

Recent update on the environmental impact of geopolymers

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Abstract

The contribution of building materials' production to environmental impacts becomes significant in the new energy efficient buildings that are currently built. Among those materials, cement represents a major part of the embodied carbon footprint of buildings. Development of alternatives to traditional Portland cement have emerged over the last decades; however, their environmental assessment over their life cycle have resulted in conflicting results, which give a confusing picture of the strategic path to be followed. This paper focuses on the alkali-activated cement and concrete and points out the variability linked with the choice of energy source for the production of the activators and precursors. A review of existing studies is then performed in order to highlight the main benefits of these alternative cements in terms of reduction of greenhouse gas emissions. Finally, the results presented here highlight further research trends for new cements and concrete.

Keywords: Concrete; LCA; Geopolymer; Alternative Cement; CO₂

1 Introduction

Concrete production has an impact on the climate as it accounts for 5-8% of total anthropogenic CO₂ emissions [1]. 95% of this CO₂ is produced during the fabrication of cement, half of it being released by the decarbonation of the limestone during cement fabrication. Furthermore, the rapid urban development in emerging countries will push forward the cement demand and recent studies estimate that cement production could represent 10 to 15% of global CO₂ emissions by 2020 [2]. Thus, there is an urgent need to reduce the carbon footprint of cement and develop more environmentally friendly, economically viable and socially relevant cements.

The cement industry has already achieved impressive improvement in terms of energy efficiency and clinker substitution by supplementary cementitious materials [3]. Furthermore, due to a better understanding of packing optimization [4, 5] as well as the development of super-plasticizers, reduction of cement content at the concrete scale while achieving similar performance can now be done [6]. Finally, structural optimization, either by better design through structurally informed form-finding software [7] or by high performance material used in smaller volume than low performance concrete [8], can further reduce the need for cement production when the structure scale is considered [9]. However, all these initiatives are not sufficient to achieve the Intergovernmental Panel on Climate Change (IPCC) recommendations, due to the tremendous growth of consumption, especially in emerging countries.

Overall, it can be expected that these technological improvements will reduce the CO₂ emissions by a factor of 2 [10, 11], which is far from the factor 4 reduction objectives of IPCC. Alternative cements are therefore crucially needed.

Calcium sulfoaluminate cement (CSA) requires a lower kiln temperature between 1250°C and 1350°C temperature as compared to 1450°C for clinker production, which slightly reduces CO₂ emissions [12]. Alkali-activated cements (AAC) are another promising alternative. AAC are binders formed by the alkali activation of alumino-silicate sources, which can be natural or synthetic materials or an industrial waste [13]. They were originally called geopolymers to account for the reaction between alumino-silicate oxides and alkali polysilicates yielding polymeric tridimensional Si-O-Al bonds network at room temperature [14] but this specific material belongs to a broader binder categories defined as AAC in the recent book published by Provis and Van Deventer [15] as well as two recent RILEM Technical Committees [16, 17]. To compare the environmental impact of these alternatives to a standard concrete, life cycle assessment (LCA) is the most appropriate method [18]. However, published results on LCA of AAC give conflicting results. Recently, Davidovits made a comprehensive review on environmental impact of geopolymers and showed how variations in data for geopolymer components influence largely the final impacts [19]. The objective of this article is hence to provide an additional understanding of this raw data variability question as well as to propose more specifically a method for comparing concrete with similar functional unit. Finally, the results presented here highlight further research trends for new cements and concrete.

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2 The environmental impact of the components

Most of the AAC are formed by activating an aluminosilicate powder (precursor) with an alkaline hydroxide and/or silicate solution (activator). Over the last 10 years, different studies have been conducted on the environmental assessment of geopolymers and AAC [19-26]. Most of the studies are done from cradle to gate, limiting their system boundaries to the production of components and the appropriate mix design, sometimes including the transport of the components. One study considers a wider system, up to construction site, including the mixing of concrete, its curing and placement on site [24]. This study confirms what is known for conventional concrete production, namely that the main impacts of concrete over its life cycle are concentrated on the production of concrete components, except for waste related impacts driven by end of life choices. Therefore, it is a priority to focus on having impacts for components, which are accurate and shared among the different studies.

2.1 Precursors

Among the various precursors used for AAC, we decided to distinguish calcined clay precursors, which have been mainly used for geopolymer fabrication [27] and industrial waste precursors, which are widely used in the broader alkali activated material family [13].

Metakaolin

Calcined kaolinite, so called metakaolin, is the most widely used clay-based precursor. This is due to the fact that the activation temperature is lower for tetrahedral-octahedral (T-O) layered clays such as kaolinite than for tetrahedral-octahedral-tetrahedral (T-O-T) layered clays such as illite or smectite, which gives a wider window of opportunity between the temperature of amorphisation and the temperature of recrystallization in non reactive forms such as mullite, feldspars or pyroxene depending on the initial chemical composition of the clay [28]. Furthermore, NMR studies have shown that it is only with kaolinite that a 5-coordinated Al can be identified during heating [29]. This metastable Al form increases the reactivity of the calcined clay. In terms of environmental impacts, conflicting values have been highlighted recently by Heath and co-authors [26]. They pointed out that values ranging from 435 g CO_{2-eq}/kg to 92.4 g CO_{2-eq}/kg could be found and that this would be due to the fact that the lower values did not consider clay extraction as well as the extra amount of unfired clay that need to be burnt to reach 1kg of metakaolin. The 92.4 g CO_{2-eq} value [28] actually included all these aspects and the low value came from the fact that biogas from agricultural waste was used to calcine the clay, as done in the Argeco production plant in the South of France (Tardy, personal communication, 2010). To better illustrate this higher sensitivity of metakaolin environmental impact to fuel source rather than production processes, we calculated the

global warming potential of metakaolin production considering a wide range of efficiency processes as well as different fuel type. The foreground data are shown in Table 1. They are based on different studies, in particular a detailed study conducted on the metakolin production plant of Argeco [30] as well as the main reference from NLK [31] used in Heath study and a recent case study on calcined clay cement in Cuba [32]. Note that, for the worst case scenario, no waste recovery has been considered bringing the energy consumption to 4100 MJ/t of metakaolin produced. This value is extremely high as half of it is for the drying of the raw material before calcination. A low-grade metakaolin has also been considered [30], where 2.1 t of raw materials are extracted to produce 1 t of metakaolin. The figure 1 shows the results of Monte Carlo simulation (10'000 simulations) performed with Simapro 8, using Ecoinvent v3 and the efficiency parameters of Table 1.

Table 1. Material and energy requirement for metakaolin production. Variability in technology is considered. Data are coming from: ^a[32]; ^b[26]; ^c[30]; ^d[31].

	Average	Min	Max
Mining			
Clay extraction (t)	1.30 ^a	1.16 ^b	2.10 ^c
Clay transport	150 km by truck		
Calcination			
Electricity (MJ)	143.9 ^{a,c}	48.6 ^a	239.2 ^c
Heating (MJ)	2997.3 ^{c,d}	1894.7 ^c	4100 ^d

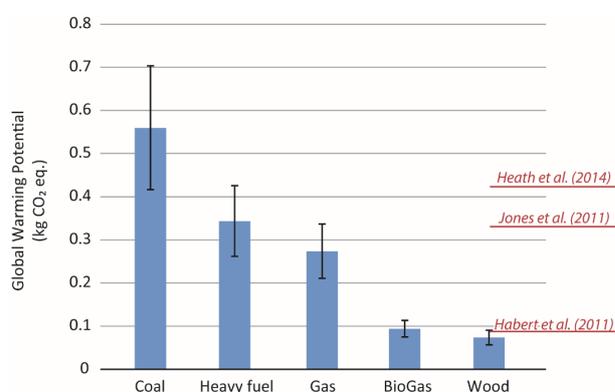


Figure 1. Global Warming Potential in kg CO_{2-eq} for the production of 1 kg of metakaolin depending on the type of fuel used for calcination. Variability in technology is shown through error bars showing 2.5 % and 95 % results of Monte Carlo simulation.

The results show clearly the origin of metakaolin impact variability. The efficiency of the process has much less influence than the choice of the heating source and the main difference between the previous studies came most probably from a choice of fossil-based or bio-based energy source. For other clay types such as illite or bentonite, higher energy consumption will be required as the calcination temperatures are higher (800 to 850°C) [33]. Furthermore, for swelling clays, the water present in the inter-layer induces additional energy consumption for drying as well as a higher amount of initial unfired clay in order to have 1 kg of material after calcination [26].

Industrial waste

Another type of aluminosilicate precursor is waste from other industrial sectors. Among them, fly ash (FA) resulting from coal power plants and granulated blast furnace slags (GBFS) resulting from crude iron production in a blast furnace are the most widely used. This option allows for a significant, both environmental and economic cost reduction in the AAC manufacturing process by saving burning costs of clay-based precursors. However, this interesting alternative is raising an accounting problem in the LCA method. Actually, according to ISO standards used in LCA [18], when a production system produces more than one product, it is necessary to attribute an environmental burden to each product. This is the case for industrial waste such as FA and GBFS, which are, by definition, not the main products of the industrial plants, but by-products. This question of the value of the environmental load has been emphasized by many authors as it concerns the AAC production as well as the main cement industry practices where clinker is substituted by supplementary cementitious materials [34, 35]. The current status would be to consider these precursors as waste and to allocate no impacts from the main process and only the impacts from the processes, which are directly linked to their transformation from a waste to a valuable precursor. For instance for GBFS, the quenching of the slag to increase the reactivity, as well as grinding, drying, storage and transport will be included. However, this situation is evolving, particularly in Europe where a European Union directive [36] considers a possibility for an “end of waste” status.

“A waste may be regarded as by-product if the following conditions are met:

- Condition a) further use of the substance or object is certain;*
- Condition b) the substance or object is produced as an integral part of a production process;*
- Condition c) the substance or object can be used directly without any further processing other than normal industrial practice;*
- Condition d) further use is lawful, i.e. the substance or object fulfills all relevant product, environmental and health protection requirements for the specific use and will not lead to overall adverse environmental or human health impacts.”*

This directive is very relevant for GBFS and FA and these two materials can thus no longer be considered as waste but instead as by-products in Europe.

Different scenarios can be promoted to calculate the environmental profiles of the by-products. A mass allocation where impacts are divided by the relative mass values of the different products (steel/slugs) induces a very high environmental load for by-products and a division in relation to the relative economic value of the products seems more in accordance with the perception of what could be the environmental load of these precursors [34]. Actually, the recent Environmental Product Declaration (EPD) for the CEM III cement of CEMBUREAU took an economic allocation

for FA and GBFS [37]. However, they considered that, as the contribution of these by-products to the overall revenue of steel or electricity production is very low (<1 %), the environmental impacts from the joint process on the co-product can be neglected in the cement LCA. The subsequent processes (e.g. granulation and grinding of blast furnace slag) are still entirely allocated to the co-products in this EPD. This assumption of neglecting the allocation because it represents less than 1 % of the economic value is problematic and not appropriate because Van den Heede and de Belie showed that with 2 % of steel production allocation on GBFS, the CO₂ emissions were already of 0.13 kg CO_{2-eq}/kg GBFS while 1 % of allocation of electricity production induced 0.19 kg CO_{2-eq}/kg FA [35]. This very small economic contribution can actually not be considered as negligible because the comparison is not between the impact of the main product and the co-product, but between the co-product and the clinker that it will replace. For instance 1 % of the 2 t of CO₂ released by the production of 1 t of crude iron (Ecoinvent v3) need to be allocated to the 0.25 t of GBFS produced at the same time as the 1 t of crude iron. It leads to 0.08 t CO_{2-eq}/t GBFS (10% of clinker CO₂ emissions) with only 1% of economic allocation. As a consequence, economic allocation needs to be considered and implemented even with 1 % of revenue associated. The main problem with the economic allocation is the question of price variability as illustrated in Habert [38]. However, using global revenue from the industry, as done in the ECRA's EPD [37], and not a price per ton can help to reduce the variability.

2.2 Activators

The activators used for AAC are mainly sodium or potassium hydroxide and silicate and in some mixes sodium carbonate. In the recent paper by Heath et al. [26], values are provided per kg and per mole, which shows that the Global Warming Potential (GWP) per mole is similar for all sodium silicate solutions and that sodium hydroxide has a lower impact than sodium carbonate, both being still more than two times lower than silicate options. The new version 3 of the Ecoinvent database provides environmental impact data for alkaline solutions per kg of the dry component (i.e. without the water). These values are presented in Table 2. They show that, as for metakaolin, the choice of the energy source has a strong influence. For instance, the same diaphragm cell production process for sodium hydroxide induces a GWP of 0.991 kg CO_{2-eq} per kg of sodium hydroxide in Europe while the impact for non-European countries is equal to 1.434 kg CO_{2-eq}/kg. A recent study on LCA of sodium hydroxide production in China showed comparable results (1.59 kg CO_{2-eq}/kg) and highlight the sensitivity of the result to the choice of electricity mix [39]. On the contrary, the differences between production type (diaphragm cell, membrane cell or mercury cell) have smaller influences (table 2). Concerning sodium silicate solution, the values of ecoinvent version 3 are still based on the initial Fawer study [40] except that, differing from version 2, the exclusion of

the water considerably reduces the environmental impact as already noticed by Davidovits [19]. All these data are presented with CML calculation method in order to allow a comparison with previous studies [22].

3 The environmental impact of geopolimer mixes

3.1 A common comparison method

With the above presented data on precursors and activators, it is possible to calculate the environmental profile of AAC fabrication. However, if we want to compare this value with cementitious materials, a single functional unit is required for the two products. As AAC and cement-based concrete share, most of the time, the same type of aggregates, we can therefore focus on the binder comparison of both concretes and find a reliable method to calculate the amount of cement necessary to provide the same strength as the alkali activated binder.

In cement based concrete technology, it is known that the ingredients for a bad concrete are exactly the same as those required for a good concrete and that it is the relative proportions that matter [41]. Furthermore, since Féret or Bolomey, it has been established that the compressive strength depends mainly on the water to cement ratio (w/c) and not so much on the specific amount of cement used in one cubic meter [42]. In the following presentation, the Féret equation will be used, but similar concepts could be developed with Bolomey or similar equations. The advantage of the Féret equation is that the mathematical form can be physically justified [4]. The Féret equation relates the cement content to the compressive strength:

$$f_c \approx KR_{C_{28}} \left(\frac{V_{cement}}{V_{paste}} \right)^2 \tag{1}$$

where f_c is the compressive strength, K is a parameter that characterizes the aggregates quality, $R_{C_{28}}$ is the specific mechanical strength of cement, V_{cement} is the volume of cement and V_{paste} is the volume of paste which includes air, water and cement.

Table 2. Environmental impacts of the different activators (1 kg). CML IA method is used on Ecoinvent v3.

Environmental impact categories	Sodium hydroxide, without water, in 50% solution state, diaphragm cell (Europe)	Sodium hydroxide, without water, in 50% solution state, membrane cell (Europe)	Sodium hydroxide, without water, in 50% solution state, mercury cell (Europe)	Sodium hydroxide, without water, in 50% solution state, diaphragm cell (Global)	Sodium hydroxide, without water, in 50% solution state, membrane cell (Global)	Sodium hydroxide, without water, in 50% solution state, mercury cell (Global)	Sodium silicate, without water, in 37% solution state, furnace liquor (Europe)	Sodium silicate, without water, in 37% solution state, furnace liquor (Global)
Abiotic depletion (kg Sb _{eq})	5.69E-06	5.72E-06	5.70E-06	5.66E-06	5.70E-06	5.67E-06	1.06E-05	1.05E-05
Abiotic depletion (fossil fuels) (MJ)	1.23E+01	1.09E+01	1.22E+01	1.66E+01	1.46E+01	1.65E+01	1.13E+01	1.08E+01
Global warming (GWP100a) (kg CO ₂ _{eq})	9.91E-01	8.86E-01	9.89E-01	1.43E+00	1.26E+00	1.43E+00	1.08E+00	1.06E+00
Ozone layer depletion (ODP) (kg CFC-11 _{eq})	7.84E-07	7.81E-07	7.85E-07	7.68E-07	7.69E-07	7.69E-07	8.26E-08	7.47E-08
Human toxicity (kg 1,4-DB _{eq})	6.30E-01	5.89E-01	6.35E-01	6.65E-01	6.19E-01	6.69E-01	7.33E-01	7.35E-01
Fresh water aquatic ecotox. (kg 1,4-DB _{eq})	6.07E-01	5.47E-01	6.07E-01	5.20E-01	4.77E-01	5.20E-01	5.11E-01	5.11E-01
Marine aquatic ecotoxicity (kg 1,4-DB _{eq})	1.69E+03	1.51E+03	1.69E+03	1.97E+03	1.76E+03	1.97E+03	1.16E+03	1.17E+03
Terrestrial ecotoxicity (kg 1,4-DB _{eq})	3.03E-03	2.77E-03	2.64E-02	2.85E-03	2.63E-03	2.62E-02	1.81E-03	1.80E-03
Photochemical oxidation (kg C ₂ H ₄ _{eq})	2.24E-04	2.06E-04	2.24E-04	4.19E-04	3.70E-04	4.18E-04	2.74E-04	2.79E-04
Acidification (kg SO ₂ _{eq})	5.15E-03	4.73E-03	5.14E-03	1.03E-02	9.05E-03	1.02E-02	6.20E-03	6.32E-03
Eutrophication (kg PO ₄ ²⁻ _{eq})	5.92E-03	5.10E-03	5.89E-03	2.77E-03	2.50E-03	2.76E-03	1.73E-03	1.72E-03

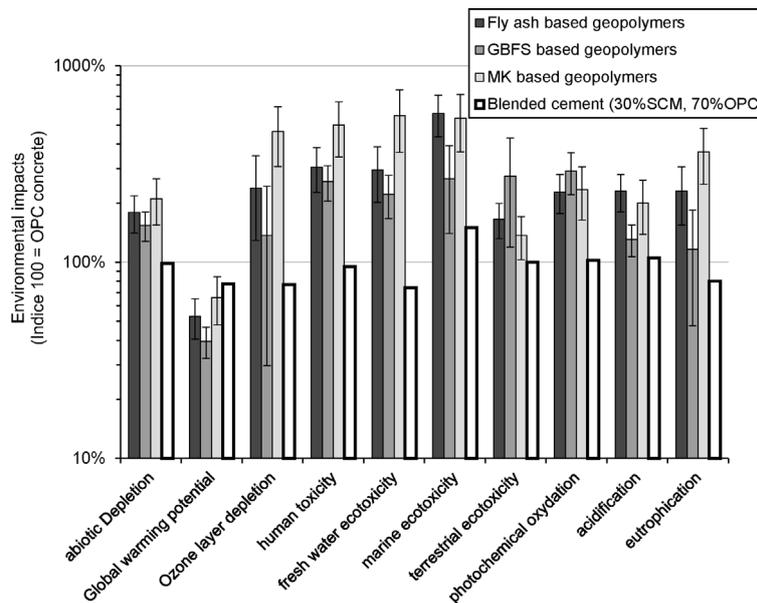


Figure 2. Environmental impact of sodium silicate activator based geopolimer with different precursors.

Secondary order parameters have been established, such as the maximum paste thickness [4], which is a key parameter for high performance concrete, but is more difficult to identify for regular concrete [43]. When the Féret equation is used to calculate the appropriate amount of cement that will provide the same strength as the studied AAC, it is interesting to note that the main unknown parameter in addition to the volume of cement is the volume of paste; which means that, if we consider the same amount of binder paste in both concretes (i.e. ordinary Portland cement and alkali activated concrete), we can directly define one single solution for the amount of cement. This method allows then to compare two concretes with a same efficiency for both pastes.

Actually, we can use the compressive strength f_c and the paste volume of the geopolymer concrete. Furthermore, we need to calculate appropriate values for K , which is related with the quality of the aggregate and can be calculated with a cement based concrete using similar aggregates as the alternative concrete but without having necessarily the same compressive strength. This concrete mix is just used to calibrate the K value [44].

3.2 Conventional geopolymers

Geopolymer concrete have been compared with ordinary Portland cement (OPC) concrete considering the same paste volume in the previous papers of Habert et al. [22, 44]. Other papers on environmental impact of AAC compares concrete with similar strength but that do not always have the same amount of aggregates, which induce a bias in the results. This lead for instance Yang et al. [25] to compare concrete with OPC, concrete with blended cement and concrete with alkali activated cement and to conclude that the concrete with blended cement is the worst. However, it comes mainly from the fact that more cement was used while keeping the same water/cement ratio.

Among the papers that compare concretes with similar paste content, there is a consensus on the fact that AAC have a lower GWP than OPC. The amount of improvement is variable. Turner and Collins reported a 10 % improvement compared to OPC [24]. Habert and co-authors had a 45 % improvement as average value for FA-based AAC, more than 80 % for average value for GBFS-based AAC [22]. MK-based geopolymer provided however no improvement. Finally, Weil et al. results are in accordance with 70 % improvement compared to OPC concerning GWP [21]. This is close to the aspirational values presented by Van Devanter et al. [45]. For the other impacts categories, it seems less clear. Results from Weil et al. [21], Habert et al. [22] and recently Heath et al. [26] show no clear improvement.

The release of the new Ecoinvent version 3 where kg of activating solution are clearly calculated without water pushes to recalculate previous data that were calculated with a too high value for the sodium silicate solution, as Davidovits mentioned in his recent review [19]. The updated results, with a sodium silicate solution impacts divided by

nearly 3 as well as the updated CML calculation method for all other components are shown in figure 2. Concerning the industrial waste precursors, they have been considered with an economic allocation with the same assumption of by-product from other industries as in Habert et al. [22].

These results improve the comparison with cementitious materials. In particular MK-based geopolymer have now similar GWP as blended cement based concrete. However, the trend is not drastically changed and confirms early results. AAC provides a clear alternative solution in terms of reduction of greenhouse gas emissions. For the other environmental impact categories, benefits are less clear. It is worth noting that among the main issues recently identified to have a safe operating planet, climate change is the only one that is related to the construction sector. The other ones such as nitrogen cycle and biodiversity loss are more related to non-sustainable agricultural practices [46]. As a consequence, the fact to have a ground-breaking building material in terms of greenhouse gas emission is highly valuable and should pave the path toward a low carbon built environment. Actually, some of the studied AAC allowed to reduce the GWP by a factor of 4 compared to OPC, which is the goal that need to be achieved to keep the global temperature below 2°C, e.g. [47].

3.3 Future perspectives

In this section, we would like to conclude with alternative mixes that push the alkali activation concept further. The first type of alternative to alkali-activated binders is the so-called hybrid cements. The main underlying concept is that the clinker used in these binders provides a first and fast reaction that allows early strength but also provides heat to help the activation of fly ash and to hasten the polymerization reaction. To assess the environmental impact of these binders, we used the mixes detailed in [48] and the adapted equation from de Larrard for cement paste calculation further detailed in [44]

The GWP of the studied hybrid cement is 70% lower than of an OPC binder. This result is in the same range as the current AAC. However, in terms of implementation in the market, the presence of a small amount of clinker might facilitate the adaptation of standards as well as the acceptance from the construction sector. This technique of hybrid cement appears then to be an interesting bridge between the classic clinker substitution technology and the still emerging alkali-activation technology.

Another promising initiative is the so-called “one part geopolymer” where a manufactured slag with high alkali content is used as activator of a blast furnace/fly ash precursors. It is known that alkali alumino-silicate melts have a low melting temperature. Albite melt ($\text{NaAlSi}_3\text{O}_8$) has the lowest one at only 700°C, compared to 1200°C for feldspathic melt with Ca ($\text{CaAl}_2\text{Si}_2\text{O}_8$). As a consequence, making a slag of albite and sodium hydroxide requires much less energy than producing a Ca-alumino-silicate slag. This sodic alumino silicate slag provides a highly reactive solid

product which rapidly releases alkali, Si and Al in the solution. This argument is explained in [49] and more detailed example can be found in an early patent [50]. The first example from the patent, where only 5 % of alkali- and thermally- activated polymerization aid was needed, is presented in Table 3. To calculate the environmental impact associated with the production of this polymerization aid, we have converted the given mineral composition of the potash feldspar to 69.4 % orthoclase, 29.6 % albite and 1.0 % anorthite. The specific heat capacities were 628 J/kg/K for orthoclase, 730 J/kg/K for albite and 711 J/kg/K for anorthite [51] and a latent heat between 250 and 450 kJ/kg was taken for the different feldspars [52]. If we consider 20 % heat loss in the process, our calculation shows that for 1 m³ of one part geopolymer, 38 MJ of energy would be needed. Using natural gas in the furnace would release 2.6 kg CO_{2-eq}. The one-part geopolymer is used as the paste and aggregate will be added to make concrete. The modified Féret equation for cement paste strength calculation is used [4].

Table 3. Example of a one-part geopolymer and its cement equivalent, data from [46].

	Mass (kg)	GWP (kg CO _{2-eq})	
		No allocation	Economic allocation
Fly Ash	621	3	118
GBFS	621	10	81
Potash feldspar	41	0.1	0.1
Sodium hydroxide	25	23	23
Free water	476	0.1	0.1
Energy to activate K-Feldspar: 38 MJ		2.6	2.6
Total GWP		39	224
Cement equivalent (OPC)	1168	1016	1016
GWP comparison with OPC		4%	22%

As a consequence, the one part geopolymer cement contributes to less than 5 % of the GWP of cement made with 100 % OPC (when no allocation on GBFS and FA is considered) and allow to achieve 80 % reduction even with an economic allocation. A very recent application of one part geopolymer in a four storey building was done at the University of Queensland by Wagner company [53]. The precast geopolymers beams were used as a structural floor element and as an architecture feature. They were reinforced with steel, but the later applications were prestressed. These early examples can work as lighthouse projects for the implementation of these one-part geopolymers which are easy to use on site and achieve tremendous reductions in terms of greenhouse gas emissions.

4 Conclusions

In this study, we have been able to confirm that, despite large uncertainties on the exact environmental impact of their different components as well as on the mix design, the new concrete based on alkali-activated cements provide a groundbreaking solution for climate change issues. They could contribute to the overall objective of reducing by a

factor of 4 the CO₂ emissions associated with the concrete production.

Considering the other environmental impact categories, which are indeed less critical for the construction industry, geopolymers and alkali activated cements do not provide benefits compared to the conventional cement industry.

Finally, it should be noted that this study focused only on the environmental impacts associated with the production of the constituents and that durability aspects need to be integrated to assess the environmental impacts over the life cycle of the concrete. The current RILEM TC 247-DTA will help to provide answers on this key point.

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