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False Values on CO_2 Emission for Geopolymer Cement/Concrete published in Scientific Papers

Joseph Davidovits

Adapted from the article originally published in Elsevier's internet site *materialstoday* at http://www.materialstoday.com/polymers-soft-materials/features/environmental-implications-of-geopolymers/, 29 June 2015, titled *Environmental implications of Geopolymers*. See also the presentation at the Geopolymer Camp 2015.

Introduction

My first study on the environmental impact (LCA), with respect to Global Warming Potential (GWP) related to the CO_2 emission comparison between Portland cement manufacture and geopolymer cement started as early as 1990, at PennState Materials Research Laboratory, Pennstate University, USA. Unfortunately, American Agencies (DOE and EPA) stated that this was not an important issue and both institutions declined to support research proposals. The theoretical studies were presented at several conferences, Davidovits (1993, 1994). Ordinary Portland cement is a serious atmospheric pollutant. Our studies had shown that approximately 0.85 to 1.0 tonnes of CO_2 are released into the atmosphere for every tonne of Portland cement which is made anywhere in the world.

The Portland cement industry reacted strongly to our publications by lobbying the legal institutions who delivered CO_2 emission numbers, which did not include the part related to calcium carbonate decomposition (0.55t), focusing only on combustion emission (0.45t). In 1997, UN's Intergovernmental Panel on Climate Change put the industry's total contribution to CO_2 emissions at 2.4%; the Carbon Dioxide Information Analysis Center at the Oak Ridge National Laboratory in Tennessee quoted 2.6 %, instead of including both sources: energy (representing 45% of the CO_2 emission) and decarbonation of calcium carbonate (the main raw material) for 55%, totalizing 5,80 % of [world] 1997 CO_2 emission. Eighteen years later, the situation has worsened with Portland cement CO_2 emissions approaching 3 billion tonnes a year (Hasanbeigi et al. 2012).

In 2002 at the World Climate Congress in Rio de Janeiro, statistics integrated the actual values and the Portland cement industry started introducing so-called "blended cements", involving the addition of mineral ingredients such as coal fly ash, so that today, 2015, the reduction is in the range of 20% of initial value of 1 tonne CO_2 emitted in the production of 1 tonne of Portland clinker.

LCA of commercialised geopolymer cement/concretes are seldom. This is due to proprietary reasons. Presently they are based on Type 2 slag/fly ash/alkali-silicate system (see Technical papers #21, #22, #23 in the Library). Geopolymer Type 2 concrete and standard Portland concrete are similar in non-binder materials used and behaviour after production; there is some dilution of the benefits when measured over the full life cycle (LCA). The greenhouse gas emissions during the life cycle of Geopolymer Type 2 concrete are approximately 62%-66% lower than emissions from the reference concrete. The Type 2 geopolymer cement has ca. 80% lower embodied greenhouse gas intensity than an equivalent amount of ordinary Portland cement binder used in reference concrete of a similar strength, confirming the data published by the Geopolymer Institute, where the reductions are in the range of 70 % to 90 % (see Technical paper #21). These values do not include any additional external constraints like transport from or to the utility. They reflect the actual potential as soon as industrialization starts in full swing. On the opposite, some published scientific LCA papers claim that, in terms of CO_2 emission, geopolymer cement was not better than Portland cement, and worse for other parameters. These statements are based on methodological errors and false calculations of the CO_2 emission values for geopolymer cement/concrete. The problem is that these false values are taken for granted by other scientists without any further consideration.

The Manufacture of Geopolymer Cement/Concrete

Geopolymer cements are manufactured in a different manner than that of Portland cement. They do not require extreme high temperature kilns, with a large expenditure of fuel, nor do they require such a large capital investment in plant and equipment. Naturally occurring alumino-silicates (geological resources available on all continents) are providing suitable geopolymeric raw materials.

In the past, commercial geopolymer cement called either Pyrament or Geopolymite 50 (Davidovits, 1991) had ratios MK-750/slag/K-silicate being 1/1/2 respectively. Due to the high amount of K-silicate, this first geopolymer cement can no longer be proposed as a valuable competitor for Portland cement because it is too expensive. It is exclusively marketed for special niche applications. In addition, as will be discussed below, the high amount of alkali-silicate is not appropriate with respect to the Global Warning Potential in the environmental impact assessment LCA.

Therefore, as early as 1993, we set our effort in R&D projects dedicated to the strong reduction of the alkali-silicate amount for concrete and building applications, resulting in CO_2 emission reduction up to 80% when compared with Portland cement.

The geopolymer cement of the second generation is coined "*Rock-based geopolymer cement*". The manufacture includes the components with the ratios MK-750/slag/volcanic tuff/ alkali silicate being 1/1/2/1. Compared with the first generation, the amount of alkali-silicate solution is reduced from 50% by weight to 20% by weight. A more competitive geopolymer cement with lower CO_2 is obtained, according to the Davidovits patent (2003) when instead of making a mixture of MK-750 and feldspathic-volcanic rock, one uses naturally occurring geological products containing these two elements in-situ. Indeed, kaolinite is the result of the weathering of feldspar and it is naturally found in weathered granitic residual rocks. The weathered residual granitic rock consists of 20 to 40 percent by weight of kaolinite and 80 to 60 percent by weight of feldspathic, and quartzitic residual sand containing reactive silica. In order to have a maximum reactivity, the weathered residual granitic rock in which kaolinization is very advanced, is calcined at a temperature ranging between 650°C and 800°C and, on the one hand, ground at an average grain size of 15-25 microns for the feldspathic and quartzitic parts, the kaolinitic part, on the other hand, having naturally quite a lower particle size. In that case, the make up of this rock-based geopolymer cement comprised the ratios slag/weathered granite/ alkali silicate being 1.5/3.5/1. Compared to the first generation, the amount of alkali-silicate solution is reduced from 50% by weight to 17% by weight.

It must be noted that these rock-based cements have very high mechanical strength, in the range of 100-125 MPa compressive strength at 28 days. It seems obvious that a reduction of the most expensive element, namely alkali-silicate solution, may go down to 10 % for a regular cement/concrete of the type 30-35 MPa. The energy needs and CO_2 emissions calculations for this second generation of geopolymer cement (Davidovits, 2013) are significantly reduced. In the most favourable case — slag availability as waste (no allocation) — there is a reduction of 59% of the energy needs in the manufacture of Rock-based geopolymer cement in comparison with Portland cement. In the least favourable case —slag manufacture (allocation) — the reduction reaches 43%.

As for CO_2 emission, in the most favourable case — slag availability as waste (no allocation) — there is a reduction of 80% of the CO_2 emission during manufacture of Rock-based geopolymer cement in comparison with Portland cement. In the least favourable case —slag manufacture (allocation) — the reduction reaches 70%.

There is a third category of geopolymer cement based on another industrial waste, coal fly ash, essentially low calcium fly ash of class F. As early as 1994, we mentioned the potential for this fly ash-based geopolymer cement. Here is the excerpt of the paper available in the Geopolymer Institute Library, Technical paper #5 *Global Warming Impact on the Cement and Aggregates Industries*: " ... *Development means implementing the use of electricity, on the one hand, and building infrastructures and houses on the other hand; in short, electricity and concrete. The by-product of electricity production with coal firing is fly ash. The innovative step would be to produce electricity and low- CO_2 cement (geopolymeric cement), in the same plant, by adapting and implementing fly-ash production into Geopolymeric raw material, without any supplementary chemical- CO_2 emission. .... , this would allow electricity utilities to produce million tonnes of low- CO_2 fly ash-based Geopolymer cement. .... In other words, implementing such a new technology would give a wide potential for any further development of electricity production with coal firing plants."*

Presently, we have two types based on Class F fly ashes:

- *Type 1: alkali-activated fly ash material:* it uses high caustic NaOH (user-hostile) + fly ash. In general, it requires heat hardening at 60-80°C and is used to directly manufacture fly-ash based concrete, at the laboratory scale.

This alkali-activation of fly ash is very often qualified with the term "geopolymer" which is totally wrong. Despite the proven fact of their dangerousness, their causticity, they are recommended as the "Current State of the Art" in several review papers. Although, during the Geopolymer 2005 Conference in Saint-Quentin, end users' representatives complained about this situation, several scientists do not take this situation into account and continue to promote highly corrosive systems in their alkali-activated-materials. This could explain why their cement technology does not achieve any applications at all.

- *Type 2: slag/fly ash-based geopolymer cement (user-friendly):* it uses the incongruent covalent bonding concept developed in Davidovits' book *Geopolymer Chemistry and Applications*, which allows the fabrication of fly ash-based geopolymer with non-corrosive conditions in a user-friendly system.

One obtains a room-temperature cement hardening with user-friendly silicate solution + blast furnace slag + fly ash. The fly ash particles are embedded in a geopolymeric matrix with Si:Al= 2, (Ca,K)-poly(sialate-siloxo). This material resulted from the EU-sponsored R&D project "*Understanding and mastering coal fired ash geopolymerization process in order to turn potential into profi*t", known under the acronym GEOASH (2004-2007). In this project, since the idea is to use the geopolymer as a cement, the curing is taking place at ambient temperature, with a modified (Ca,K)–based system that does not include MK-750. One finds a dedicated paper in the Geopolymer Institute Library, Technical paper #22: *GEOASH, ambient temp. hardening of fly ash-based geopolymer cements*. Industrial Geopolymer cement/concretes are presently exclusively of this Type 2, see in Technical paper #23

The make up comprises the ratios slag/fly ash/ alkali silicate being 1/5/1. This is for cement developing a compressive strength in the range of 100 MPa at 28 days. Compared with the first generation, the amount of alkali-silicate solution is reduced from 50% by weight to 15-20% by weight. For a lower strength, in the range of 40 MPa, the ratios are 1/8/1, i.e. a reduction of the amount of alkali-silicate solution down to 10-15 % by weight.

The strong reductions in the amount of alkali-silicate (Na and K-silicate) that occurred in the various developments of the geopolymer cement types, since 1985 until 2006 are summarized in Table 1.

Table 1: Evolution of the amount in potassium-silicate % by weight of geopolymeric formulation for room temperature hardening geopolymer cements since 1983-85 (see explanation in the text).

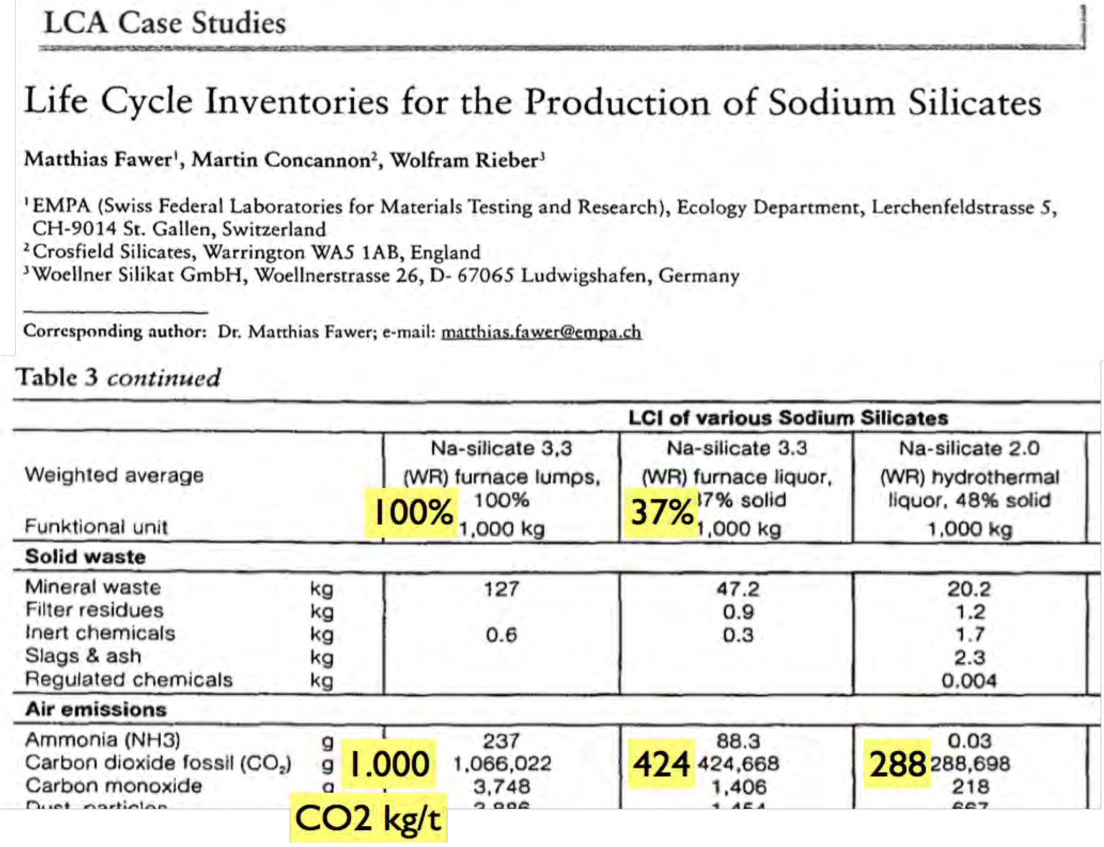
Pyrament (1985)	Geopolymite 50 (1987)	Rock-based (1997) 100 MPa	Rock-based (2002) 50 MPa	Fly Ash-based (2006) 100 MPa	Fly Ash-based (2006) 40 MPa
50 %	50 %	20 %	17 %	14 %	10 %

False published values for Geopolymer cement/concretes CO2 emissions.

Notwithstanding this fact, the majority of published Environmental Impact LCA studies are dealing with alkali-activated materials. They ignore the evolution of the formulations for geopolymer cement since its invention in 1983-85 (see Table 1), simply because it pertains to the industrial and commercial implementation, not to "regular" scientific studies. It is a fact that the scientific community continues to neglect the patent literature.

Actually, the most important document in our study is the LCA paper by Fawer et al. (1999) *Life Cycle Inventories for the Production of Sodium Silicates*, Int. J. LCA 4 (4) 207-212 (1999). Life Cycle Inventories were compiled by EMPA St. Gallen / Switzerland from 12 West European silicate producers covering about 93% of the total alkaline silicate production in Western Europe. The production routes for five typical commercial sodium silicate products were traced back to the extraction of the relevant raw materials from the earth. See Table 2 below.

Table 2: Copy of Fawer *et al.* paper with its Table 3 and highlighted values for CO_2 emission of sodium silicates



LCA Case Studies

Life Cycle Inventories for the Production of Sodium Silicates

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Table 3 *continued*

		LCI of various Sodium Silicates		
Weighted average Funktional unit		Na-silicate 3,3 (WR) furnace lumps, 100% 1,000 kg 100%	Na-silicate 3.3 (WR) furnace liquor, 7% solid 1,000 kg 37%	Na-silicate 2.0 (WR) hydrothermal liquor, 48% solid 1,000 kg
Solid waste				
Mineral waste	kg	127	47.2	20.2
Filter residues	kg		0.9	1.2
Inert chemicals	kg	0.6	0.3	1.7
Slags & ash	kg			2.3
Regulated chemicals	kg			0.004
Air emissions				
Ammonia (NH3)	g	237	88.3	0.03
Carbon dioxide fossil (CO_2)	g	1,066,022 1.000	424,668 424	288,698 288
Carbon monoxide	g	3,748	1,406	218

CO2 kg/t

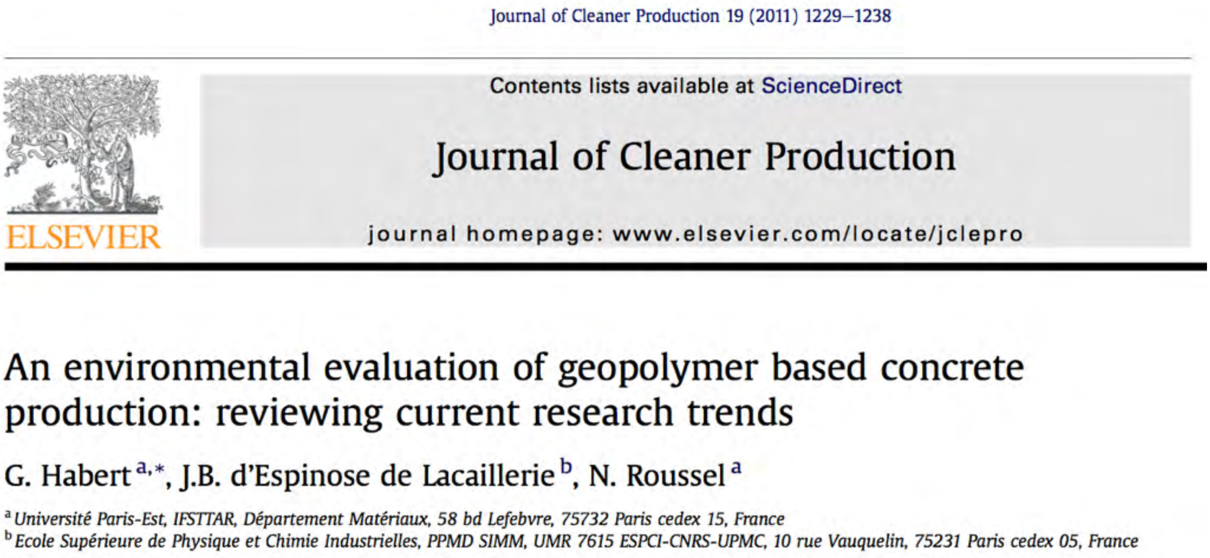
The CO_2 emissions for the two major sodium silicates discussed in this paper are:

- 1,066 kg/tonne (approx. 1,000 kg/t) for the glass sodium silicate Na-silicate 3.3 (WR) furnace lumps (100%),
- 424 kg/tonne for the solution Na-silicate 3.3 (WR) furnace route, 37% solid.

1) The false values in Habert et al. paper

The first LCA was presented at the Geopolymer Camp 2010 by Habert et al. (2010) (see at http://www.geopolymer.org/conference/gpcamp/gpcamp-2010) and it was a shock for the attendance. They claimed that, in terms of CO_2 emission, geopolymer cement was not better than Portland cement, and worse for other parameters. One of their studies involved a mix design containing metakaolin MK-750 and Na-silicate and, because of the high amount of alkali silicate needed in the formulation, they claimed that geopolymer cement emitted twice the amount of Portland cement. This statement was taken for granted by other scientists without any further consideration. Habert et al. did not recognize that this formulation was not geopolymer cement but rather a geopolymer resin/binder.

We mentioned that the presentation at the Geopolymer Camp 2010 by Habert (2010) was a shock for the attendance. We could get precise details in Habert et al. (2011) paper *An environmental evaluation of geopolymer based concrete production: reviewing current research trends*, Journal of Cleaner Production 19 (2011) 1229-1238.



Journal of Cleaner Production 19 (2011) 1229–1238

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An environmental evaluation of geopolymer based concrete production: reviewing current research trends

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It is a well-documented survey, which lists numerous mix-designs, the majority of them pertaining to alkali-activated-materials, not to genuine geopolymer cements/concretes. In fact they took each mineral ingredient separately, blast-furnace slag, fly ash, metakaolin, each of them being "activated" by a Na-silicate solution. They studied their LCA implications and stated that current alkali-activated mix designs made from fly ash alone or blast-furnace slag alone emit less CO_2 than Portland cement. However, this reduction is not sufficient to achieve the objectives. This study also highlights that the environmental impact of alkali-activated-materials stems from the use of the sodium silicate solution. In this case, the sodium silicate solution would lead to a pollution transfer within all of the other environmental impact categories.

The authors concluded that the ideal solution would be to strongly diminish the amount of alkali-silicate and, consequently, to follow the genuine geopolymer methods discovered and implemented by Davidovits. We quote: "*...the solution, proposed by Davidovits, has the advantage of using less [silicate] and slag than pure GBFS geopolymer concrete. This is beneficial from an environmental point of view...*" In other words: away from alkali-activated-materials.

In fact, another paper by Ouellet-Plamondon and Habert (2014) confirms that their study was dedicated to alkali-activated materials, not to geopolymer cements at all. It is found as Chapter 25, of the Handbook of Alkali Activated Cements, Mortars and Concretes, and is titled: *Life Cycle Assessment (LCA) of alkali-activated cements and concretes*.

The solution Na-silicate 3.3 (WR), 37% solid, is precisely the major raw ingredient in all mix designs listed in Habert's paper. Its CO2 eq. is given in Table 3 below:

Table 3: Copy of Habert *et al*. paper with its Table 2 and the kg CO2 eq. for Sodium silicate solution (37%) and Portland CEMI.

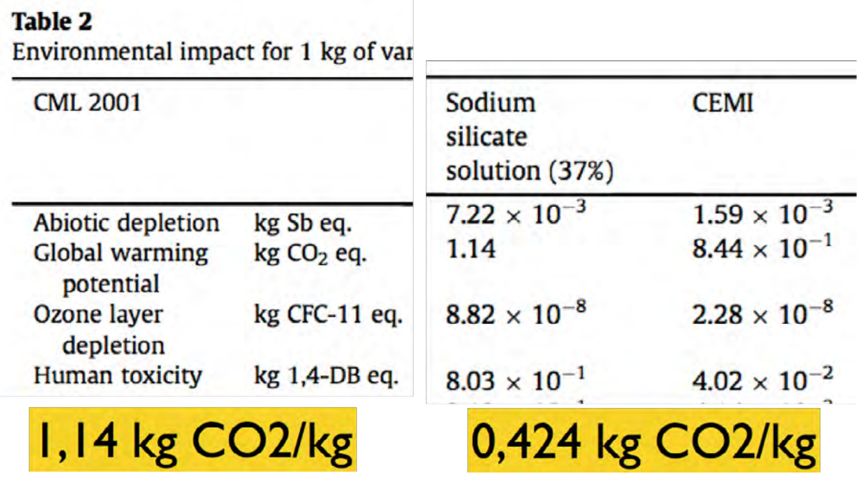


Table 2
Environmental impact for 1 kg of var

CML 2001		Sodium silicate solution (37%)	CEMI
Abiotic depletion	kg Sb eq.	7.22×10^{-3}	1.59×10^{-3}
Global warming potential	kg CO_2 eq.	1.14	8.44×10^{-1}
Ozone layer depletion	kg CFC-11 eq.	8.82×10^{-8}	2.28×10^{-8}
Human toxicity	kg 1,4-DB eq.	8.03×10^{-1}	4.02×10^{-2}

1,14 kg CO2/kg

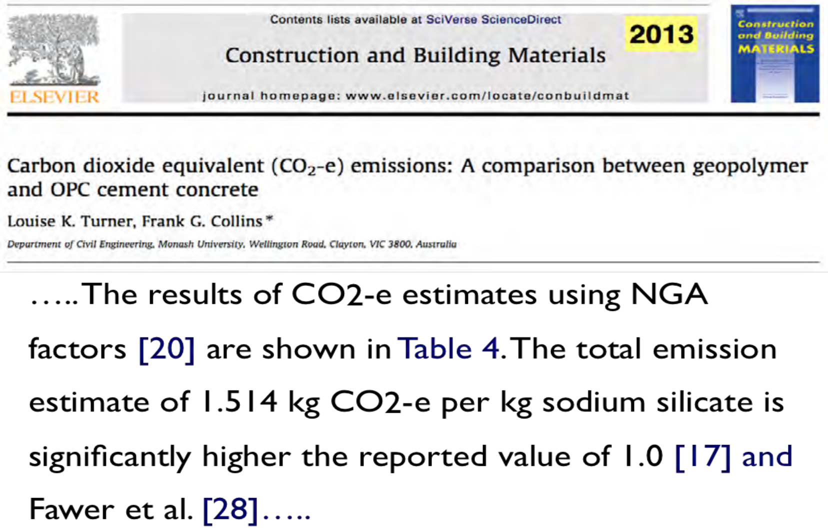
0,424 kg CO2/kg

Although Habert et al. write in their paper, quotation, " *data for sodium silicate solution come from Fawer et al. (1999)*", the value given in their Table 2 for the solution (37% solid) is 1.14 kg CO_2 eq. This is in the range of the value for the solid glass (100%), not diluted in water, in Fawer's paper, instead of the expected 0.424 kg CO_2 eq for the solution.

This is a methodological flagrant error. We may therefore conclude that all the CO_2 emissions and environmental impacts calculated in Habert et al. paper are wrong and must be roughly divided by 2.

2) The false values in Turner and Collins paper

We have the same situation in another paper by Turner and Collins (2013), *Carbon dioxide equivalent (CO_2-e) emissions: A comparison between geopolymer and OPC cement concrete,* Construction and Building Materials 43 (2013) 125–130.



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2013

Carbon dioxide equivalent (CO_2-e) emissions: A comparison between geopolymer and OPC cement concrete

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…..The results of CO2-e estimates using NGA factors [20] are shown in Table 4. The total emission estimate of 1.514 kg CO2-e per kg sodium silicate is significantly higher the reported value of 1.0 [17] and Fawer et al. [28]…..

The Australian team could not get any data from silicate producers. They calculated theoretically the CO_2 emissions for the Na-silicate glass (100%), got 1.222 kg CO_2 eq/kg for the emission arising during manufacturing (i.e. a value higher than Fawer and Habert), added 30% more for transport, and ended with a total emission estimate of 1.514 kg CO_2 eq per kg sodium silicate glass (100%).

The problem is that, like in Habert et al. paper, they used this value, estimated for the 100% solid lumps, on the place of the actual value of the diluted silicate solution (45% solid). In their Fig.3 reproduced here in Table 4, the quota for sodium silicate solution (45% solid) is 156 kg CO_2/m^3, the total for fly ash-based Geopolymer concrete is 320 to be compared with total 354 kg CO_2/m^3 for OPC.

Table 4: Copy of Turner and Collins paper with its Figure 3 and the kg CO2 eq. for Sodium silicate solution (37%) and Portland CEMI.

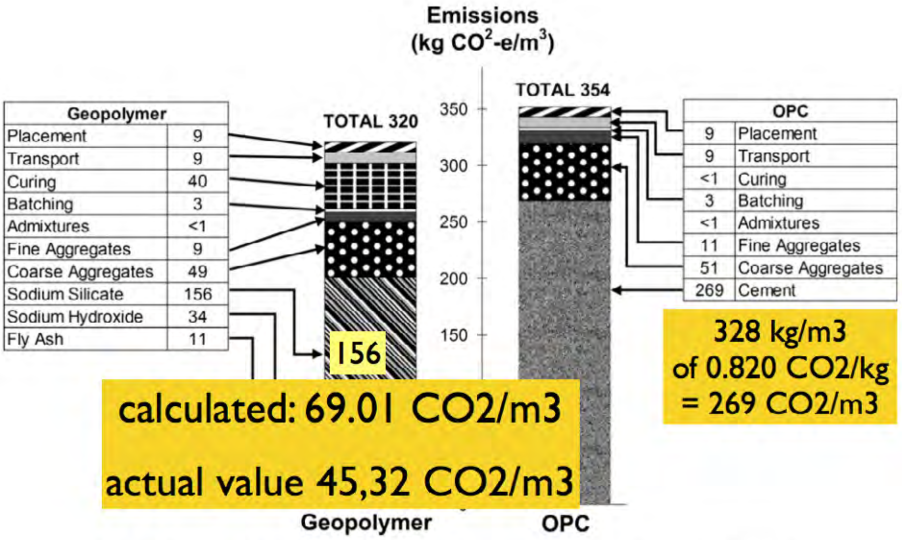
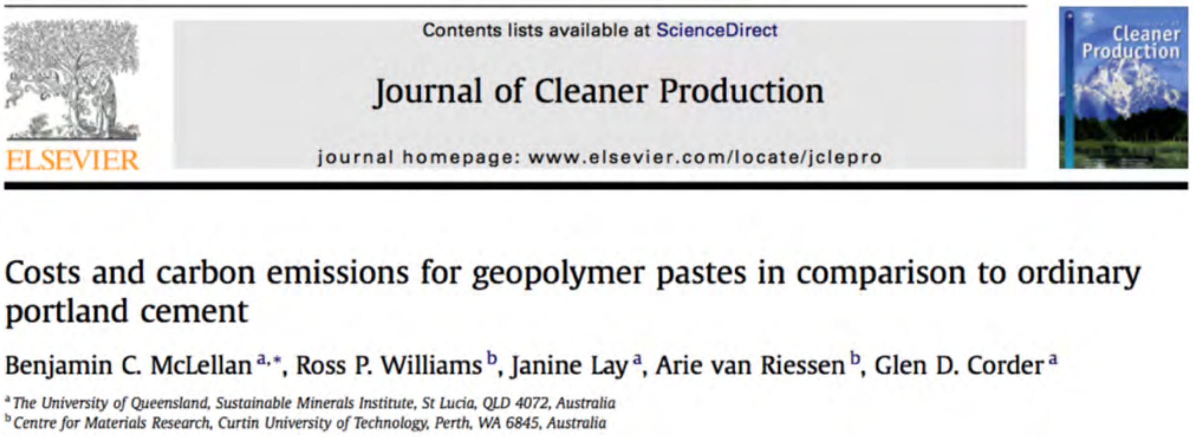


Fig. 3. Summary of CO_2-e for Grade 40 concrete mixtures with OPC and geopolymer binders.

Their Geopolymer concrete contains 103 kg/m³ of sodium silicate solution (45%, i.e. 0.67 kg CO_2 eq/kg). The amount of 156 kg CO_2/m^3 for Sodium Silicate solution in Turner and Collins paper is false; their correct calculated quota for the Geopolymer concrete would be lower, namely 69.01 kg CO_2/m^3. In fact, the actual number deduced from Fawer paper would give 45.32 kg CO_2/m^3, yielding a total of 209 kg CO_2/m^3 for this fly ash-based Geopolymer concrete. However, they used the heat-cured fly ash system and calculated an additional 40 kg CO_2/m^3 for heat cure. Well, industrialized/commercialized fly ash-based geopolymer concrete of Type 2 is an ambient cured system. The final value should be in the range of 169 kg CO_2/m^3 to be compared with their initial 320 kg CO_2/m^3.

Here too, the CO_2 emissions calculated are wrong and must be roughly divided by 2.

3) Other papers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Journal of Cleaner Production

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Costs and carbon emissions for geopolymer pastes in comparison to ordinary portland cement

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The paper by McLellan et al. (2011) *Costs and carbon emissions for geopolymer pastes in comparison to ordinary Portland cement*, Journal of Cleaner Production 19 (2011) 1080-1090, provides accurate values for the silicate solution.

The value found for the solid form, 100% concentration, was multiplied by 0.37 to get the value for the solution at 37% concentration. Yet, the conclusions are highly affected by the enormous transport distances found in Australia. The source locations for the sodium silicate are China, India, UK, and USA. They conclude that compared with emissions from Portland cement concrete, emissions from geopolymer concrete can be 97% lower up to 14% higher. Each application for geopolymers therefore needs to be assessed for its specific location, given that the impact of location on overall sustainability is one of the determining factors.

Another paper providing the actual numbers for the sodium silicate solution, namely 0.445 CO_2 kg-eq, was published by Heath et al. (2014), *Minimising the global warming potential of clay-based geopolymers*, Journal of Cleaner Production 78 (2014) 75-83. This value is in the range of the number given in Fawer et al. paper. Their target was to replace the expensive metakaolin MK-750 by cheaper calcined meta-clays.

Conclusion

Some published scientific LCA papers claim that, in terms of CO_2 emission, geopolymer cement was not better than Portland cement, and worse for other parameters. These statements are based on methodological errors and false calculations of the CO_2 emission values for geopolymer cement/concrete. The problem is that these false values are taken for granted by other scientists without any further consideration.

All LCAs published are also focusing on the amount of CO_2 that must be added to the original manufacture emissions in order to reflect the long distances that the raw ingredients and chemicals (metakaolin, slag, alkali-silicates) have to go all over before reaching their destinations. Sometimes, these distances are enormous: 6000 km for metakaolin or Na-silicate. This could contribute to a doubling of the Global Warming Potential numbers.

We feel, there is something misleading in these calculations. Special local environmental impact assessments are generalised to serve as references for the entire world. But the most striking element is that each paper compares a well-established 170-year-old industry involving hundreds of cement plants and terminals, with a start-up situation. Thinking in terms of innovation and R&D results implementation, the authors would have been better inspired in calculating at least 2 cases: first, their present laboratory situation, second, the one that will prevail in 5-10 years from now when industrialisation starts in full swing. There is a lack in the methodology as well as in standard procedures.

For people involved in R&D and innovation, the logic would have been to consider the market forces. As a matter of fact, business will foster the manufacturing of the chemicals and ingredients to take place as close as possible to the market. We know for example that a major global alkali-silicate manufacturing company has launched the marketing of a slag/fly ash-based geopolymer cement/concrete of Type 2. It is logical to understand why their target is to cover the emerging countries, India, Africa, and others, with alkali-silicates production sites located close to the market and to the geopolymer cement manufacturing sites.

The greenhouse gas emissions during the life cycle of Geopolymer Type 2 concrete are approximately 62%-66% lower than emissions from the reference concrete. The Type 2 geopolymer cement has ca. 80% lower embodied greenhouse gas intensity than an equivalent amount of ordinary Portland cement binder used in reference concrete of a similar strength, confirming the data published by the Geopolymer Institute, where the reductions are in the range of 70 % to 90 %.

We hope this selection will inspire additional, and much-needed, research on the environmental implications of genuine geopolymer cement mix designs, bearing in mind that industrialization and commercialisation already started with the production of structural geopolymer concretes for public buildings and infrastructure (airport). See at Geopolymer Institute News pages http://www.geopolymer.org/news and the Technical paper #23 GP-AIPORT (2015) *EFC Geopolymer Concrete Aircraft Pavements at Brisbane West Wellcamp Airport.* by Tom Glasby, John Day, Russell Genrich and James Aldred.

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