Life cycle assessment of red mud-based geopolymer production at industrial scale

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Abstract. Nowadays the production of traditional building materials continues to be highly energy consuming and polluting. Therefore, the development of sustainable materials that allow the reuse of industrial waste could lead to a faster ecological transition of the construction industry. Among lab-scale developed materials, geopolymers are widely recognized as a future and sustainable alternative to traditional Portland cement, allowing to reuse various wastes from different industrial sectors.

This paper aims to perform an environmental analysis of geopolymers large-scale production based on the use of red mud, a waste product from the aluminum supply chain. Previous studies on red mud-based geopolymers have focused mainly on optimizing the laboratory formulations, without analyzing their real environmental advantage over traditional building materials. Therefore, this study uses the life cycle assessment approach to verify the environmental benefit of the large-scale production for the circular economy. Starting from the literature analysis, a four-steps scale-up procedure was applied to identify a potential industrial production.

Results showed a clear reduction in CO_2 emissions compared to the production of conventional Portland cement and highlighted the need to reduce the use of alkaline activators in geopolymers production.

Keywords: Sustainable manufacturing, Industrial scale-up, Ex-ante LCA, Red mud, Geopolymers.

1 Introduction

Construction industry is widely recognized as responsible for the generation of high quantities of waste and pollutant emissions into the environment [1]. Every year the production of cement-based materials causes the release of thousands of tons of greenhouse gas (GHG) emissions along with the consumption of a large volume of raw materials. In particular, for each ton of cement produced the GHG emissions are about 0.9 tons and raw materials consumption around two tons [2].

Aiming to the transition towards a circular and sustainable economy, the need of greener materials has led scientific research to focus on the development of alternative solutions. To face serious environmental degradation, sustainable development of new ecological binders that replace conventional cement has become increasingly

important. In particular, the reduction of both waste and CO_2 emissions is a driver for the design of any innovative production [3]. Over the last years, a new class of sustainable binders, known as geopolymers (GPs), has shown mechanical performance comparable to traditional ones [4]. GPs are alkali activated binders able to replace cement in the production of innovative mortars [5,6]. Geopolymer binders are obtained by activating aluminosilicate precursors with an alkaline solution. Different waste materials or industrial by-products can be used as resources in the production of GPs [7]. The valorization of such waste might result in a reduction of both energy and natural resources consumption in the construction sector [7,8]. It has already been demonstrated that the replacement of cement concrete with GPs may reduce the carbon emission up to 80% depending on aluminosilicate source and alkaline activators used [9].

Different aluminosilicate sources can be used as precursor for the GPs production, such as kaolinite, and some wastes from industrial and agricultural processes such as fly ash, blast slag and rice husk [10,11,12]. The potential use of wastes and/or by-products in the production of sustainable construction materials might lead to the identification of industrial symbiosis opportunities, with a significant impact on energy saving and emission reduction of involved industries [13]. In this context, Red Mud (RM) is an interesting waste produced by aluminum industry, whose chemical composition makes it a potential resource for the construction sector [14]. RM is usually stored in tailing dams and it can go through a dehydration and drying process to reduce its volume and maintenance costs [15]. Due to high alkalinity and toxicity of its constituents, the storage of red mud is a significant environmental problem, as it is dangerous for the earth, ecosystems and groundwater [16]. It is estimated that the RM quantity globally stored is around 3 billion tons, with an annually production rate of 150 million tons [11]. Therefore, safe disposal and valorization of red mud have become important issues for the aluminum industry [17]. Many literature contributions have reported studies and performance analyses on the use of RM in the production of building materials such as cement [18,19], concrete [20], brick [21,22], ceramics [23] and geopolymers [11,24].

As regards RM use for geopolymers production, different laboratory studies developed many RM-based geopolymers formulations, with the aim of studying mainly their mechanical performance by changing the quantity of the components or the process parameters, such as temperature of curing [25,26]. Nevertheless, these studies have focused only on the analysis of physical or mechanical performance, without providing any information on the technical feasibility and environmental footprint of industrial production of the developed samples. In particular, a predictive analysis of the environmental impact of the industrial production of red mud-based geopolymers is required to investigate their real environmental advantage over traditional building materials.

With this recognition, the present paper aims to verify the potential environmental benefit of the industrial production of a specific category of RM-based geopolymer samples presented in literature. The life cycle assessment (LCA) approach was used to identify the main criticalities of the large-scale production system and to highlight the potential advantages provided by the reuse of aluminum supply chain waste for the circular economy of the construction sector. Due to low alkaline reactivity of red mud in the geopolymerisation, different precursors including fly ash, slag, metakaolin and rice husk ash are generally used to optimize geopolymer production process [11,26,27]. This study considered only the red mud and fly ash-based (RM/FA-based) geopolymer specimens.

Contrary to most LCA studies analyzing existing industrial systems, this study aims to perform a LCA analysis of a hypothesized industrial production system, technologically consolidated only at laboratory scale. Some authors have already dealt with this type of LCA study, defining it "prospective LCA" [28] or "ex-ante LCA" [29]. Both definitions indicate the study of the large-scale performance of an emerging laboratory technology using the LCA approach, and the comparison with established technologies at full scale [30]. Despite the major limitations represented by the lack of reliable data on large-scale processes, technology developers can use ex-ante LCA to predict environmental and technical performance and optimize the analyzed processes.

In this study, the approach used to carry out the life cycle analysis is based on the framework proposed by Piccinno et al. [31], who provide a scale-up procedure to perform an ex-ante LCA. Figure 1 summarizes the phases of the approach used.



Fig. 1. Steps of the scale up procedure, based on Piccinno et al. [31] approach.

The first step consists in the definition of the laboratory scale process data obtained from the analysis of literature contributions, which develop samples of RM/FA-based geopolymer binders. All phases and parameters of the lab-scale production process should be identified in this step. The information collected is then used to design the plant flow diagram, which shows the sequence of the industrial scale production processes. After, the scale up of material and energy flows is carry out and finally the LCA study is performed. The methodological approach applied for every step of the scaleup procedure is specified in each dedicated section of this work.

The remainder of this paper is organized as follows. Section 2 reports the literature analysis and the definition of lab-scale process data. In Section 3 the industrial scale up is presented, while Section 4 is dedicated to the LCA analysis of the production system proposed in Section 3. The discussion of the results is given in Section 5, while the conclusions are summarized in Section 6.

2 Laboratory scale process data

This section presents the literature analysis and the definition of the lab-scale process data of the RM/FA-based geopolymer production. Due to the large number of literature works developing geopolymer samples based on the use of red mud, selection criteria have been applied to limit the boundaries of the literature analysis. First of all, a search based on the Scopus database was carried out. Three different keywords (i.e. "geopolymer", "red mud" and "fly ash") were used to identify a specific category of literature works, which develop only geopolymer formulations using a mix of red mud and fly ash as precursors. The number of articles was further reduced by taking into consideration only articles published in the last twenty years in journals or conferences. After, the abstract of the hundred articles obtained was carefully analyzed, selecting those which presented production processes and laboratory tests of the produced samples. Among these, a further selection was performed by analyzing the "material and methods" section, with the aim of taking into consideration only those papers that clearly show the production process by presenting the different stages and characteristic process parameters. Finally, only those papers that develop samples with uniaxial compressive strength (UCS) around 30 MPa were selected, aiming to compare formulations with the same performance characteristics.

Table 1 summarizes the process parameters that generally provide the best mechanical performance of RM/FA-based geopolymer samples developed at laboratory scale. Most of the previous work deals with optimizing these parameters in order to obtain formulations with the best performance.

Almost all the previous works have studied the effect of the amount of red mud and fly ash on the mechanical strength of geopolymers. The main result was that as the RM/FA ratio increases, the compressive strength decreases and samples show an increasingly ductile behavior [32,33]. Since compressive strength generally shows a peak for RM/FA ratios of 1:1 [26,32,33,34], in this study an equal quantity of red mud and fly ash will be considered for the industrial production hypothesis. Regarding the alkaline activator solution, most previous studies have investigated the effect of the type of alkaline solution used to activate the geopolymerization. They obtained that a binary solution with sodium hydroxide (i.e. NaOH) and sodium silicate (i.e. Na2SiO3) provides the best performance [26,33,35]. Sodium hydroxide is usually dissolved in deionized water and then mixed with sodium silicate. Almost all the works analyzed used a NaOH/Na2SiO3 ratio of 1:2.5. The molar concentration of sodium hydroxide has a great influence on the compressive strength and microstructure of the geopolymer. Generally, the higher the molarity of the alkaline solutions, the higher the degree of geopolymerization and the mechanical strength [36,37]. Most of the studies analyzed agree that a molar concentration within the range of 8-10M gives excellent mechanical strength to the geopolymeric binder [25,36,32,33,38]. After mixing the red mud and fly ash (i.e. the solid precursors of aluminosilicate) and preparing the alkaline solution (i.e. the liquid solution), the geopolymerization process is started by mixing the RM/FA mix with the alkaline solution. Literature contributions have highlighted the importance of liquid/solid ratio on compressive strength. In particular, Hu et al. [26] found that a lower liquid/solid ratio offers more advantages regarding the strength and the overall cost of the production, as using a higher quantity of alkaline activators results in higher costs. However, they also obtained that too low liquid/solid ratios can cause workability problems. In general, in the literature a value of about 0.5 is used [26,32,33,35]. As regards the curing conditions, the curing time is compulsorily fixed at 28 days by the standards, while the effect of temperature has been investigated. In general, curing at room temperature has been shown to be more effective in terms of greater compressive strength,

although some studies have obtained better performance by carrying out the first 24 hours of curing at 60 °C [25,34,38].

Red mud and fly ash also require a pre-treatment in order to optimize the geopolymerization process and improve both the microstructure and the mechanical strength of the geopolymers. It should be noted that the type of pre-treatment depends on the nature of the red mud and fly ash. If the moisture content is high and/or the size is excessively coarse, the raw materials must be dried at 105 °C for 24 hours and subsequently ground and sieved to obtain fine particle size [33,34].

Parameters	Values	References
Red mud/Fly ash ratio	1:1	[26,32,33,34,40,41,42]
NaOH/ Na2SiO3 ratio	1:2.5	[25,26,33,38,40]
NaOH Molarity	10 M	[25,32,33,38]
Liquid/solid ratio	0.5	[26,32,33,34,35,41,42]
Temperature of curing [°C]	20	[32,33,35,39]

Table 1. Process parameters proposed by previous literature works

3 Plant flow diagram design and industrial scale up

Once the laboratory process data have been analyzed in detail, this section aims to design the plant flow diagram of the industrial production system. Figure 2 presents the flow diagram of the RM/FA-based geopolymer binder production, developed on the basis of the considerations discussed in Section 2. The main process parameters are shown in Table 2. These have been chosen in order to obtain a product with an average compressive strength of 30 MPa, suitable for application in the structural field.



Fig. 2. Flow diagram of the RM/FA-based geopolymer production system

Once at the plant, red mud and fly ash must undergo pre-treatment processes. First, they should be dried at a temperature of 105°C for 24 hours according to laboratory scale procedure. However, the process time at industrial scale must be minimized due to the high production rates required. The new large-scale drying parameters can be calculated by considering the heat transfer theory, summarized in equation (1).

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$$Q = A \cdot U \cdot \Delta T \cdot t_p \tag{1}$$

A is the heat transfer surface, U is the overall heat transfer coefficient, ΔT is the temperature difference between the heat exchanger bodies and t_p is the process time. After a market analysis of the electric industrial ovens used for the drying process carried out on the website www.directindustry.it [43], it was assumed that the industrial oven has the same U coefficient as the laboratory one; while the heat exchange surface is 8 times larger. Based on these data, the drying time required to supply the same thermal energy was 3 hours.

Afterwards, red mud and fly ash must be ground to reach an average particle size of 0.355 mm [33] and finally sieved (process P2 and P3). After the mechanical-thermal pretreatment, equal quantities of red mud and fly ash are mixed for 10 minutes until a uniform mixture is achieved (process P4). The alkaline solution is obtained by dissolving anhydrous sodium hydroxide in deionized water at a molar concentration of 10 M, stirring the solution in a magnetic stirrer for 12 h (process P5) [1]. Note that this process

is highly exothermic and therefore a container made of material resistant to high temperatures must be used. Sodium silicate is then added to the solution in the mass ratio NaOH/Na₂SiO₃ equal to 1:2.5, mixing the whole for 5 minutes (process P6). Finally, the solid source of aluminosilicate consisting of the RM/FA mix and the alkaline solution are mixed in the liquid/solid volume ratio of 0.5 for 5-10 minutes. Mixing must be done always in the same direction until uniformity is achieved (process P7). Table 2 summarizes the main process parameters adopted and machinery to be used.

Process	Parameters		Machinery		
P1 – Drying	Temperature	105 °C	Continuous rotary oven		
	Time	3 h			
P2 – Grinding	2 – Grinding Particles size		Crushing machine		
P3 – Sieving	Mesh size	0.355 mm	Sieving machine		
P4 – Mixing	Time	~ 10 min	Rotor-stator type		
	Ratio RM/FA	1:1	nomogenizer		
P5 – Mixing	Time	12 h	Magnetic stirrer		
	Molar concentration	10 M			
P6 – Mixing	Time	~ 5 min	Mechanical mixer		
	NaOH/Na2SiO3 ratio	1:2.5			
P7 – Mixing	Time	~ 5-10 min	Rotor-stator type		
	Liquid/solid ratio	0.5	8		

Table 2. Main process parameters adopted and machinery to be used at industrial scale.

As regard the material flows at industrial scale, it was assumed an annual production capacity of 500,000 tons, equal to the average production capacity of Italian cement plants [44]. Assuming 330 working days per year, the daily production rate is around 1,500 tons. Based on the GP formulation parameters reported in Table 2, Table 3 summarizes the daily quantities of raw materials required for the assumed annual production.

Table 3. Raw materials daily quantity required

Materials	Quantity per day	
Red mud	555 tons	
Fly Ash	555 tons	
Sodium hydroxide	74 tons	

Sodium Silicate	185 tons
Deionized water	185 tons

The sizing of the energy flows was based on the approach proposed by Piccinno et al. [31]. In particular, the energy vectors required by the industrial processes are heat for drying and mechanical energy for the grinding, sieving and mixing processes. It was assumed to power all industrial processes with electricity and to have an energy conversion efficiency of 0.9.

The heat for drying was calculated based on the heat transfer theory. Equation (2) gives the total heat required, that is equal to the sum of the heat (Q_{heat}) to bring the material to the process temperature and the heat (Q_{loss}) needed to keep the temperature constant, due to losses.

$$Q_{drying} = Q_{heat} + Q_{loss} = c_p M(T_p - T_o) + \frac{Ak(T_p - T_o)}{s} t_p$$
⁽²⁾

The parameters involved are the specific heat c_p and the quantity M of the material to be heated, the process temperature T_p and the room temperature T_o , and finally the thermal resistance parameters assumed for the oven (i.e. the heat exchange surface A, the thermal conductivity k and the thickness s of the insulation layer).

The mixing energy was calculated according to equation (3) and depends on the diameter d and the type N_p of the impeller, the rotational speed N, the density ρ of the mixture, the mixing time t and the efficiency η_{mix} of the machine.

$$E_{mix} = \frac{N_p \rho_{mix} N^3 d^5 t}{\eta_{mix}}$$
(3)

As regards grinding energy, it mainly depends on the size of the final particles, the material to be grinded and the type of grinding. Piccinno et al. [31] suggest an average value in the range of [8-16] kWh/ton. In the absence of further data, it was assumed to take the maximum value of 16 kWh/ton, in order to have a conservative approach. The same approach was used for the sieving energy calculation, which is influenced by several parameters, and it is estimated by Alt [45] in the range [1-10] kWh/ton.

4 Life cycle analysis

The LCA analysis of the developed industrial production system is the final step of the approach used. The life cycle study was carried out in accordance with ISO guidelines [46] and with the help of Simapro software [47]. The life cycle inventory (LCI) was based on the industrial scale-up data calculated in the previous stage, on the Ecoinvent database (https://ecoinvent.org/the-ecoinvent-database/) and on literature references. The adopted life cycle impact assessment method was the ReCiPe 2016 Midpoint (H) consisting in the following impact categories: Global warming potential (GWP100), Ionizing radiation, Terrestrial ecotoxicity, Freshwater ecotoxicity, Marine ecotoxicity, Human carcinogenic toxicity, Human non-carcinogenic toxicity, Land use, and Fossil resource scarcity.

4.1 Goal and scope

The main goal of this study is to evaluate the environmental impact of the industrial production of RM/FA-based geopolymers and compare it with that of conventional Portland cement. The functional unit chosen for the comparison is 1 m³ of binder to use in the construction sector. As regards the system boundaries, the cradle-to-gate approach was used. This one involves the extraction of raw materials, transport and manufacturing phases until the final product is obtained. Since the analyzed production system involves the reuse of waste (i.e. red mud and fly ash), the "cut-off system model" was considered as multi-functional processes approach. It envisages to consider waste as "cut off" from the primary production system and, therefore, free of environmental burdens due to its previous life.

4.2 Life Cycle Inventory

The LCI refers to the scale-up calculations based on the framework presented in the previous sections of this work. Table 4 shows the material and energy flows resulting from the scale-up procedure of Section 3 and the data source of the inventory. It also reports the assumed data for the transport phase (i.e. 300 km for red mud and fly ash and 75 km for chemicals). All calculations were performed manually using Excel. Data are reported per functional unit of 1 m³ of GP binder produced.

Life cycle phase	Elementary flows	Quantity/FU	Source
Materials	Red mud	0.75 tons	Burden free
	Fly ash	0.75 tons	Burden free
	Sodium Hydroxide	0.1 tons	Ecoinvent
	Sodium Silicate	0.25 tons	Ecoinvent
	Deionized water	0.25 tons	Ecoinvent
Manufacturing	Drying heat (P1)	44.3 kWh	Ecoinvent:
	Grinding energy (P2)	12 kWh	Electricity, high
	Sieving energy (P3)	7.5 kWh	voltage {Europe
	Mixing energy (P4)	0.19 kWh	without Swit-
	Mixing energy (P5)	1.09 kWh	zerland} mar-
	Mixing energy (P6)	0.12 kWh	ket group for
	Mixing energy (P7)	0.35 kWh	Cut-off, S
Transport	Red mud and fly ash	300 km	lorry 16-32 metric ton

Table 4. Material and energy flows per functional unit

Sodium	hydroxide an	d so-	75 km	light	commer-
dium silicate				cial ve	hicles

5 Results and discussion

Table 5 shows the results of the main environmental impact indicators of the developed industrial scale-up compared with that of conventional Portland cement production. In particular, it reports the environmental characterization factors of each indicator per functional unit of binder produced. The cement impact relates to the cradle-to-gate production inventory of Portland cement in Europe, contained in Ecoinvent database.

 Table 5. Comparison between the potential environmental impact of 1 m3 of GP binder and

 Portland cement

	GWP [kg CO2 eq]	Terrestrial acidification [kg SO2 eq]	Water consump- tion [m3]	Mineral resource scarcity [kg Cu eq]	Fossil resource scarcity [kg oil eq]
Materials	312.9	1.49	6.24	3.11	78.37
Transport	119.1	0.39	0.24	0.53	41.73
Manufacturing	25.9	0.11	0.47	0.03	7.35
GP binder	457.9	1.98	6.95	3.67	127.45
Portland cement	1208.8	1.85	2.85	6.86	102.66

Looking at the GWP indicator results, the RM/FA-based geopolymer binder allows to reduce GHG emissions by approximately 62% compared to Portland cement production. This result is due to the greater amount of energy required for the production of cement. In fact, the firing of clinker at very high temperatures is responsible for the majority of CO₂ emissions into the atmosphere. On the other hand, the main advantage of producing geopolymer binders is that the geopolymerization process takes place at room temperature without the consumption of energy.

According to the findings of the other indicators, the production of GP binder requires 2.5 times as much water as Portland cement, since large quantities of water are consumed in the production of the alkaline activators. The mineral consumption of GP production is about half of the cement production. The last one consumes large quantities of limestone and clay, while the main raw materials of GP production are recovered waste. Finally, the equivalent consumption of fossil resources in GP production is higher (i.e. +24%), while the terrestrial acidification indicator result is almost the same. Analyzing in detail the impact of GP production, the main contribution is given by the material phase as the production of alkaline activators is highly energy-intensive. Regarding GWP indicator it represents about 68% of the total, although the use of red mud and fly ash is considered burden free. In fact, they are waste reused and their environmental cost is zero. This result shows that the main limit of using geopolymers in the construction sector is represented by the use of alkaline activators. The contribution of the production phase is only 5%, since the production system mainly consists of low energy-intensive processes. However, it should be noted that the results of the contribution analysis are affected by the assumption on the transport distances shown in Table 4. This effect could be reduced if the red mud-based geopolymer manufacturing facility was located adjacent to the aluminum manufacturing facility.

6 Conclusions

Recent effects of climate change have motivated the governance of the European Union to promote a major ecological transition plan towards a circular and sustainable economy, which involves all productive sectors. In construction industry, the scientific community has focused on the development of more ecological materials capable of reusing different types of waste. With this recognition, this study aims to investigate the potential environmental performance of red mud-based geopolymers, developed and analyzed only at laboratory scale. To this goal, this work proposed a four steps scale-up approach composed by: 1. the literature analysis to identify the main production parameters on lab-scale; 2. the design of a plant flow diagram; 3. the scale up of material and energy flows; 4. the LCA analysis.

The used procedure allowed to identify the main characteristics of the geopolymer formulation optimizing the mechanical resistance. After scaling up the manufacturing processes, LCA results showed that RM/FA-based geopolymers have a GWP index of approximately one-third that of conventional Portland cement and consume half the mineral resources. The main impact is caused by the use of alkaline activators, whose production is highly energy-intensive. However, the mechanical strength increases as their quantity increases. Therefore, future research should focus on developing formulations that use less amount of alkali solution without reducing the mechanical strength. From this point of view, increasing the quantity of red mud could be a solution, as it has a high alkaline content and could partially replace the use of sodium hydroxide [48]. Another solution could be to power the production of alkaline activators with renewable sources.

As regards the economic aspect, the usage of red mud in geopolymers production could have significant advantages for both sectors. In fact, dumping red mud usually requires a large amount of land: e.g. an alumina production plant with a production rate of 1 million tons per year requires 1 km² of land every 5 years [49]. This means a high cost to the alumina industry of about \$10 per ton [48]. Therefore, the start-up of an industrial symbiosis between the assumed geopolymer production plant and an alumina factory could allow to avoid the dumping of the red mud and to save about 1,800,000 dollars per year. Future work should perform a detailed economic analysis regarding investment and operating costs and potential revenue of the developed production system.

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