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Environmental Evaluation for Cement Substitution with Geopolymers

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ABSTRACT

Portland cement production could represent not less than 5 % of the total CO_2 emissions from human activities. Geopolymeric binders appear to be an alternative to traditional Portland cement. However, although intensive research has been done in this field since 40 years, few studies have evaluated the environmental impact of geopolymer binders when they are used as a replacement of Ordinary Portland Cement. In this study a detailed life cycle impact assessment of standard geopolymer production is performed and shows that the use of sodium silicate solutions has large environmental impacts. Fly ash and Blast furnace slag based geopolymers have low environmental impact as soon as fly ashes and slags are considered as waste. Geopolymers done with calcined clays have much higher impacts. Finally geopolymer concretes made with by-products from other industries or with metakaolin do not achieve a significant CO_2 emission reduction to reach factor 4 objectives.

INTRODUCTION

Concrete is the most commonly used construction material; its usage by the communities across the globe is second only to water. Customarily, concrete is produced by using the Ordinary Portland Cement (OPC) as the binder, which is a highly energy intensive product and releases carbon dioxide (CO_2). Actually cement production could represent nearly 10% of total anthropogenic CO_2 emissions in the close future due to the world-wide demand increase for OPC. Numerous studies have dealt with mitigation perspectives in the cement industry [Liu et al. 1995; Worrell et al. 2000; Szabo et al. 2006; Taylor et al. 2006]. A recent study showed that it is possible to achieve a reduction by a factor 2 from the 1990 CO_2 emission level with improvement in the current cement technology, but that a reduction by a factor 4, as it is recommended by the Intergovernmental Panel Group for Climate Change (IPCC), would not be possible without a technological turn around [Habert et al. 2009]. New binders are therefore needed to meet the demand and still achieve the CO_2 reduction goals. Among these new binders it is commonly accepted that sulfo-aluminate clinkers and geopolymers are high potential options. However, although geopolymers are presented by many authors as one of the options for green concrete [Davidovits 1999; Duxson et al. 2007], few studies have quantified the environmental impacts of geopolymers, and no complete environmental evaluation by Life Cycle Impact Assessment (LCIA) method can be found in literature. LCIA is a methodology for evaluating the environmental load of processes and products during their life cycle, from cradle to grave. This methodology is based on international standards of series ISO 14040 [ISO 2006]. LCIA has been used in the building sector since 1990 [Fava 2006] and is now a widely used methodology [Ortiz et al. 2008].

The objective of this study is to perform a detailed life cycle impact assessment of standard geopolymer production and to compare it with the production of OPC based concrete. Geopolymer concrete mix designs found in the literature are used in this paper. A distinction is made between fly ash based, slag based and metakaolin based geopolymers. The different environmental impact categories are then evaluated.

MATERIALS AND METHODS

The LCIA method is separated in 3 main steps before results discussion. Firstly, the *functional unit* which is the function that needs to be kept constant throughout the different processes that will be compared, and the *system boundaries* have to be defined. Once this framework is set, it is possible to collect all the flows of materials and energy that pass through the defined system. This will be the *Life Cycle Inventory* (LCI). And finally, once the LCI is built, it can be transferred into *environmental impact*s with chosen environmental impact indicators. These three steps are detailed in this section.

Functional unit and system boundaries

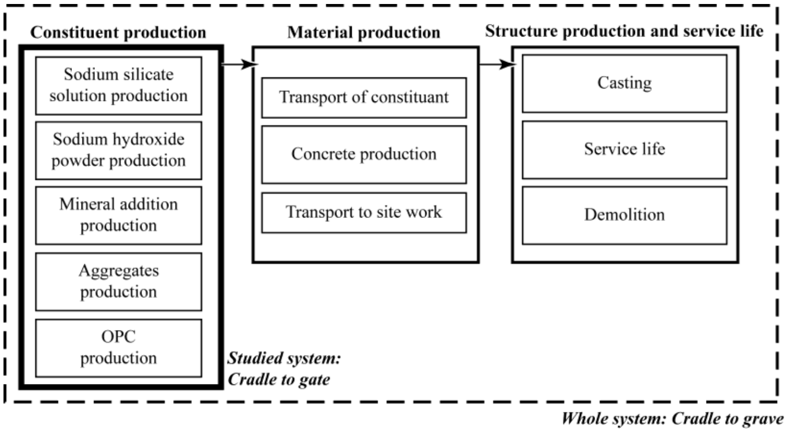

Fig. 1. Boundaries of the System

The studied system is reduced to the production of the constituents needed to make concrete. The analysis does not include every stage of the product's life cycle (*cradle to grave*) but ends at an intermediate stage (*cradle to gate*) as shown in **figure 1**. This can be done, when one analyses a production, such as concrete, which has multiple specific applications in civil engineering (beams, pillars, pavements, houses, bridges, etc.) and therefore disallows a unique life cycle to be defined. This type of partial analysis is useful for the further construction of complete life cycles for specific concrete end-products on a larger scale.

To compare geopolymer based concrete and OPC based concrete it has been chosen to define the *functional unit* as 1 cubic meter of concrete that has the same final compressive strength. The Féret equation [De Larrard 1999] has been used to calculate the cement content of OPC concretes to have the same strength resistance as the geopolymer concretes found in the literature **(equation 1)**.

$$f_c \approx K.Rc_{28}.\left(\frac{V_{cement}}{V_{paste}}\right)^2 \qquad \textbf{(1)}$$

Where f_c is the compressive strength, K a parameter that characterise the aggregate quality, Rc_{28} the specific strength of cement, V_{cement} the volume of cement and V_{paste} the volume of the paste that includes air water and cement. In this study K and Rc_{28} have been set at 4.91 and 65 respectively [De Larrard 1999]. As standard concrete already uses 30% of clinker substitution [Habert and Roussel 2009], geopolymer concretes have been compared with cement based concrete with the same final strength resistance and with a binder made only with OPC or with 30% clinker substitution by fly ash.

Environmental and technical data collection

Geopolymer concrete mix designs come from literature [Bakharev et al. 2001; Lee and Van Deventer 2002; Hardjito and Rangan 2005; Pacheco-Torgal et al. 2005; Sofi, et al. 2007; Silva et al. 2007]. To calculate the amount of cement equivalent to these geopolymers, the geopolymer paste volume is calculated and a similar paste volume is chosen for cement based concrete and used in **equation 1**.

Primary data on cement production have been built by Chen [2009] and presented in Chen et al. [2009a]. Data for aggregates production have been built by Chen [2009] with primary data from Martaud [2008] and Ecoinvent database [Kellenberger and Althaus 2003]. Data for sodium powder manufacture have been calculated with the original system boundary of Althaus et al. [2007] and data for sodium silicate solution come from Fawer et al. [1999]. For fly ash and blast furnace slags, a distinction is made between the production of both products and by-products (Iron industry, coal power plants), and the specific treatments made on the by-products for their introduction in concrete. Inputs and outputs data are from Althaus [2003] for Iron production and Dunlap [2003] for slag treatment, Doka [2005] and Dones [2007] for coal power plants process and Surschiste [2009] for fly ash treatment. Finally data for metakaolin come from Engelhard [2009].

Environmental impact calculation

Environmental impacts were evaluated according to the baseline method of CML01 [Guinnée et al. 2002] that evaluates 10 environmental impacts (abiotic depletion, global warming, ozone layer depletion, fresh and marine water ecotoxicity, terrestrial ecotoxicity, human toxicity, eutrophication, acidification and photochemical oxidation).

No classification in order to assign inventory results to impact categories has been done. This can introduce potential double counting and magnify the impacts of a particular burden [Reap et al. 2008]. However, these classifications need a spatial differentiation [Finnveden and Nilsson 2005], which is difficult in all inclusive studies. A site-generic impact modelling where all sources are considered to contribute to the same generic receiving environment has then been chosen [Guinnée et al. 2002]. Furthermore an impact allocation has been tested to evaluate the environmental impacts of by-product such as fly ash and blast furnace. Actually, they are usually considered as wastes and their environmental impacts are therefore only associated with the specific treatments needed for their use in concrete (grinding, drying and stock). However, in France and more generally in Western Europe, this vision can be questioned as most of the slags produced are used in cement and that the mass of fly ashes used in concrete is equal to 130% of the present mass production. In other words, it seems natural to question whether or not these products can still be considered as waste. If not, a percentage of the impacts from primary production process (Blast furnace, coal power station) have to be affected to the by-products [ISO 2006]. This allocation problem has been raised by numerous studies [Ekvall and Finnveden 2001; Weidema 2001; Chen et al. 2009] and can not be detailed here. The allocation method that has been chosen in this study is an allocation by economic value, which means that the relative economic value of product and by-product is chosen to evaluate percentage of the *primary production process* impact that has to be affected to the by-product.

RESULTS

Environmental profile of Fly ash based geopolymer concrete compared to OPC concrete

From the 28 mix designs of fly ash based geopolymer concrete [Lee and Van Deventer 2002; Hardjito and Rangan 2005; Pacheco-Torgal et al. 2005; Sofi, et al. 2007] the mix design in **table 1** represents a mean value. A concrete made with OPC that has the same strength resistance contains 415 kg/m^3 of CEM I. The environmental impacts of this mix design are presented in **table 2**.

Table 1. Mix Design of a Classic Fly Ash Based Geopolymer Concrete

Constituent (kg/m^3)	Fly ash	Sodium hydroxide powder	Sodium silicate solution	Sand	Gravel	Compressive strength (Mpa)
geopolymer concrete mix design	477	13	120	554	1294	57

In the **table 2** it is clear that sodium silicate solution produces the main impacts for the geopolymer concrete. Note that this simulation is made with no allocation for fly ash. The results, as shown later will be different when an economic allocation is applied.

Table 2. Environmental Impacts of a Classic Fly Ash Based Geopolymer Concrete

Impact category	Sodium hydroxide powder	Sodium silicate solution	Aggregates	Fly ash	Geopolymer concrete	OPC concrete
Abiotic depletion (kg Sb eq.)	$3.6\ 10^{-2}$	0.8	$4.7\ 10^{-2}$	$9.6\ 10^{-2}$	$9.8\ 10^{-1}$	$6.6\ 10^{-1}$
Global warming (GWP100) (kg CO_2 eq.)	5.4	$1.3\ 10^{2}$	6.9	3	$1.4\ 10^{2}$	$3.3\ 10^{2}$
Ozone layer depletion (ODP) (kg CFC-11 eq.)	$1.2\ 10^{-7}$	$1.0\ 10^{-5}$	$6.7\ 10^{-7}$	$1.6\ 10^{-6}$	$1.2\ 10^{-5}$	$9.4\ 10^{-6}$
Human toxicity (kg 1.4-DB eq.)	3.1	83	4.6	$7.5\ 10^{-1}$	91.4	20
Fresh water aquatic ecotox. (kg 1.4-DB eq.)	$5.8\ 10^{-1}$	24	1	$8.4\ 10^{-2}$	25.3	2.6
Marine aquatic ecotoxicity (kg 1.4-DB eq.)	$3.4\ 10^{3}$	$5.8\ 10^{4}$	$2.8\ 10^{3}$	$9.2\ 10^{2}$	$6.5\ 10^{4}$	$1.0\ 10^{4}$
Terrestrial ecotoxicity (kg 1.4-DB eq.)	$1.6\ 10^{-2}$	$8.7\ 10^{-1}$	$4.3\ 10^{-2}$	$8.0\ 10^{-3}$	$9.4\ 10^{-1}$	$4.9\ 10^{-1}$
Photochemical oxidation (kg C_2H_4 eq.)	$1.7\ 10^{-3}$	$2.5\ 10^{-2}$	$1.6\ 10^{-3}$	$9.2\ 10^{-4}$	$2.9\ 10^{-2}$	$1.8\ 10^{-2}$
Acidification (kg SO_2 eq.)	$5.0\ 10^{-2}$	0.6	$3.9\ 10^{-2}$	$1.6\ 10^{-2}$	$7.0\ 10^{-1}$	$4.8\ 10^{-1}$
Eutrophication (kg PO_4^{2-} eq.)	$6.4\ 10^{-3}$	$5.5\ 10^{-2}$	$7.0\ 10^{-3}$	$2.4\ 10^{-3}$	$7.1\ 10^{-2}$	$7.3\ 10^{-2}$

When this geopolymer concrete is compared with hydraulic cement based concrete, it is evident that this new type of binder allows for for strong reuction of the global warming potential. From 330 kg of equivalent CO_2 per m^3 for OPC based concrete, the geopolymer concrete releases only 144 kg of equivalent CO_2 per m^3, which represents a saving of 60%. It is interesting to note that even with no allocation on fly ash production, this type of geopolymer concrete can not achieve the factor 4 objective for the concrete industry, and therefore, fly ash based geopolymer concrete as they are manufactured today do not represent the breaktrough technology that will allow the concrete industry to reduce CO_2 emissions by a factor 4.

Concerning the other environmental impacts geopolymer concrete, due to the use of sodium silicate solution have always higher impacts that OPC concrete as shown in **figure 2**. The economic allocation on fly ash does not change the main pattern but slightly increases all impact categories. Therefore this study highlights the fact that using geopolymer concrete to reduce CO_2 emission in the building sector will increase environmental impacts for other aspects that are not currently prioritary impact categories but that were before important (Ozone layer destruction, Acid rains) or that could become important in the future. The use of sodium silicate solution in concrete to substitute clinker based concrete can then induce a pollution transfer from Global warming considerations towards all the other environmental impacts.

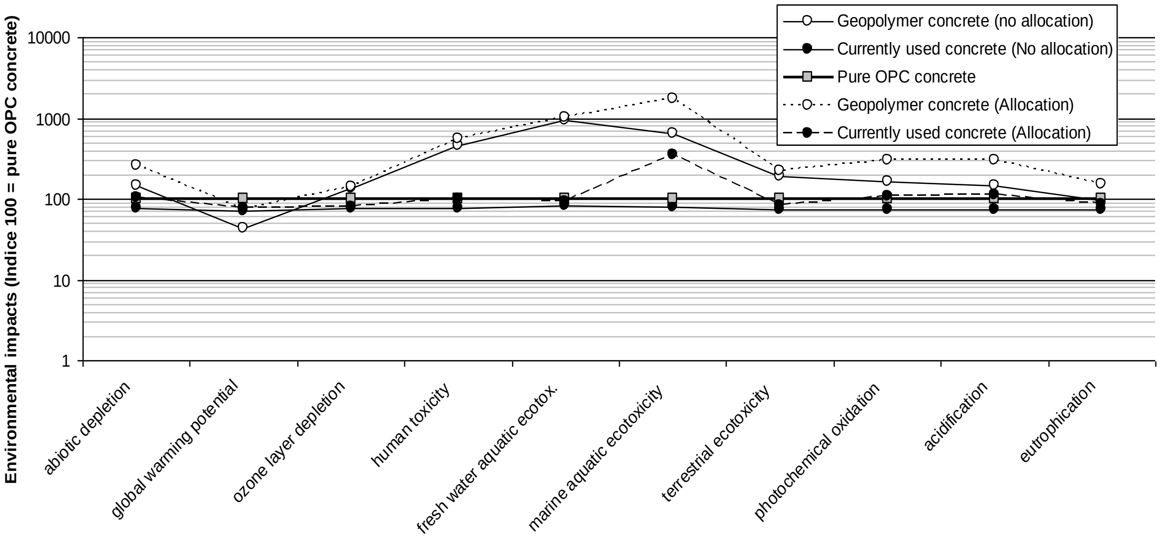

Fig 2. Eco-Profile of Fly Ash Based Geopolymer Concrete Compared to OPC Concrete.

The pure OPC concrete binder is made exclusively with CEM I whereas the standard concrete binder is made with 70% CEM I and 30% fly ash. Two calculation methods are shown: 1) fly ashes are considered as a waste, and no allocation is done; 2) fly ashes are considered as a by-product, and an economic allocation is used.

Environmental impact of geopolymer concrete made with Fly ash, Slag and metakaolin

28, 2 and 10 geopolymer concretes made respectively with fly ash (FA), blast furnace slags (BFSG) and metakaolin (MK) have been studied [Bakharev et al. 2001; Lee and Van Deventer 2002; Hardjito and Rangan 2005; Pacheco-Torgal et al. 2005; Sofi, et al. 2007; Silva et al. 2007). The environmental impacts of these concretes can be calculated and for each type of geopolymer concrete a mean environmental impact and a standard deviation can be calculated. All these concretes have been compared to an OPC concrete with the same strength resistance. When the impact for OPC concrete is set at 100% geopolymer concretes can be compared in percentage variation. The results of the impact for the global warming indicator are presented in **figure 3**. From these results it is clear that FA and BFSG based geopolymer concretes have lower impacts than MK based geopolymer concretes. **Figure 3** shows also that the fact to consider FA and BFSG as by products from coal power and iron industry respectively and therefore perform an economic allocation, increases the impacts of geopolymer concretes. As a result geopolymer concretes made with by-products from other industries or with MK do not represent a significant CO_2 emission reduction that would allow the achievement of the factor 4 objectives.

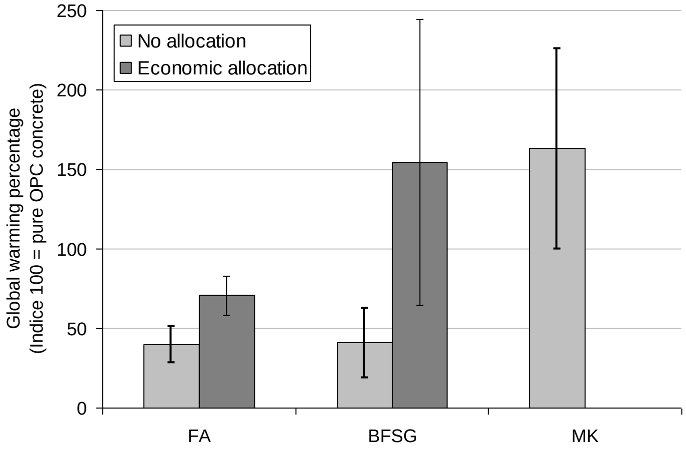

Fig 3. Global Warming Potential of Geopolymer Concretes Compared with OPC Concrete. The OPC global warming impact is set at 100% and geopolymer concrete values are expressed in percentage variation.

DISCUSSION AND PERSPECTIVES

In this study it has been shown that FA and BFSG based geopolymer concrete have low CO_2 emissions compared to OPC concrete but still do not achieve factor 4 objectives. It has also been shown that the sodium silicate solution used represents the main impacts for geopolymer concrete and induces a pollution transfer to all other environmental impact categories. It seems, therefore, that a way to achieve the CO_2 reduction objective in the concrete industry with geopolymer concrete and still do not induce pollution transfer is: 1) to use true waste from industry that do not have allocation impacts or to use thermally activated clays such as metakaolin that have low impacts for the production; 2) to drastically reduce the use of sodium silicate. This can be done by substituting sodium silicate solution by sodic slags. This has been already been developed in the Geocistem program [1997] and further works should be done in this field.

CONCLUSION

In this study, a detailed environmental evaluation of geopolymer concrete has been done using the Life Cycle Impact Assessment method. Except for global warming, geopolymer concretes have higher impacts than hydraulic concrete due to the important impacts of sodium silicate solution. For the global warming impacts, this study has shown that FA and BFSG based geopolymer have lower impact than MK based geopolymer. However, when impacts of the production of FA and BFSG is included as by-products from iron and electric industry, these geopolymers have a global warming potential similar than usual concrete. Furthermore, even without this allocation step, these geopolymer concretes do not allow us to reach the IPCC objective expressed as a diminution of a factor 4 from 1990 emissions.

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