalination, as studied by Ferrini et al. (2009), De Vito et al. (2012) and Ma and Yoon (2013), are locally available and close to each other in an integrated power–water supply system.

These integrated desalination–power plants are often used in the Middle East. For example, Qatar has the highest CO_2 emissions per capita due to the production and consumption of fossil fuel. The Qatar National Vision 2030 includes climate change policies for the reduction of greenhouse gases emissions, at the same time aiming for a *high* standard of living (Alhorr et al., 2014). This *high* standard of living is reflected in the high consumption of water per capita, around 600 L per inhabitant per day, one of the highest in the world. At present all drinking water has to be produced by desalination using multi-stage flash distillation, using recovered waste heat from natural gas power plants (Darwish et al., 2013).

The research team of the University of Aberdeen, in cooperation with the Gulf Organisation for Research and Development (GORD) in Qatar, has developed a self-cementing material based on Mg carbonates from desalination brines and CO_2 gas from the exhaust of power plants (Morrison et al., 2014). The development of a construction product of neutral or even negative carbon footprint, using this new type of cement, would reduce the increasing environmental pressure of the intense construction activity and of the built environment in the Gulf region. Through this paper we present: (i) the theoretical framework for calculation of the impact of such a material; (ii) the greenhouse gas balance of the process and its resource efficiency (in terms of metrics for water and energy consumption) in a cradle-to-gate perspective, to identify the main hotspots of production of this carbon negative material; and (iii) the evaluation of several scenarios to assess the system performance against process efficiency, raw materials source and the potential substitution of commercial products.

2. Methodology

Given the background and the nature of the problem to be assessed, the optimal methodology considered is Life Cycle Assessment, LCA, as defined by ISO standard 14040:2006. However, due to the fact that there is no industrial facility producing the target material and the lack of knowledge on the main environmental hotspots of the production process, we decided to perform a simplified LCA as defined by Guinée (2004).

2.1. Definitions

In terms of impact assessment, three terms, very commonly used in the life-cycle assessment and carbon capture fields are defined in this section:

- An "*embodied*" property refers to the total amount of that property is required in the extraction, processing, manufacture and delivery of a product. It is commonly used in the assessment of construction materials as a metric of the environmental impact of materials in a building system (ICE, 2015). In this paper, it is used exclusively for energy; for the carbon dioxide and water balances the term "*footprint*" will be used instead.