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Comparative life cycle assessment of reactive MgO and Portland cement production

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ABSTRACT

Reactive magnesia (MgO) cements are proposed as a potentially sustainable binder due to their lower production temperatures (~800 vs. 1450 °C) than Portland cement (PC) and ability to fully carbonate and gain strength during setting. Reactive MgO is mainly produced via the calcination of magnesite. Environmental implications of reactive MgO production must be analyzed before any final conclusions can be made regarding their contribution to the sustainability of the cement industry. This study evaluates the environmental impacts of reactive MgO production and provides a comparison with PC production using a life-cycle assessment (LCA) approach. The advantages of MgO production with respect to radiation, ozone layer, eco-toxicity, acidification/eutrophication, minerals and fossil fuels outweigh the disadvantages when compared to PC production. MgO has a lower impact on the overall ecosystem quality and resources than PC, but poses a larger damage to human health due to the high coal usage by most plants. The decomposition of magnesite releases a higher amount of CO_2 than limestone (~1.1 vs. 0.78–0.83 t/t), creating a higher climate change score for MgO despite its lower production temperatures. However, when the carbonation capability of MgO cements is considered, their net CO_2 emissions are ~73% lower than PC. The introduction of a strength ratio modification to MgO production results in ~13% and ~32% lower CO_2 intensity and damage impact on human health when compared to PC production, respectively. The influence of energy type and amount on different impact and damage categories associated with MgO production was revealed by scenario modelling. The quantity of raw materials had a negligible effect on the overall environmental impact of reactive MgO production, whereas increasing the emissions led to an increase in climate change. Energy was identified as the key parameter with the highest influence on the environmental burdens of reactive MgO production. Results indicate that the use of alternative fuels could further improve the overall sustainability of reactive MgO cement production.

1. Introduction

Climate change induced by increased concentrations of anthropogenic carbon dioxide (CO_2) emissions is the largest environmental threat of the 21st century. Current CO_2 levels in the atmosphere are rising at a rate of 1.85 ppm/year and have reached 400.11 ppm in 2015 (NOAA). This has led to a growing interest in sustainable practices in line with the increased pressure on industries to develop alternative technologies with reduced CO_2 emissions. Due to the use of large quantities of raw materials and high energy consumption processes, the manufacture of Portland cement (PC) contributes to ~5–7% of the total anthropogenic CO_2 emissions globally (Allwood et al., 2010; Friedlingstein et al., 2010; Huntzinger and Eatmon, 2009; Van Oss and Padovani, 2002). As it is a major contributor to climate change, an accurate and thorough quantification of the environmental impacts of the cement industry can facilitate the understanding of how to substantially reduce emissions. This can have a significant influence in mitigating global warming (Boesch and Hellweg, 2010).

Around 60% of the CO_2 emitted by cement plants is derived from the calcination process (i.e. decomposition of $CaCO_3$ into CaO and CO_2), whereas the rest is from the energy consumed during this process (Allwood et al., 2010; Van Oss and Padovani, 2002). The extent of CO_2 emitted is heavily dependent on the manufacturing method and the type of fuel utilized during production (Hendrik and van Oss, 2010; Marceau et al., 2006; Taylor et al., 2006; Valderrama et al., 2012). In addition to the CO_2 discharge, nitrogen oxides (NO_x), sulfur oxides (SO_x), cement kiln dust (CKD),

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particulate matter (PM), carbon monoxide (CO) and heavy metals are emitted into the atmosphere during cement production (Boesch and Hellweg, 2010; Lei et al., 2011; Magrama, 2012; Valderrama et al., 2012; Wang, 2013). A major source of air contaminants, 40% of PM emissions are from industrial origins. PC plants account for approximately 15–27% of PM, 3–4% of sulfur dioxide (SO_2) and 8–12% of NO_x emissions in China (CEYEC, 2001; Lei et al., 2011; Li and Li, 2013). Similarly in Europe, the production of 1 tonne of Type I PC results in the average emissions of 2.4 kg of NO_x, 0.6 kg of SO_2 and 2.7 kg of dust (Josa et al., 2007). These emissions are mainly due to the impurities present within the raw materials and the combustion of fuels used during cement production (Agency, 1994; Oss and Padovani, 2003; Steinberg, 1985). Crude limestone contains a high amount of impurities (i.e. Fe_2O_3, SiO_2, Al_2O_3 and MgO), leading to their release during calcination. Therefore, there is an urgent need to identify potential solutions for cement production with lower environment impacts.

One promising solution is the development of cement binders with alternative compositions, an example of which is reactive magnesium oxide (MgO) cements. These binders have the ability to absorb CO_2 in the form of stable carbonates during curing and gain strength accordingly (Harrison, 2008; Unluer and Al-Tabbaa, 2014). MgO is found in minerals such as dolomite ($CaMg(CO_3)_2$) and magnesite ($MgCO_3$). Magnesium (Mg) is also the third most abundant element in seawater (Shand, 2006). MgO is obtained by two main methods:

(i) Calcination of magnesia-based minerals (e.g. magnesite or dolomite), and
(ii) Synthetically from brine or seawater

A majority of the MgO utilized today is currently produced from the calcination of $MgCO_3$, which is found in two physical forms: (i) cryptocrystalline or amorphous magnesite; and (ii) macrocrystalline magnesite (Cardarelli, 2008). Pure magnesite contains 47.6% MgO and 52.4% CO_2 by weight. Small amounts of calcite ($CaCO_3$) and siderite ($FeCO_3$) can also be detected in natural magnesite, which can coexist with dolomite generally found in the form of sedimentary rocks (Alderman and Von der Borch, 1961; BREF, 2010; Cardarelli, 2008). Raw magnesite is widely used in ceramics, surface coatings, landscaping and flame retardants (Cardarelli, 2008).

It is estimated that about 14 million tonnes/year of MgO is produced world-wide. For comparison, production of PC exceeds 4 billion tonnes/year (USGS, 2014). Although magnesite resources are widely available, 75% of the global magnesite production is from China, North Korea, Slovakia, Turkey, Russia, Australia and India (Shand, 2006). The total production of MgO from magnesite is ~8.5 million tonnes/year (USGS, 2012). The 3.7 billion tonnes of magnesite reserves in China account for ~29% of the world's total reserves, ranking first in the world (Xu, 2013). In addition to having the largest magnesite reserves in the world, China is the largest producer, consumer and exporter of MgO. An annual yield of 3.8 and 4.8 million tonnes of reactive and dead-burned MgO were reported by China in 2011, respectively. A majority of the production takes place in the Liaoning province, where 85% of the total magnesite resources in China are located and 16 million tons of various grades of magnesite are mined annually (Chen and Wei, 2013). Calcination of magnesite usually takes place in shaft or tunnel kilns. Low quality control measures practiced in China result in inconsistent calcination conditions (temperature and residence time), which can cause large temperature variations at different locations within the kiln, producing MgO with heterogeneous reactivity.

Similar to the production of PC from limestone, magnesite is heated to produce MgO, meanwhile emitting CO_2. Calcination of $MgCO_3$ to form reactive MgO occurs at temperatures between 550 and 800 °C and is an endothermic reaction (i.e. total energy requirement of ~2415 kJ/kg), as indicated in Equation (1). The exact calcination temperature depends on several conditions such as the presence of impurities and the physical properties of the parent material. Higher calcination temperatures may be necessary as these parameters become more prominent. Nevertheless, the lower calcination temperatures used in the production of reactive MgO cement compared to PC (800 vs. 1450 °C) allows the use of alternative fuels with relatively low heating values.

$$MgCO_3 \rightarrow MgO + CO_2 \tag{1}$$

Sintering occurs during the calcination of $MgCO_3$, which reduces the porosity of the final product. Calcination conditions such as temperature and residence time are key parameters that influence the final properties such as the specific surface area (SSA) and reactivity of the MgO produced (Shand, 2006; Thomas et al., 2014). Increasing the calcination temperature and residence time decreases MgO reactivity. This is accompanied by a reduction in its SSA and an increase in particle size (Eubank, 1951; Mo et al., 2010).

MgO is classified under four categories depending on the calcination temperature used during its production via the dry process route. All the different grades, properties and applications of MgO currently used in different industries are presented in Table 1 (BREF, 2010; Unluer and Al-Tabbaa, 2015). Different from dead-burned MgO, whose utilization as a binder can cause large and localized volume expansions that can create cracking in the hardened cement due to its delayed hydration, reactive MgO undergoes hydration at a similar rate as PC, thus avoiding any compatibility issues (Vandeperre et al., 2008).

Reactive MgO is produced at lower calcination temperatures than PC (800 vs. 1450 °C) and has a high SSA and reactivity. Apart from the growing interest on its use as a cement binder, it is generally used in a range of applications within the pharmaceutical, agricultural and paper industries. Hard-burned MgO has a relatively low SSA and reactivity and is typically used in applications that exploit its expansion properties. Dead-burned MgO, which also has a low SSA and is unreactive, is mainly used in refractories due to its high melting point. Fused MgO possesses low hydration energy, high thermal shock resistance and a compact structure and is used in monolithic refractories and electrical insulating. Out of the several different Mg-based binder compositions currently being developed or commercially available on the market (i.e. magnesium carbonate, phosphate, oxychloride, oxysulfate and silicate-hydrate), magnesium carbonate cements utilizing reactive MgO propose the most promising future due to their ability to sequester CO_2 in the form of stable carbonates.

Reactive MgO has a purity ranging between 75% and 96% (Tececo, 2008). Its main impurities include CaO, Fe_2O_3, Al_2O_3 and SiO_2, whose concentrations depend on the raw materials used during the production process. When compared to the calcination of $MgCO_3$, the synthetic process involving the production of MgO from $MgCl_2$ rich brine or seawater through the wet process route is much more complex. The higher energy requirements and the associated cost of this production route can be justified by the higher purity (>97%) of the final MgO (BREF, 2010; Cardarelli, 2008; Ferrini et al., 2009; Hassan, 2014), which may be desirable in certain applications within the pharmaceutical or food industries.

Although there is an abundance of literature investigating the environmental impacts of PC production (Boesch and Hellweg, 2010; Lei et al., 2011; Marceau et al., 2006; Valderrama et al., 2012; Van Oss and Padovani, 2002), a very limited number of studies have focused on the implications of MgO production. Most of the publications and technical databases on the production of