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Additional Information

# **Carbon footprint of geopolymeric mortar: Study of the contribution of the alkaline activating solution and assessment of an alternative route**

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## **Abstract**

CO<sub>2</sub> emissions associated with geopolymeric mortar prepared using spent fluid catalytic cracking catalyst (FCC) were compared to those calculated for plain ordinary Portland cement (OPC) mortar. Commercial waterglass used for preparing alkaline activating solution for geopolymeric mortar was the main contributing component related to CO<sub>2</sub> emission. An alternative route for formulating alkaline activating solution in the preparation of the geopolymeric binder was proposed: refluxing of rice husk ash (RHA) in NaOH solution. Geopolymeric mortar using rice hull ash-derived waterglass led to reduced CO<sub>2</sub> emission by 63% compared to OPC mortar. The new alternative route led to a 50% reduction in CO<sub>2</sub> emission compared to geopolymer prepared with commercial waterglass. Replacement of commercial waterglass by rice hull ash-derived waterglass in the preparation of the geopolymer did not cause a significant decrease in the mechanical strength of mortar. CO<sub>2</sub> intensity performance indicators ( $C_i$ ) for geopolymeric mortars were lower than that found for OPC mortar, indicating that the new route for activating solution led to the lowest  $C_i$  value.

**Keywords:** geopolymer, alkaline activating solution, rice husk ash, CO<sub>2</sub> emissions, cement, waterglass, spent fluid catalytic cracking catalyst.

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## 1. Introduction

Cement is an essential component of the building and construction industries since it forms the ‘glue’ that holds the sand and aggregate particles in concrete and mortar together. Although cement usually comprises only 10–15% of the mass of concrete, this minor constituent plays a significant role in determining the cost of concrete, its environmental impact and the properties of both fresh and hardened concrete. In particular, ordinary Portland cement (OPC) has become the centrepiece of the modern construction industry. Major environmental impacts associated with Portland cement include the energy required for production and transportation of clinker, the emission of greenhouse gases either directly or indirectly during manufacture, and the impact of mining, resource depletion and waste generation. The most serious environmental problems (such as CO<sub>2</sub> and NO<sub>x</sub> emissions) have a global impact and are the most difficult to combat.<sup>1</sup>

In order to increase the sustainability of concrete production, Parvulescu *et al.* demonstrated that bio-glycerol is a highly efficient renewable-based additive in the grinding process of concrete production and helps reduce energy costs and improves the quality of the resulting product.<sup>2</sup>

Several alternatives have allowed the development of new environmentally friendly construction materials. For example, Red Mud (the mineral processing waste from bauxite) can be used in the construction of roads, building foundations, in tile and brick production, *etc.*<sup>3</sup>

One route to remedy the problems of global warming due to greenhouse gas emissions and waste management, among others, is accelerated carbonation of wastes, giving reaction products that can increase the speed of hardening and the production of monolithic materials.<sup>4</sup>

Geopolymer, an alternative binder system, is emerging as a material with very attractive properties from an environmental and technological point of view. The development of geopolymeric cement is an important step towards the production of environmentally friendly cements. In the literature,

interesting books and review articles setting out its main features, reaction mechanisms, mechanical and durability aspects and applications can be found.<sup>5-10</sup>

Geopolymers are inorganic polymeric materials that were studied in depth by Joseph Davidovits in the 1970s. Geopolymerization involves a chemical reaction between alumino-silicate raw materials (in an amorphous state) and highly concentrated alkali silicate/hydroxide solutions yielding amorphous to semi-crystalline three-dimensional polymeric structures.

Geopolymerization has potential for the production of 'green' concrete and construction materials with a lower carbon footprint. They can be produced from a broad range of solid and fluid raw materials of significantly different qualities and chemical properties. However, the environmental profiles depend largely on the raw materials used. There are significant differences between resource-intensive primary solid raw materials (*e.g.* metakaolin) and less resource-intensive secondary solid raw materials (*e.g.* fly ash from coal thermoelectric power plants), and between resource-intensive primary fluid raw materials (*e.g.* NaOH solution, sodium silicate solution) and less resource-intensive primary fluid raw materials (*e.g.* water).

Some wastes are being used for preparing geopolymers by means of alkali activation: slag, fly ashes and others. Some authors have demonstrated the viability of using spent fluid catalytic cracking catalyst (FCC), a waste from the petroleum industry.<sup>11-14</sup> FCC waste was used as a raw material in the preparation of geopolymers by activating with a mixture of sodium hydroxide and commercial waterglass at 65°C. A stable binder was prepared and a mortar with compressive strength of 68 MPa was developed after 3 days of curing. The use of waste in the geopolymeric systems is highly desirable for two main reasons: first, it avoids the problem of disposal and, secondly, the CO<sub>2</sub> emissions associated with the production of these materials can be considered zero, because they are byproducts of industry.

The literature shows that the production of most standard types of geopolymeric concrete has a lower impact on global warming (mainly CO<sub>2</sub> emissions) than standard OPC concrete.<sup>15</sup> However, the production of geopolymer concrete has a higher environmental impact regarding impact categories

other than global warming.<sup>16</sup> Environmental impacts are associated directly with the production of reagents for preparing the alkaline activating solution, particularly the synthesis of the sodium silicate solution (commercial waterglass).

Turner and Collins reported the results of a comprehensive analysis of CO<sub>2</sub>-equivalent (CO<sub>2</sub>-e) estimates for both geopolymer and OPC concrete.<sup>17</sup> The study includes all the activities necessary for obtaining raw materials, concrete manufacturing, and construction of one cubic metre of concrete in metropolitan Melbourne. The CO<sub>2</sub>-e footprints generated by concrete comprising geopolymeric binders and 100% OPC concrete were compared. The CO<sub>2</sub> footprint of geopolymer concrete (320 kg CO<sub>2</sub>-e m<sup>-3</sup>) was approximately 9% less than comparable concrete containing 100% OPC binder (354 kg CO<sub>2</sub>-e m<sup>-3</sup>), which was much less than predictions by earlier studies. The key factors that led to the higher than expected emissions for geopolymer concrete include mining, treatment and transport of raw materials for manufacture of alkali activators of geopolymers, expenditure of significant energy during manufacture of alkali activators, and the need for elevated curing temperature of geopolymer concrete to achieve reasonable strength.

In order to assess the potential for the production of 'green' concrete and construction materials, the environmental impact of geopolymers can be quantified through Life Cycle Assessment (LCA) studies. Recently, a detailed environmental evaluation of geopolymer concrete production using the LCA principle was carried out.<sup>18</sup> The geopolymer concrete life cycle studied was restricted to the production of constituents used in concrete (cradle-to-gate approach). The other stages of concrete's life cycle (cradle-to-grave approach) were not considered. It is assumed that once concrete is cast in the structure, the impacts during the rest of its life cycle (maintenance and demolition) are similar for a geopolymeric concrete and for OPC concrete. A comparison of the ratio of raw material mass to the share of environmental impacts represented by the indicator GWP100 (Global Warming Potential) of a geopolymer prepared with 11.2% mass of sodium silicate solution reveals that this compound is the largest contributor to GWP (75.6% of total emissions).

Thus, the main environmental impact of geopolymer concrete is related to the use of sodium silicate solution (commercial waterglass) as a chemical activator. Therefore, it is desirable to find an alternative to this chemical reagent.

Rice husk ash (RHA), produced by the controlled burning of rice husk, has been used as a highly reactive pozzolanic material, leading to a significant improvement in strength and durability of normal concretes.<sup>19, 20</sup> RHA obtained by controlled combustion contains a high volume of silica ( $\text{SiO}_2$ , >90%) mostly in amorphous form. It was observed that the optimum combustion temperature for obtaining highly reactive RHA is 600°C.<sup>21</sup> Using RHA as a source of silica, Bejarano *et al.* obtained sodium silicate solutions by a hydrothermal treatment of the ash with NaOH.<sup>22</sup> The results showed that the best combination of variables for further dissolution of silica and suitable alkalinity was obtained using temperatures close to 100°C, molar ratio  $\text{H}_2\text{O}/\text{SiO}_2$  about 10, molar ratio  $\text{NaOH}/\text{SiO}_2$  about 2 and reaction times longer than 1 hour. In these conditions, the silica conversion to silicate was higher than 90%. In a recent paper published by Bouzón *et al.*,<sup>23</sup> a mixture of RHA, NaOH and water was boiled in a reflux system to dissolve silica and to obtain sodium silicate solution. This mixture was used as an alkaline activator instead of commercial waterglass, which is usually used in the manufacture of most geopolymers. All mortars with alkaline activator containing RHA showed compressive strength (cured at 65°C for 1 day) in the range 31–41 MPa, which is similar to control mortar prepared using an equivalent mixture of NaOH and commercial waterglass.

The aim of this paper was to compare the  $\text{CO}_2$  emissions of a geopolymer prepared by activation of FCC by using commercial waterglass–sodium hydroxide solution with those for a geopolymer prepared using an activating solution where the commercial waterglass has been replaced by an alternative source of active silica, namely RHA. This option allows integration of green chemistry principles into the preparation of new binders by reusing waste materials, reducing emissions of greenhouse gases and contributing to sustainable development.

## 2. Results and discussion

To calculate the CO<sub>2</sub> emissions associated with the preparation of geopolymeric mortars, the Intergovernmental Panel on Climate Change (IPCC) Guidelines for National Greenhouse Gas Inventories were followed.<sup>24</sup> The general methodology employed to estimate emissions associated with a particular process involves the product of activity level data, *e.g.* the amount of material processed or the amount of energy consumed, and an associated emission factor per unit of consumption/production according to:

$$E_i = A_i \times EF_i \quad (1)$$

where:  $E_i$  = the process emission (kg) of CO<sub>2</sub> from each ingredient or operation ‘i’;  $A_i$  = the amount of activity or processed material ‘i’; and  $EF_i$  = the emission factor associated with CO<sub>2</sub> emission per unit of activity or process material ‘i’.

Calculations were applied to the following mortars: (A) conventional OPC mortar, prepared by mixing Portland cement (OPC), water and sand; (B) geopolymeric mortar, prepared by mixing FCC, commercial waterglass, NaOH, water and sand; and (C) geopolymeric mortar, prepared by mixing FCC, RHA, NaOH, water and sand. The compositions of the compared mortars are summarised in Table 1.

**Table 1.** Compositions of prepared mortars.

Mortar Key	Component	Quantity/g
A	OPC	450
	H <sub>2</sub> O	225
	Sand	1350
B	FCC	450
	H <sub>2</sub> O	108
	NaOH pellets	54.87
	Commercial waterglass	253.15
	Sand	1350
C	FCC	450
	H <sub>2</sub> O	270
	RHA	78.75
	NaOH pellets	81
	Sand	1350

Three different cases were assessed: 1) CO<sub>2</sub> emissions from the binder only, whereby transportation processes were not included; 2) CO<sub>2</sub> emissions considering the external grinding of FCC and RHA, and transportation of materials from the origin; and 3) CO<sub>2</sub> emissions from the manufactured mortars.

In the preparation of mortar A, Spanish OPC (CEM I–52.5R) supplied by Cemex company (Buñol, Valencia, Spain) was used. This OPC did not contain any mineral addition. Chemical composition is given in Table 2.

**Table 2.** Chemical composition of OPC.

Compound	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	CaO	MgO	SO <sub>3</sub>	K <sub>2</sub> O	Na <sub>2</sub> O	LOI
Wt./%	17.69	4.37	3.35	67.19	1.47	3.38	1.23	0.47	2.35

LOI = loss on ignition.

For the second and third types of mortar (B and C), NaOH pellets were used. Commercial waterglass solution (used in mortar B) had a density of 1.35 g cm<sup>-3</sup> and its composition was as follows: 64% H<sub>2</sub>O, 28% SiO<sub>2</sub> and 8% Na<sub>2</sub>O. FCC came from BP OIL plant in Grao de Castellón (Castellón, Spain) and RHA was supplied by Dacsa (Almàssera, Valencia, Spain). Chemical compositions for FCC and RHA are summarized in Table 3.

**Table 3.** Chemical compositions of FCC and RHA (Wt./%)

Sample	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	CaO	MgO	SO <sub>3</sub>	K <sub>2</sub> O	LOI	Na <sub>2</sub> O	P <sub>2</sub> O <sub>5</sub>	TiO <sub>2</sub>	Cl <sup>-</sup>
FCC	47.76	49.26	0.60	0.11	0.17	0.02	0.02	0.51	0.31	0.01	1.22	-
RHA	85.58	0.25	0.21	1.83	0.50	0.26	3.39	6.99	-	0.67	-	0.32

LOI = loss on ignition.

Sand (siliceous type) was supplied by a factory located in Liria (Valencia, Spain). It has a fineness modulus of 4.1 and has similar characteristics to those specified in the UNE–EN–196–1:1996 standard. It has moisture content below 0.1% and a specific mass of 2680 kg m<sup>-3</sup>. In mortars B and C, the SiO<sub>2</sub>/Na<sub>2</sub>O molar ratio of the activating solution was kept constant at 1.17. The water/binder ratio

was 0.5 in mortar A and 0.6 in mortars B and C, being OPC the binder in mortar A and FCC the binder in mortars B and C.

**Case 1.** As mentioned above, this refers to a simplified calculation to estimate CO<sub>2</sub> emissions associated only with the components of the binder, not considering their mixing or curing processes. In addition, transport of materials to the laboratory from its origin was not considered.

- Mortar A, case 1. OPC is the only binder of this mortar. According to Årskog *et al.*,<sup>25</sup> emissions associated with it are 1 kg CO<sub>2</sub> per kg of cement. Other authors provide this same data.<sup>1, 26-28</sup> Using Equation 1, the emission associated with the use of 450 g of cement in mortar was calculated. Finally, CO<sub>2</sub> emission per mass unit of mortar (kg CO<sub>2</sub> per kg mortar) was given.
- Mortar B, case 1. The binder of this mortar is formed by FCC and alkaline activating solution. The solution consists of a mixture of commercial waterglass, NaOH and water. Since the FCC is an industrial waste, for the calculation of the associated CO<sub>2</sub> emission, the manufacture of NaOH and commercial waterglass was considered (no emission associated with water supply). Both emission factors, corresponding to NaOH and commercial waterglass, were obtained from the database of the SimaPro7.1 program (demo version, Pré Consultants Company of the Netherlands, LCA software specialists). The program draws from several recognised databases from different European Union countries. The emission factors were 1.12 kg CO<sub>2</sub> per kg of NaOH and 1.2 kg CO<sub>2</sub> per kg of commercial waterglass solution. Applying these factors to the amounts of these reagents used in the preparation of mortars, the quantities of CO<sub>2</sub> emitted per mass unit of mortars were calculated.
- Mortar C, case 1. Commercial waterglass replacement by RHA implies that the latter should be ground to reduce its particle size to increase its solubility in the hot NaOH solution. Then the mixture must be heated to boiling for 1 hour; this thermal treatment produces a suitable amount of available sodium silicate in solution. We must consider the CO<sub>2</sub> emissions associated with energy consumption of all of these operations. To grind the RHA, a ball-mill

with 0.3 kW electric power and with capacity for 450 g was used. The grinding time was 30 minutes. NaOH pellets were dissolved in water, and, after cooling, the ground RHA was added. Then this suspension was heated to boiling for 1 hour in a heater with 0.25 kW electric power. For calculation, we used the CO<sub>2</sub> emission factor for the electricity supplier to the Universitat Politècnica de València, ALPIQ, which is 0.33 kg CO<sub>2</sub> per kWh supplied. Thus, the proportional part of emissions for the amount of RHA used in the preparation of the mortar was calculated.

**Case 2.** In this case, CO<sub>2</sub> emissions associated with the binder were calculated, considering the external grinding of FCC and RHA, and transportation of all materials from the origin. As indicated by McLellan *et al.*,<sup>29</sup> transportation of materials is of key importance, and CO<sub>2</sub> emissions can be greatly affected by the distance and mode of transport. In Table 4, transportation data are shown.

**Table 4.** Transportation data for used materials.

Material	Itinerary and description	Total distance from origin to Valencia/km	Transport characteristics
OPC	Buñol – Valencia	45.3	Van (emission factor: 0.184 kg CO <sub>2</sub> per km) <sup>30</sup>
NaOH	Barcelona – Valencia	363	
Commercial waterglass	Madrid – Valencia	366	
FCC	Castellón – Onda Grinding in Onda Onda – Valencia	98.9	
RHA	Almàssera – Onda Grinding in Onda Onda – Valencia	135.4	

FCC was ground in Onda (Castellón, Spain), in a mill with 20,000 kg capacity and a power of 132 kW. The grinding time was 40 hours. RHA was also ground in Onda, in a mill with 70 kg capacity and a power of 2.5 kW. The grinding time was 4 hours. For both milling operations, we take as the

emission factor of energy consumption the national average value provided by IDAE,<sup>30</sup> which is 0.25 kg CO<sub>2</sub> per kWh.

Moreover, according to the PAS 2050 standard,<sup>31</sup> in the calculation of emissions associated with transportation, it is necessary to consider the return trip of the empty vehicle, not just the outward journey. Finally, applying the emission factors to the quantities of materials used in the preparation of mortars (A, B and C) and adding the emissions calculated in case 1, the total CO<sub>2</sub> emissions per unit mass of mortar for case 2 were calculated.

**Case 3.** This case considers all of the above plus the emissions associated with the sand, and the processes of mixing, compaction and curing of mortars. First, the factor emission of sand production is 8.46 g CO<sub>2</sub> per kg sand (from the database of the SimaPro7.1 program). This sand was transported from Liria to Valencia (29.9 km) in the same van as specified in Table 4.

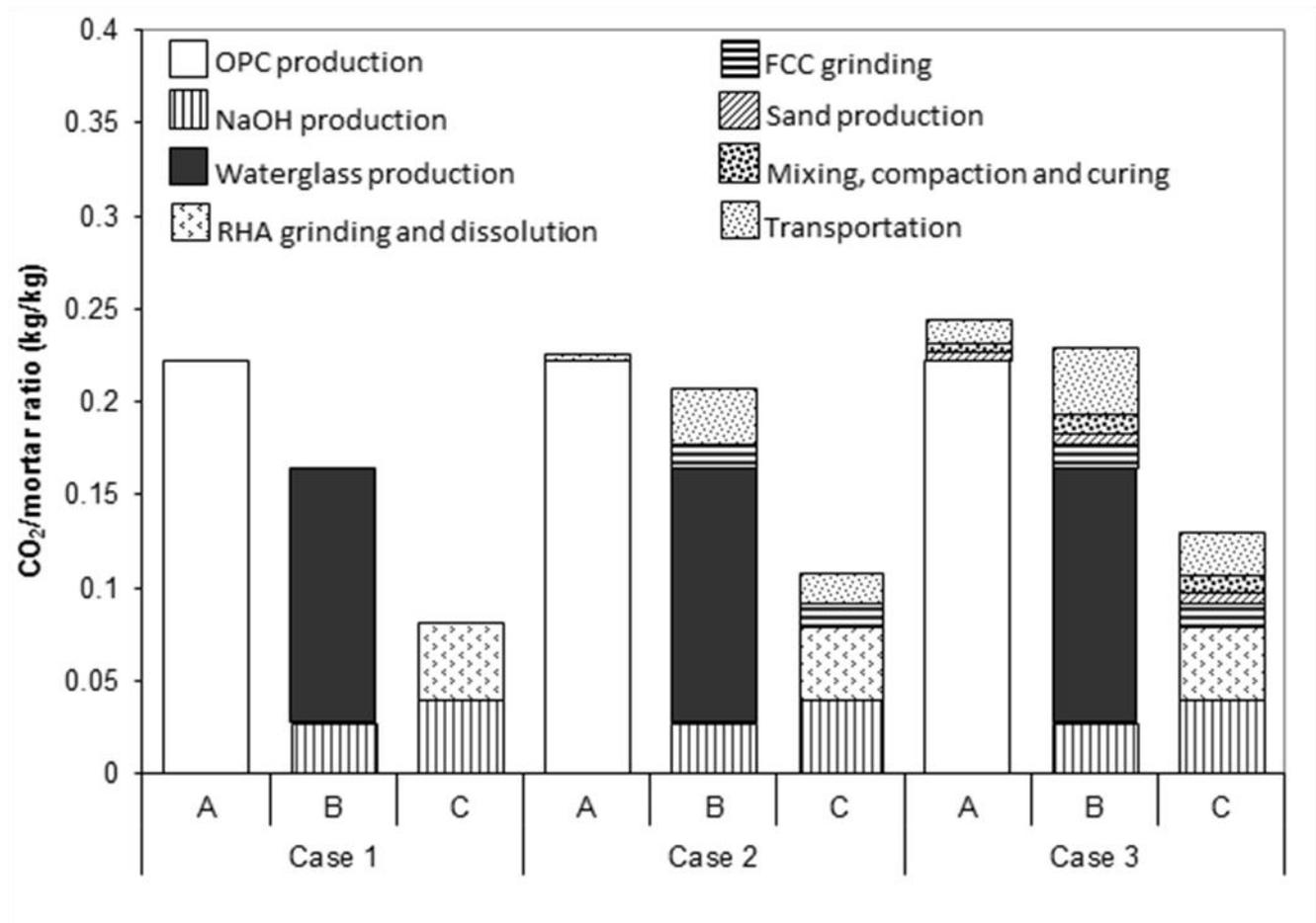
- Mortar A, case 3. To prepare an OPC mortar, we used a mixer of 0.25 kW of power for 4 minutes. Then the mortar requires compacting, which is carried out on a shaking table of 360 W of power and takes two cycles of 1 minute each.
- Mortars B and C, case 3. We used the same mixer described above (0.25 kW of power) to prepare geopolymeric mortars. It takes 7 minutes' mixing. The compacting is performed on a more powerful shaking table as above, namely 750 W of power, for 3 minutes for each mortar.

We will not consider emissions associated with the curing of the mortar, as it is performed at room temperature.

To compare the performance of prepared mortars, compressive strength was measured using a universal testing machine, according to UNE-EN-196-1:1996 standard, at 28 days curing age.

Fig. 1 shows the results for the calculation of CO<sub>2</sub> emissions for the three studied cases. For each case and mortar, values of kg of CO<sub>2</sub>/kg of mortar are given. For simplicity, we will use the symbol  $\varphi$  to identify the kg CO<sub>2</sub>/kg mortar ratio. Taking into account that we have three types of mortars and three

case calculations, we will use the following symbol:  $\phi_c^t$ , where 't' is the type of mortar (A, B or C) and 'c' is the case calculation (1, 2 or 3).

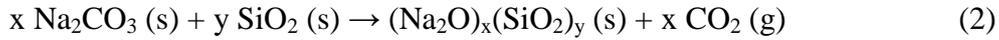


**Fig. 1.** Comparison of CO<sub>2</sub> emissions for the mortars studied in cases 1, 2 and 3.

For mortar A (OPC), the highest  $\phi_c^A$  values were found: the  $\phi_1^A$  value was 0.222, with the most important part being related to the synthesis of OPC binder. This value was slightly increased when transport and sand contributions were included ( $\phi_2^A = 0.226$  and  $\phi_3^A = 0.245$ ). This means that the main contribution for this mortar is due to the manufacturing of binder (more than 98% for case 2 and more than 90% for case 3).

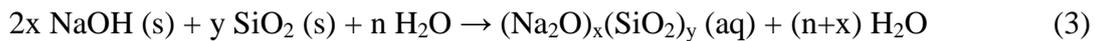
For mortar B, prepared using commercial waterglass as one of the components for preparing the alkaline activator, the  $\phi_1^B$  was 0.165 (25% less than mortar A). The contribution of commercial waterglass to the  $\phi$  value was the main one, being more than 80% of the total CO<sub>2</sub> emissions. The

synthesis of commercial waterglass is usually carried out by means of a high-temperature process: reaction of quartz and sodium carbonate at temperatures higher than 1300°C. In this reaction, the decomposition of sodium carbonate also produced CO<sub>2</sub>:



These results highlight the fact that the associated emissions related to commercial waterglass become the key for assessing and developing more friendly binders in the geopolymer field.

Finally, for mortar C, the most important contributions to  $\varphi^C_1$  were from the use of NaOH and the refluxing process of the mixture NaOH and ground RHA in water. In this process, a solubilisation of the silica in RHA was carried out (at *ca.* 130°C due to boiling-point elevation for the highly concentrated NaOH solution, *ca.* 30%) according to the following equation:



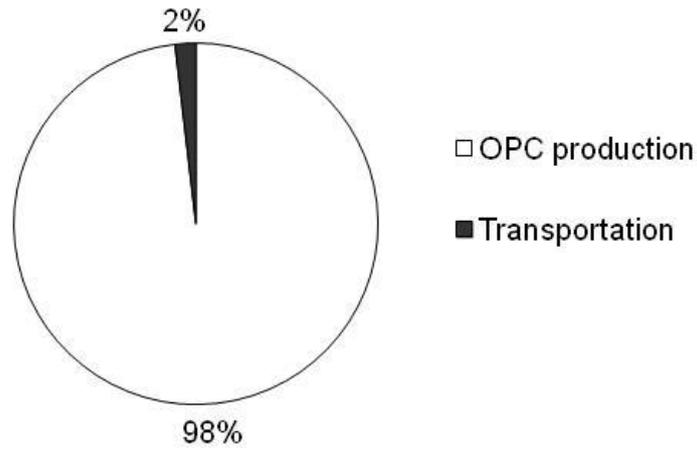
In this case, the value of  $\varphi^C_1$  was equal to 0.082, which is only 37% of the CO<sub>2</sub> emission produced from mortar A. Grinding and transport contributions for C mortar increased the value of  $\varphi^C_2$  to 0.108, and for case 3 the value  $\varphi^C_3$  was 0.130. This last value represents only 53% of the CO<sub>2</sub> emission produced from mortar A in case 3.

The greatest difference is observed in case 1, in which we have considered only CO<sub>2</sub> emissions associated with the binder, so that the nature of the material itself becomes the key to preventing global warming; the lowest emissions are associated with the use of RHA as the source of silica in the preparation of the alkaline activator. Replacement of commercial waterglass by rice hull ash-derived waterglass in the preparation of the geopolymer led to reduced CO<sub>2</sub> emission by 50%. Geopolymeric mortar using rice hull ash-derived waterglass led to reduced CO<sub>2</sub> emission by 63% compared to OPC mortar.

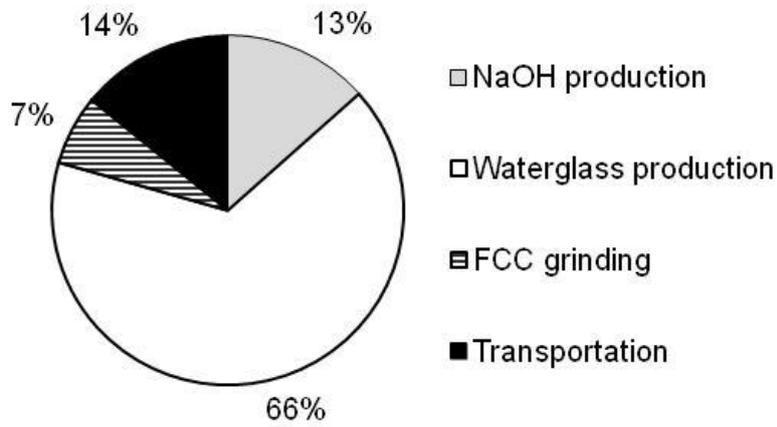
In case 2, as expected, the inclusion of transport in the calculations elevated CO<sub>2</sub> emissions in all three mortars. It is noted that the difference between the mortars A and B, in this case, is less than observed in case 1. This is because the OPC comes from Buñol (45.3 km away), while the reagents NaOH and

commercial waterglass come from Barcelona and Madrid, respectively, rather larger distances. In fact, in this case, we found that the sum of CO<sub>2</sub> emissions from the production of commercial waterglass and their transport accounted for 75% of the total CO<sub>2</sub> emitted for mortar B. The importance of the use of local resources is evident, as this can avoid unnecessary CO<sub>2</sub> emissions from transport.

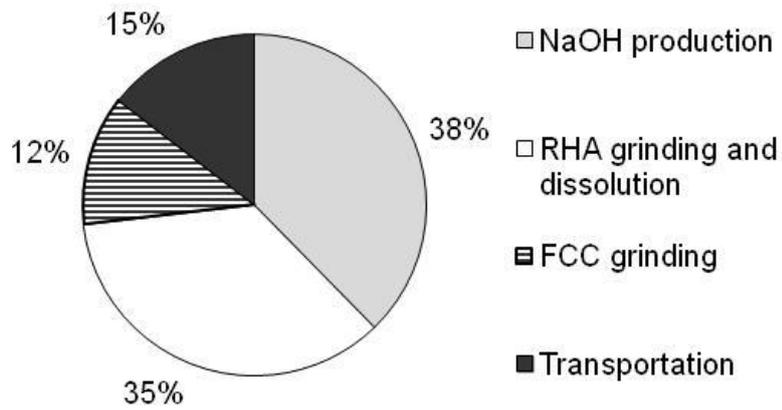
In Fig. 2, a comparison between the contribution to CO<sub>2</sub> emissions of the different operations and materials for each mortar in case 2 is shown. The main contribution to emissions in mortar A corresponds to the production of OPC, in mortar B corresponds to the production of commercial waterglass, and in mortar C, the production of NaOH closely followed by the production (grinding and dissolving) of RHA. Contribution percentages to CO<sub>2</sub> emissions corresponding with the preparation of rice hull ash-derived waterglass are: 3% for milling and 97% for the dissolution of RHA.



A



B



C

**Fig. 2.** Percentages of contribution to CO<sub>2</sub> emissions from operations and materials for mortar A, B and C, in case 2.

Notice that the percentage of emissions for RHA in mortar C is 35%, almost half of the emissions associated with the production of commercial waterglass (67%) in mortar B. This highlights the advantage of replacing the commercial product by the waste material. The proposed route for the synthesis of sodium silicate solution by refluxing a mixture of RHA/NaOH becomes the key to the reduction in CO<sub>2</sub> emission.

However, the CO<sub>2</sub> emissions associated with the previous treatment of RHA to obtain sodium silicate solutions by grinding and dissolving are quite large (35% of total). Thus it is desirable to find a way of dissolving the silica from the RHA that involves lower energy consumption and a reduction in CO<sub>2</sub> emissions due to the refluxing step. For example, one could use the heat-recovery process of burning rice husks for heating the alkaline solution and preparing the sodium silicate reagent.

In case 3, the differences between CO<sub>2</sub> emissions associated with the three mortars are practically the same as those obtained in case 2. The absolute values are larger due to the inclusion of the operations of sand transport and mixing/compacting of the mortar.

In conclusion, the materials that most contribute to the carbon footprint are the OPC in mortar A and the sodium silicate solution in mortar B. These results agree with those of Yang *et al.*<sup>32</sup>

A targeted CO<sub>2</sub> reduction in a mortar or concrete mix design should become an essential goal alongside the targeted compressive strength. Damireli *et al.*<sup>27</sup> proposed performance indicators to determine a benchmark and to establish feasible goals for OPC concrete. Using performance indicators would be helpful in determining the type and unit content of the binder needed to reduce CO<sub>2</sub> emissions in mortar or concrete production. Furthermore, the CO<sub>2</sub> reduction efficiency in a geopolymeric binder can provide a key tool to the mix design of such mortar or concrete.

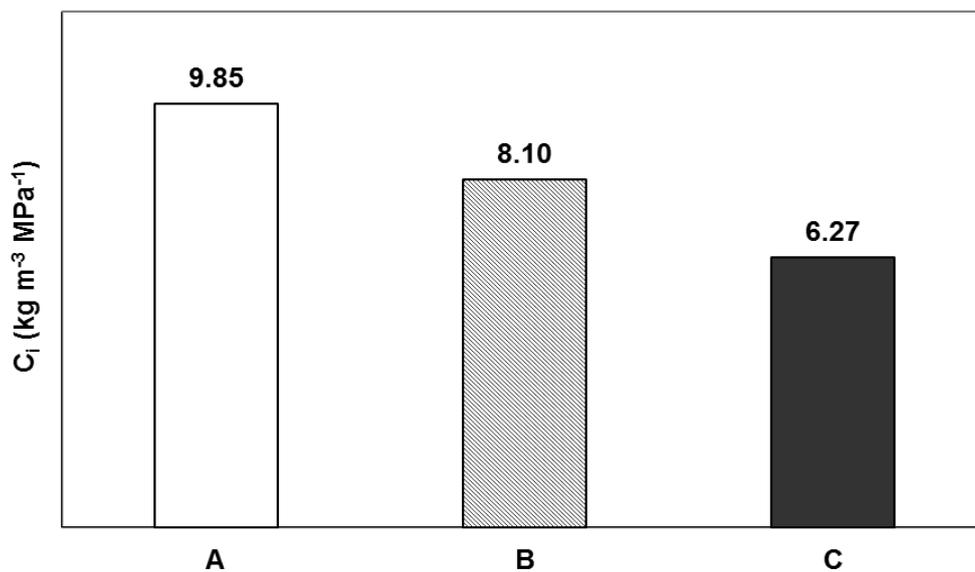
We used the CO<sub>2</sub> intensity (C<sub>i</sub>) indicator in order to assess the binder's influence on CO<sub>2</sub> emissions:

$$C_i = c/p \quad (4)$$

where  $C_i$  is the amount of  $\text{CO}_2$  emitted to deliver one unit of performance ( $\text{kg m}^{-3} \text{MPa}^{-1}$ ),  $c$  is the total  $\text{CO}_2$  emitted to produce mortar ( $\text{kg m}^{-3}$ ), and  $p$  is the compressive strength of mortar (MPa) at 28 days curing age.

To calculate  $C_i$  for the three prepared mortars, the first step was to transform the  $\varphi$  values to  $c$ , with units of  $\text{kg m}^{-3}$ , by dividing each by its corresponding density ( $2212.1 \text{ kg m}^{-3}$  for mortar A,  $2218.4 \text{ kg m}^{-3}$  for mortar B, and  $2121.1 \text{ kg m}^{-3}$  for mortar C).

The compressive strength of mortars prepared was tested after 28-days curing at room temperature. Compressive strength of mortar A was 55.0 MPa, mortar B was 59.7 MPa, and mortar C was 44.1 MPa. In Fig. 3, a comparison between  $C_i$  calculated for all three prepared mortars is shown.



**Fig. 3.** Comparison of  $\text{CO}_2$  intensity performance indicator ( $C_i$ ) for the mortars A, B and C, in case 3.

As can be observed,  $\text{CO}_2$  intensity decreases from mortar A to B and C. In terms of  $C_i$  values, one can assume that mortars B and C emit less  $\text{CO}_2$  to develop one unit of compressive strength. The calculated  $C_i$  for geopolymer C is 36.3% lower than that for mortar A, and 22.6% lower than that for mortar B, which is a significant reduction of the carbon footprint when the production of waterglass

solution (a component of the alkali activator) is carried out by thermal treatment of rice husk ash into a NaOH solution.

The results demonstrate that replacement of commercial waterglass by rice hull ash-derived waterglass in the geopolymer is not a significant detriment to their mechanical strength, which remains within acceptable values. The decrease in the mechanical performance in mortar C with respect to mortar B is probably due to the incomplete dissolution of silica from RHA in the refluxing process. However, the reduction in CO<sub>2</sub> emissions for mortar C is an advantageous option in environmental terms.

### **3. Conclusions**

- The most important parameter contributing to CO<sub>2</sub> emissions in geopolymeric mortars is the synthesis of commercial waterglass.
- Replacement of commercial waterglass by rice hull ash-derived waterglass in the preparation of the geopolymer led to reduced CO<sub>2</sub> emission by 50%.
- Replacement of commercial waterglass by rice hull ash-derived waterglass in the preparation of the geopolymer did not significantly decrease their mechanical strength. However, the reduction in CO<sub>2</sub> emissions for mortar C is an advantageous option in environmental terms, which can be seen in the results of the performance indicator CO<sub>2</sub> intensity, C<sub>i</sub>.
- The results demonstrate the great advantage of the alternative synthetic route in the preparation of rice hull ash-derived waterglass. This option allows us to integrate green chemistry principles into the preparation of new binders by reusing waste materials, reducing emissions of greenhouse gases and contributing to sustainable development.

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