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SMART-ECO-INNOVATIVE COMPOSITE MATERIALS WITH SELF-CLEANING CAPABILITY AND ENHANCED RESISTANCE TO MICROORGANISMS

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Abstract

This research focuses on the development of advanced cementitious composite or geopolymer materials that exhibit smart, eco-innovative properties, including self-cleaning capabilities and heightened resistance to microorganisms. The aim is to address environmental concerns and enhance the durability and functionality of materials in various applications. The composite materials are designed by incorporating novel nanomaterials and eco-friendly additives, leveraging their unique properties. The self-cleaning capability is achieved through the integration of photocatalytic nanoparticles, such as titanium dioxide, which harness solar or artificial light to catalyse the degradation of organic contaminants on the material`s surface. The eco-innovative aspect of the research involves the utilization of sustainable components, minimizing the environmental impact of the composite materials throughout their life cycle. The materials are designed to correspond to the Circular Economy and Sustainable Development principles by reducing overall waste generation and the study involves a comprehensive characterization of the mechanisms that allow the production of these type of materials.

Key words: *advanced composites, microorganism resistance, photocatalysis, TiO_2 nanoparticles.*

INTRODUCTION

Due to the action of external factors, pollutants in the air, water, compounds resulting from the combustion of fuels used for heating and transport, buildings in the urban environment are subject to deterioration and decay. As a result of the growing awareness of environmental and public health issues, the need to manage industrial wastes and by-products from the energy industry, metallurgy, etc. has been identified. More specifically, the global environmental context of drastic reduction of harmful effects related to pollution, destabilisation of ecosystems, global warming and all their related elements, calls for an urgent need to identify possibilities to recycle fly ash, slag and other wastes. The implementation of the Circular Economy cannot be achieved at the level of a single sector of activity and, moreover, this approach would not be successful because if in one sector of activity the product represents waste or an industrial by-product, in another sector the same product may represent a valuable and under-exploited raw material.

Worldwide, in line with the principles of Sustainable Development and the Circular Economy, there is a strong orientation towards reducing the consumption of non-renewable raw materials, increasing sustainability, reducing soil, water and air pollution and, consequently, reducing the volume of waste or identifying possibilities for its recovery. From the point of view of the construction sector, a huge consumption of cement is identified, as it is the main raw material in many technological processes specific to this sector (Zailan et al., 2016). Producing such a large volume of cement/concrete is directly associated with environmental problems - cement production is responsible for about 5-8% of total carbon dioxide emissions (Cembureau, 2023; Aitcin, 2000; Sandu, 2021; Jamaludin et al., 2022).

The cement manufacturing industry estimates that, today, applying the best available technologies for producing Portland cement, CO_2 emissions from its production could be reduced by up to 17% (Damtoft et al., 2008; Warid Wazien et al., 2016; Lloyd & Rangan, 2010). At EU level, there are currently specific documents available for the cement industry that directly address and outline guidelines on reducing environmental impacts: Best Available Techniques (BAT) Reference Document for the Production of Cement, Lime and Magnesium Oxide (BREFs) (Best Available Techniques Reference Document, 2023) and the Commission Implementing Decision of 26 March 2013 setting out the conclusions on Best Available Techniques (BAT) under Directive 2010/75/EU of the European Parliament and of the Council on industrial emissions for the production of cement, lime and magnesium oxide (BAT) (European IPPC Bureau, 2013).

In all this context, on the one hand the high pollution potential of the cement manufacturing industry is identified, even applying BAT, and on the other hand two options for the use of fly are identified: the first, less convenient in terms of the degree of environmental impact, would be the use as an auxiliary raw material in the cement production process; the second, also preferable in terms of environmental criteria, being the use as the main raw material in the production of alkali-activated geopolymer materials (Friedlingstein et al., 2010; UNSTATS, 2010).

Research in recent years has shown that one possibility for the efficient management of these types of industrial wastes and by-products is their use in the production of geopolymer composites, so-called "green concrete" or "geopolymer concrete" (Stengel et al., 2009; Weil et al., 2009; Pachengo et al., 2008; Skvara, 2007). In the light of the urgent need to develop environmentally friendly materials that, at some point, will allow the reduction of concrete and, consequently, cement consumption, while at the same time allowing the reuse of waste and industrial by-products, the development of alkali-activated fly ash geopolymer materials present an area of great interest.

On the other hand, the possibility of producing cementitious or geopolymer materials with self-cleaning properties due to the photocatalytic properties of TiO_2 nanoparticles used as an admixture or as a substitute for a cement part is currently reported worldwide (Grebenişan et al., 2023; Janus & Zajac, 2016; Fujishima & Zhang, 2006). It is known that the biocidal mechanism and self-cleaning ability of composite surfaces containing TiO_2 nanoparticles is the result of two mechanisms, that of superhydrophilicity and that of degradation, destruction of molecules of organic nature, therefore, implicitly of microorganism cells these having a structure of organic nature (Zailan et al., 2017). In this context, it is known that the growth of micro-organisms (moulds, bacteria, viruses, algae, lichens, mites) on building surfaces has negative effects, on one hand on the health of the population, especially if the growth occurs on interior surfaces, and on the other hand on the health of buildings, causing, in addition to an unpleasant appearance, maintenance and repair costs (Haleem Khan & Karuppayil, 2012; Ebbehoj et al., 2002; Zeliger, 2013; Andresen et al., 2011).

Life Cycle Assessment (LCA) in the construction sector is a systematic approach to evaluate the environmental impacts of a building or infrastructure project throughout its entire life cycle, from raw material extraction and processing to construction, use, and eventual demolition or disposal. It considers various aspects such as energy consumption, resource depletion, emissions, and waste generation at each stage. LCA in construction industry helps in making informed decisions to minimize the environmental footprint of materials, buildings and infrastructure. It aids in the selection of sustainable materials, energy-efficient designs, and construction practices (Mellado et al., 2014). LCA also supports the development of sustainable infrastructure by providing a holistic view of the environmental performance of materials. It helps inform decision-makers about the environmental consequences of different choices during the planning and design phases and encourages the adoption of more sustainable practices and materials in construction projects.

Life Cycle Assessment of geopolymers involves evaluating the environmental impact of geopolymers throughout their entire life cycle, from raw material extraction to production, use, and disposal or recycling and are often considered as a more sustainable option compared to traditional Portland cement (Badurdeen et al., 2018; McLellan et al., 2011; Mellado et al., 2014). Furthermore, the incorporation of recycled aggregates in geopolymer concrete aligns with the growing emphasis on sustainability in the construction industry. This approach addresses environmental concerns, reduces reliance on virgin materials, and contributes to the development of more eco-friendly construction practices. Proper quality control and mix design considerations are essential to harness the full potential of recycled aggregates in geopolymer concrete applications (Bostanci et al., 2018; Bostanci, 2020; Guo et al., 2018; Lăzărescu et al., 2024).

Several studies indicate that it is not possible to make a simple sustainability comparison on the use of OPC and geopolymers. This is due to the significant impact of reagent transport and variability in the source of energy and technology used to produce the reagents. Costs have been minimized over time for OPC, as it is an established product; however, geopolymers are yet to go through this cycle of scale-up. Large scale geopolymer use is likely to lead to lower costs due to large orders of reagents (McLellan et al., 2011; Chen et al., 2010). To demonstrate the potential variability in the sustainability potential of geopolymer materials compared with OPC a case-by-case investigation must be carried out, considering several parameters regarding local raw materials, availability, location and influencing cost parameters.

The aim of this paper is to present preliminary results on the production of paving block elements produced using the concept of alkaline activation of fly ash and to present results on specific physical and mechanical characteristics. At the same time, to maximize the potential of generating smart-eco-innovative character, different types of aggregates were used to produce these elements, as well as the use of TiO_2 nano-particles into the geopolymer matrix. Finally, the present work aims to highlight an LCA calculation of the resulting CO_2 equivalent emissions by comparison with similar elements produced using traditional materials (cement) and a cost-benefit analysis of the results obtained.

MATERIALS AND METHODS

Several types of alkali-activated fly ash-based geopolymer composites and cementitious composites were designed, prepared and analysed for the aim of the research. In order to assess the LCA, the control samples were considered the paving blocks produces using only natural aggregates, granular class 0/4 mm with and without the addition of TiO_2.

The following raw materials were selected for the production of the alkali-activated fly ash-based geopolymer paving blocks:

- low-calcium fly ash that was used in this study was obtained from a power plant in Romania and the chemical composition was established by the means of the X-ray fluorescence analysis (XRF analysis) (Table 1). The particle analysis, in terms of cumulative distribution of the fly ash particles was established using a HELOS RODOS/L, R5 instrument capable of dry dispersion in the free aerosol jet for laser diffraction and dynamic image analysis (Figure 1);
- the alkaline activator used in the production of the alkali-activated fly ash-based samples was a combination between sodium silicate solution (Na_2SiO_3 solution) and sodium hydroxide solution (NaOH solution). The chemical composition of the sodium silicate solution is SiO_2=30%, Na_2O=14% and H_2O=56%. The sodium hydroxide solution was prepared by dissolving the NaOH pearls, 99% purity, into water, to obtain the desired concentration of the solution. The NaOH solution concentration was set to 8M;
- granular class 0/4 mm natural aggregates, recycled glass granular class 0/4 mm and micronized quartz were used in this study to produce the alkali-activated fly ash-based geopolymer paving blocks. Particle size distribution of aggregates is shown in Figure 2 and Figure 3;
- TiO_2 nanoparticles of type AEROXIDE® TiO_2 P25, according to the manufacturer's technical data sheet, these TiO_2 nanoparticles are characterized by a purity of 99.5%, containing more than 70% anatase crystalline phase.

Ordinary Portland cement CEM I 52.5 R was used to produce the control samples for the assessment of the LCA.

For all tested mixtures, the introduction of 3% TiO_2 nanoparticles as a percentage of total ash/cement mass was considered to determine the LCA.

The chosen TiO_2 percentage is in accordance with the literature, which shows that for a TiO_2 percentage of 3%, both the physical-mechanical performance and the resistance to microorganisms' action can be improved.

Table 1. Fly ash chemical composition

Oxides	%
SiO_2	46.94
Al_2O_3	23.83
Fe_2O_3	10.08
CaO	10.72
MgO	2.63
SO_3	0.45
Na_2O	0.62
K_2O	1.65
TiO_2	0.92
L.O.I.	2.11

Based on the results regarding the possibility of developing such construction elements (Lăzărescu et al., 2022; 2024), the research initially focused on preliminary evaluation of the physical, mechanical and durability characteristics of the obtained products, for the study of their potential, different areas of applicability:
(1) Apparent density;
(2) Weathering resistance – total water absorption;
(3) Weathering resistance – freeze-thaw resistance with de-icing salt;
(4) Tensile splitting strength;
(5) Abrasion resistance;
(6) Slip/skid resistance.
All the test were performed according to norm EN 1388:2004/AC:2006: Concrete paving blocks. Requirements and test methods (ASRO, 2006), which specifies the materials, characteristics, conditions and methods of testing the paving blocks, and it is applicable for the use of pedestrians, vehicles, bicycle lanes, parking lots, roads, highways, industrial areas, etc. To be used in specific applications, they must comply with certain conditions at the time of their declaration as fit for use by the manufacturer.
The Life Cycle Assessment (LCA) was carried out based on a methodology consisting of four steps: setting the objective and defining the scope (1), life cycle inventory (2), environmental impact assessment (3) and interpretation of the results (4) (Figure 4).
For each proposed mix-design the LCA diagram, the main environmental impact element in terms of CO_{2Eq} was identified. Thus, in the case of geopolymer composites, the heat treatment required for the geopolymerization processes is identified as the main impact element, due to the electricity consumption used to achieve a temperature of 70°C in the heat treatment chamber during the first 24 hours after casting. A possible method to improve the environmental impact in this case is to optimize the heat treatment process or, through compositional optimization, even to eliminate it. In the case of cementitious composites, the main environmental impact in terms of CO_{2Eq} has been identified as cement, the main raw material.
The LCA has been calculated in terms of price/m^2, but also in terms of CO_{2Eq}/m^2. to produce type I paving blocks (22.5 x 8.8 x 6.0 cm - Figure 5).

RESULTS AND DISCUSSIONS

From the point of view of the physical-mechanical characteristics of the composites analysed the following can be said:
- Geopolymer binder-based composites have a lower density compared to cementitious ones (Figure 6). This parameter is influenced by both the type and nature of aggregates used, with recycled glass aggregates and natural aggregates contributing to the increased density of the composite. On the other hand, the addition of NT contributes to the densification of the material, causing a slight increase in density, through the changes it induces at the microstructural level, especially at the porosity level of the material.
- The use of recycled glass aggregates contributes to reducing the water absorption of the geopolymer composite, a parameter which for the use of natural aggregates or micronized quartz would be close to the water absorption characteristic of cementitious composite. The influence of TiO_2 nanoparticles is also observed this time on the characteristics of the composites, the water absorption reducing with the addition of nanoparticles, both for the geopolymer-bound composites and for the cementitious composite.

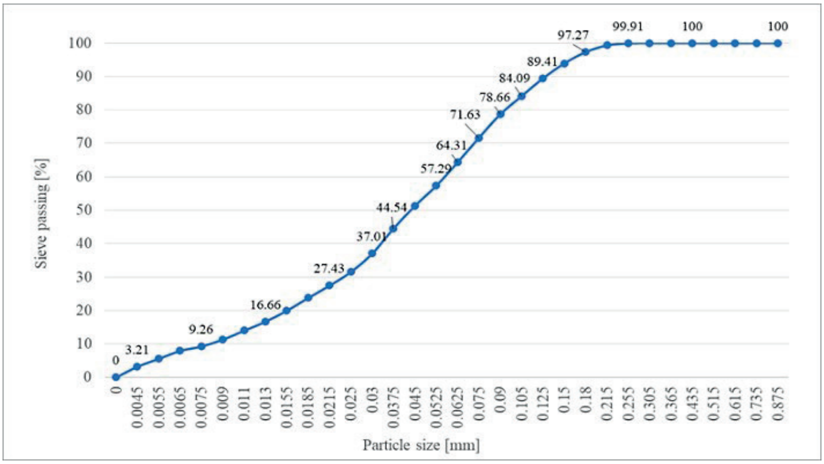

Figure 1. Particle size distribution of fly ash

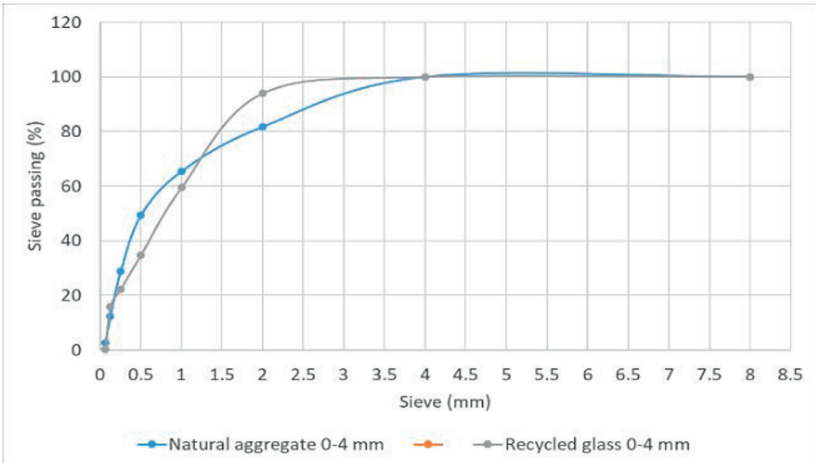

Figure 2. Particle size distribution of natural aggregates granular class 0/4 mm
and recycled glass aggregates 0/4 mm

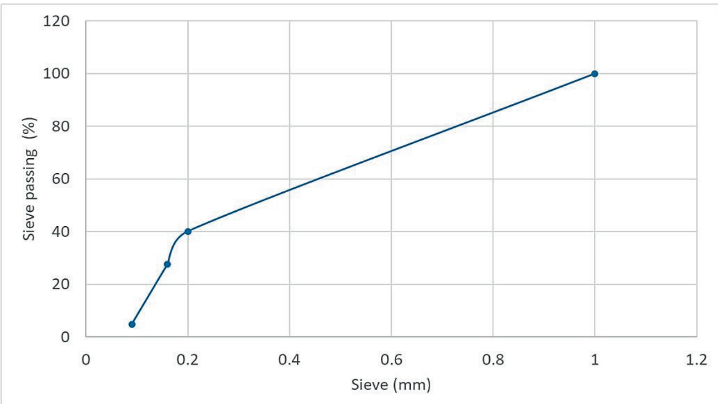

Figure 3. Particle size distribution of micronized quartz

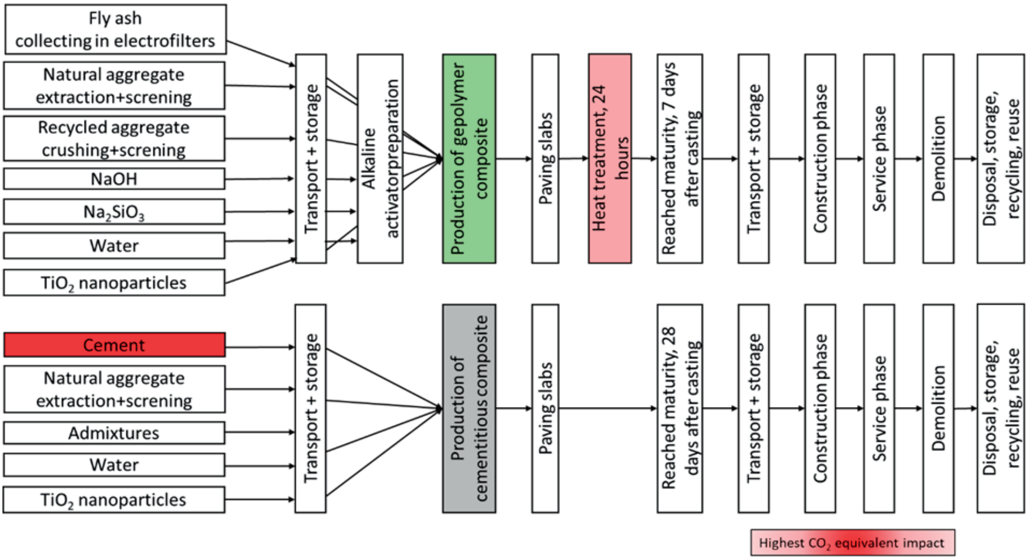

Figure 4. Life Cycle Assessment methodology- Comparative analysis of alkaline activated fly ash geopolymer paving blocks vs. OPC paving blocks

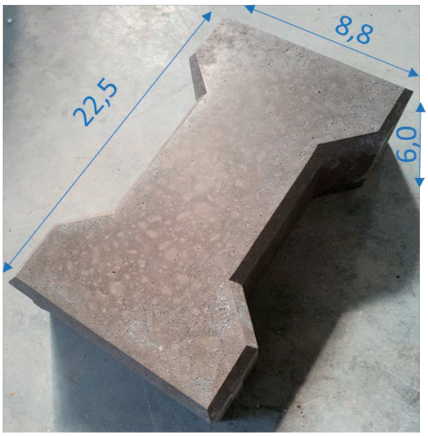

Figure 5. 22.5 x 8.8 x 6.0 cm paving block

By introducing NT as an addition (Figure 7), the water absorption of geopolymer composites decreases by 12.5-15.8% compared to similar materials without NT, and in the case of cementitious composite, the decrease is 10.3%. These changes are beneficial, as a lower water absorption is favourable to increase the resistance of the material to the action of environmental agents and, therefore, to increase the lifetime.

- The abrasion resistance (Figure 8), measured by volume loss, is relatively similar for geopolymer composites compared to cementitious, but with the introduction of TiO_2 nanoparticles, this volume loss is decreased by 1.1% for cementitious composites and by 5.3-6.2% for geopolymer composites. This time too, the influence of the nature of the aggregates is noticeable, the wear resistance being better for the use of recycled glass aggregates and even micronized quartz.
- As expected, considering the densification of the composites and the reduction of their porosity due to the introduction of NT in the composite matrices, an improvement of the freeze-thaw resistance of both the composites with geopolymer binder (30-50%) and the one with cementitious binder (25%, Figure 9) can be observed, which will again lead to an increase of the lifetime of the products through an improvement of the resistance to the action of environmental factors.
- The splitting strength (Figure 10) is lower in the case of geopolymer material compared to cementitious composite, but this parameter proves to be improvable, on the one hand by compositional changes (type and nature of aggregates), and on the other hand by the introduction of TiO_2 nanoparticles.
- Another important aspect is to reduce the slip potential. As seen in Figure 11, it can also be appreciated that changes at the compositional

level of the geopolymer composite, i.e. changes in the nature of the aggregates, but also the introduction of NT contribute beneficially. Thus, if for the geopolymer composite with natural aggregates the slip potential is classified as "moderate" (USRV 20-39), with the use of recycled glass or micronized quartz aggregates, this potential improves by one class, respectively, reaching the "low" classification (40-74). Also, the introduction of NT in the composite matrix has the effect of transitioning from the "moderate" class (USRV 20-39) to "low" class (40-74), all of which contributes to increasing the operational safety for users of precast paving products. A beneficial effect of NT is also seen in the case of cementitious composite, with a transition from "moderate" class (USRV 20-39) to "low" class (40-74) also recorded.

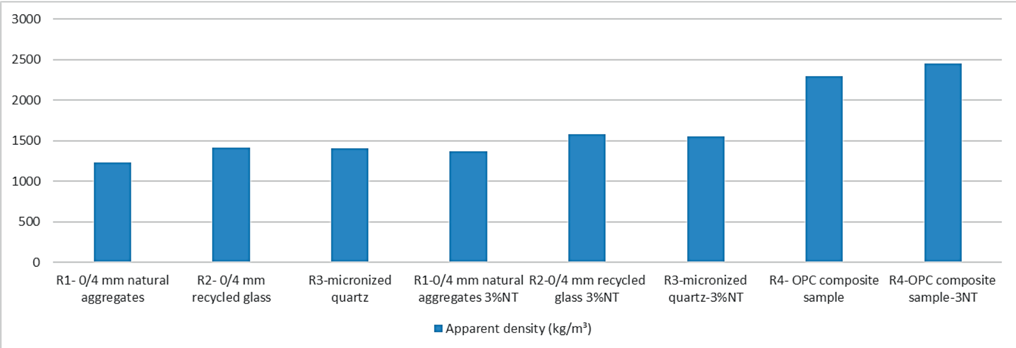

Figure 6. Apparent density of alkaline activated fly ash geopolymer paving blocks and OPC paving blocks

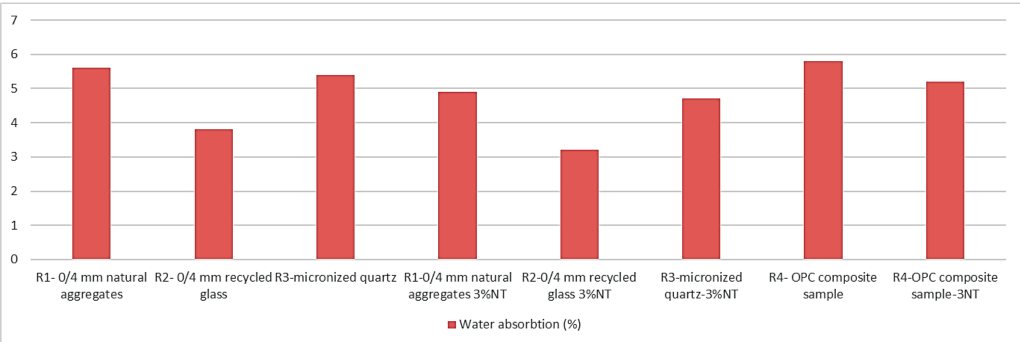

Figure 7. Water absorbtion of alkaline activated fly ash geopolymer paving blocks and OPC paving blocks

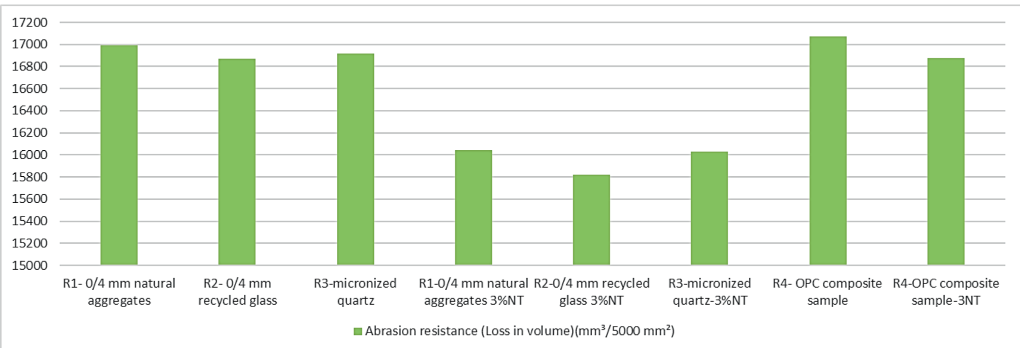

Figure 8. Abrasion resistance of alkaline activated fly ash geopolymer paving blocks and OPC paving blocks

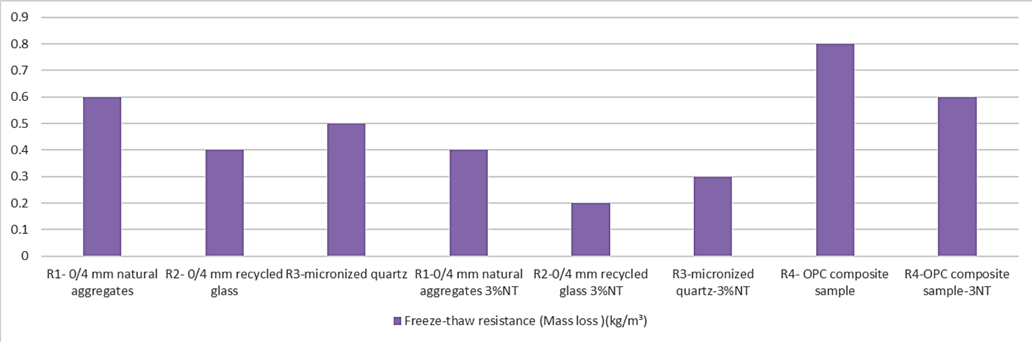

Figure 9. Freeze-thaw resistance of alkaline activated fly ash geopolymer paving blocks and OPC paving blocks

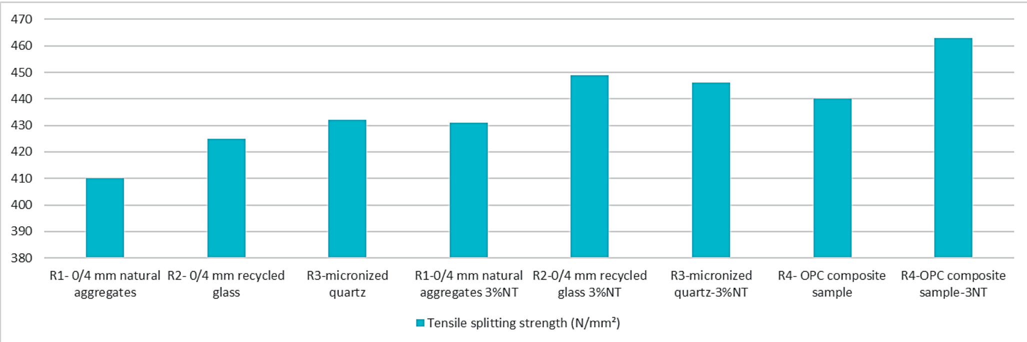

Figure 10. Tensile splitting strength resistance of alkaline activated fly ash geopolymer paving blocks and OPC paving blocks

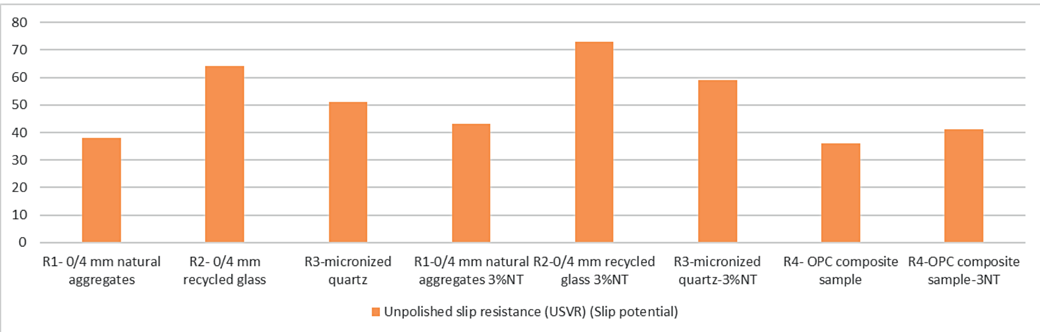

Figure 11. USVR resistance of alkaline activated fly ash geopolymer paving blocks and OPC paving blocks

Analysing each stage of the LCA in turn, the following can be said:

- The raw materials used to produce the geopolymer paving blocks have several advantages in terms of environmental impact: fly ash, the main raw material, is a waste product, collected, by means of electrofilters that consume electricity, but which would otherwise be disposed of/landfilled as waste, which would have polluting consequences. Similarly, it is considered that the issue of recycled aggregates (glass) should also be addressed. Chemical raw materials (NaOH and Na_2SiO_3) are commonly used materials in various industries, easily accessible, affordable and with medium environmental impact.
- Among the raw materials used in the preparation of the cementitious composite, the

strong polluting impact of cement stands out, while the natural aggregates, used for both variants of composites, contribute equally in terms of CO_{2Eq}, as well as TiO_2 nanoparticles. However, in the case of TiO_2 nanoparticles, although they contribute their own CO_{2Eq} contribution to the final product, their use in composites induces significant benefits on the physical-mechanical characteristics and, consequently, on the increase of the product lifetime and enhanced resistance to microorganisms.

- The phases of transport and storage, production of prefabricated elements (paving blocks), installation, use, maintenance, demolition and disposal, recycling, reuse, being carried out in a similar way for both cases analysed (geopolymer vs. cementitious), have an equal contribution in terms of environmental impact.
- The alkaline activator phase, specific to the preparation of geopolymers, is carried out by simply dosing and dissolving NaOH in water, i.e., subsequent mixing with Na_2SiO_3, and therefore does not have a significant effect in terms of CO_{2Eq}.
- The conditioning and maturation stages of the products show some differences, depending on the type of composite. Thus, in the case of geopolymers, the heat treatment phase is identified as the main environmental impact element in terms of CO_{2Eq}, as previously analysed, followed by an environmental storage phase, which does not represent an environmental impact factor. In the case of cementitious composites, although there is no heat treatment phase, the need for curing is 4 times longer than in the case of geopolymers and under specified conditions (immersion in water) indicates that even in this case, although quantitatively smaller, there is an environmental impact due to the need for handling, with electrically or auto-mechanically operated equipment, of the prefabricated elements and the achievement and maintenance of suitable environmental conditions (water temperature).
- The installation is mostly done by hand, and during the installation there is generally no maintenance/repair work that would induce a CO_{2Eq} contribution.

An estimate carried out for the production of prefabricated paving blocks needed to cover a pedestrian area equal to 1 m^2, revealed the following (Figure 12):

- as expected, the production of geopolymer composite paving blocks increases the cost price significantly, mainly since the technology is still relatively new and includes the use of more expensive raw materials (NaOH and Na_2SiO_3) to ensure constant product performance;
- partial substitution of natural aggregates with recycled waste glass aggregates does not contribute significantly in terms of price, but has a clear role in reducing the carbon footprint;
- partial substitution of natural aggregates with micronized quartz increases the cost price of geopolymer composite paving blocks, while slightly increasing the carbon footprint;
- the use of TiO_2 nanoparticles induces a price increase, as this material is expensive, and induces some increase in carbon footprint.

All these four observations, at a first analysis, would not seem to encourage the hypothesis of paving blocks produced using geopolymer composite. If all this is analysed in a broader context, on the one hand comparing the carbon footprint with that of cement composite paving blocks and on the other hand comparing the physical-mechanical and durability performance, the results of the analysis are quite different.

Firstly, in all cases the CO_{2Eq}/m^2 pavement is much lower for geopolymer composite than for cementitious composite, with 35-50% lower values than for the use of cementitious composite, which is in line with the literature, although the cost price is substantially higher. This is even more so with the use of nanoparticles, which is also consistent with the literature (McLellan et al., 2011).

If the benefits in terms of improved product performance (resistance to environmental agents, wear resistance and mechanical strength) are analysed, it can be indirectly concluded that in terms of geopolymer composite paving blocks, especially with TiO_2 nanoparticle content, further research is desirable, especially with a view to technological optimisation, so that the cost price can be reduced while maintaining the environmental benefits and an extended service life of 10-40%.

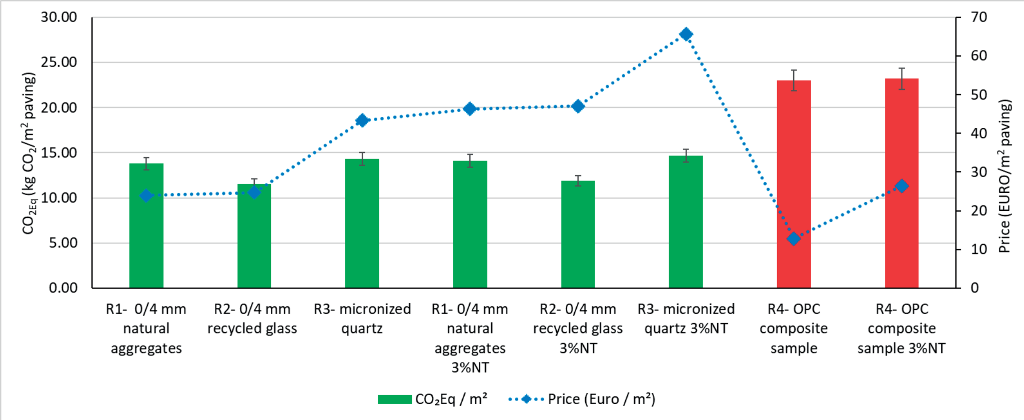

Figure 12. Price and CO_{2Eq} for the case of 1m^2 paving blocks produced using alkali-activated fly ash-based geopolymer vs. cementitious composite

CONCLUSIONS

The aim of this paper was to present preliminary results on the production of paving block elements produced using the concept of alkaline activation of fly ash, to present results on specific physical and mechanical characteristics and to highlight an LCA calculation of the resulting CO_{2Eq} emissions by comparison with similar elements produced using traditional materials (cement) and a cost-benefit analysis of the results obtained.

Conducting a Life Cycle Assessment (LCA) on geopolymer paving blocks provides valuable insights into the environmental performance of this sustainable construction material throughout its entire life cycle. The key conclusions drawn from such an assessment are:

- *Environmental benefits*: Geopolymer paving blocks exhibit the potential for significantly lower environmental impacts compared to conventional paving materials, particularly if they replace traditional concrete. The reduced carbon footprint is a notable advantage, as geopolymer binders typically require lower energy inputs and produce fewer greenhouse gas emissions during production.
- *Raw material savings*: Geopolymers often utilize industrial by-products such as fly ash or slag, diverting these materials from landfills and reducing the need for virgin raw materials. This aligns with principles of sustainable resource management and contributes to a circular economy by repurposing waste materials.
- *Energy efficiency:* The lower curing temperatures required for geopolymerization contribute to energy savings during the production phase compared to traditional cement-based materials. Reduced energy consumption in the manufacturing process leads to lower overall embodied energy in geopolymer paving blocks.
- *Durability and longevity*: Geopolymer paving blocks have shown promising mechanical properties and resistance to environmental factors, contributing to a longer service life compared to some traditional paving materials. Enhanced durability obtained using TiO_2 nanoparticles can result in reduced maintenance and replacement needs over time, further extending the overall life cycle of the paving blocks.
- *End-of-life considerations*: Geopolymer paving blocks may offer opportunities for end-of-life recycling or reuse, contributing to a more sustainable waste management strategy. Proper disposal methods, such as recycling or utilizing the blocks in other construction applications, can minimize the environmental impact associated with their end-of-life phase.
- *Challenges and opportunities for improvement:* The variability in the quality and availability of raw materials, as well as variations in production processes, can impact

the environmental performance of geopolymer paving blocks.

Ongoing research and development are essential to optimize production processes, improve material consistency, and address any potential challenges associated with the use of geopolymers.

In conclusion, a Life Cycle Assessment of geopolymer paving blocks underscores their potential as a sustainable alternative in the construction industry. While they show positive environmental attributes, ongoing research, standardization, and collaboration within the industry are necessary to optimize their production processes and further enhance their overall sustainability. Additionally, collaboration between researchers, industry professionals, and policymakers is vital to promoting the widespread adoption of geopolymer paving blocks as a sustainable construction solution.

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