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Use of Soda-lime Glass Microparticles as Supplementary Cementitious Material on the properties of Portland Cement Systems: Review

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ABSTRACT
This research aims to carry out a study of the state of the art on the influence of the use of soda-lime glass microparticles as supplementary cementitious material (SCM) on the properties of Portland cement systems. The methodology consisted of making a review of the current state of the art of recent works that approached the use of glass as SCM, in a theoretical or experimental way. As main results, the works show that glass and SCM are promising mainly due to their pozzolanic activity, microfiler effect and mitigation of the Alkali-Silica Reaction (ASR), which positively impact its mechanical properties and durability. As a conclusion, it can be seen that the particle size of glass is a fundamental parameter for it to be incorporated efficiently in Portland cement systems, allowing higher levels of substitution of cement for glass, which brings environmental benefits by reducing consumption of cement and the high CO_2 emission rates linked to it, and for giving a new destination to the high volume of glass waste that is not recycled.

Keywords – Portland Cement, Pozzolanic Activity, Supplementary Cementitious Material, Waste Glass Powder.

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I. INTRODUCTION

Concrete is the most widely used building material in the world, due to its versatility, durability and favorable cost, with cement as the basic component of its composition. The use of cement is linked to a high environmental cost due to the high emissions of carbon dioxide (CO_2) generated during its production, where about half of CO_2 emissions originate from the calcination of $CaCO_3$ (calcium carbonate), while the remaining carbon is from the energy used during this process [1-3]. To produce a ton of cement, almost a ton of CO_2 is produced (5 to 8% contribution to global CO_2 emissions). The CO_2 is the main emitting source that contributes about 65% of global warming [3-4].

Climate change and the reduction of CO_2 emissions, as well as environmental pollution, the circular economy, and the recycling of urban waste are seen as important and interrelated topics in society and science today [5]. Due to climate change generated by carbon emissions, the European Union and several multinational companies have committed to achieving neutrality in carbon emissions by 2050. Due to this worldwide trend, the concrete construction industry faces a major challenge with regard to sustainability criteria [3]. The most promising techniques to promote the reduction of the environmental impacts of cement production include the use of co-products or by-products as partial cement replacements [6]. The reuse of recycled materials in the manufacture of concrete and mortar is a solution for the preservation of natural resources and sustainable development [7-8].

Replacing part of the cement in mortar and concrete with mineral additions is an alternative to reduce cement consumption and the environmental problems associated with its production. These substitutions must be made without damaging the properties of the cementitious composites, guaranteeing equal or even superior durability. Mineral additions can be classified as reactive or inert, pozzolans are a type of reactive mineral addition, formed mostly by silicon oxide (SiO_2) in amorphous phase. The pozzolans react with the portlandite ($Ca(OH)_2$) formed with the hydration of the C_2S and C_3S of the anhydrous cement, forming more Hydrated Calcium Silicates (C-S-H), which is the binding phase of the hydrated cement, that is, with the use of pozzolans, within a limit, it is possible to form more binder phase in pastes, mortars and concretes using less cement [6,9].

Pozzolans are reactive mineral additions, or even SCMs, as they are incorporated into the traditional process of civil construction, appear as an innovation to acquire beneficial characteristics to Portland cement products, as well as to minimize the environmental impacts generated in the industry's production chain. Portland cement [9]. By-products and by-products of the industry traditionally used as SCMs, such as fly ash, silica fume and granulated blast furnace slag, are not always available in all regions and due to costs related to transport they may become economically unviable, due to this it is done the development of local alternative SCMs is necessary [10].

Soda-lime packaging glass is a recurring post-consumer waste in urban centers, although the glass can theoretically be fully recycled, there are still limitations in terms of meeting the quality criteria for remanufacturing glass, in addition to related costs transportation that can often make this type of recycling unfeasible, and then, this large volume of solid waste will be sent to landfills, reducing their useful life. Glass is a material that presents in its composition levels close to 70% of amorphous silica, presenting pozzolanic activity when finely ground, using glass as a supplementary cementitious material (SCM) opens a way to reduce both the environmental impacts generated by consumption and production cement, but also generated by low glass recycling rates [2,10-11]. Glass has the potential to be widely used as SCM, for which it is necessary extensive evaluations on its functional properties and related to durability [3].

The construction industry, including the materials sector, is a conservative sector in terms of reliance on prescriptive specifications for materials [12]. The pros and cons of using glass in cement systems are still being discussed. It is estimated that in 2025, 7% of all the world's waste is made up of glass [13]. The use of glass powder as a substitute for cement can decrease the consumption of cement and greenhouse gases belonging to the production of the global cement industry. On the other hand, proper waste management is one of the most important environmental objectives [14].

Due to the potential of using glass as a pozzolan and the lack of technical specifications for this use, different studies have investigated the use of glass waste in Portland cement compounds as SCM. Given this context, this research aimed to bring the current state of the art of the influence of the use of soda-lime glass microparticles as supplementary cement material on the properties of Portland cement systems.

II. WASTE GLASS POWDER

Packaging glass (soda-lime) is a ceramic material, amorphous, rigid, brittle, transparent and easy to manufacture. It has a wide range of applications, ranging from containers, windows and lamps to optical devices, waveguides and laser action media [15]. The glass is made from sand rich in silica, calcium carbonate ($CaCO_3$) and sodium carbonate (Na2CO3), these raw materials are mixed and burned at temperatures reaching over 1450°C [16]. The density of the residual glass powder is close to 2.52 g/cm^3 [17].

Typically, this glass consists of 70% silica (SiO_2), the rest being mainly composed of soda (Na_2O) and lime (CaO), and may have small contributions from other oxides such as K_2O (potassium oxide) and Al_2O_3, alkaline atoms presented in a glass generally act as network modifiers, which reduces the degree of polymerization of polyhedra [18]. Figure 1 shows the schematic distribution in two dimensions of its main constituent atoms, in which the absence of periodicity in the distribution of the atoms can be noted, also characteristic of solids with some degree of disorder (amorphous).

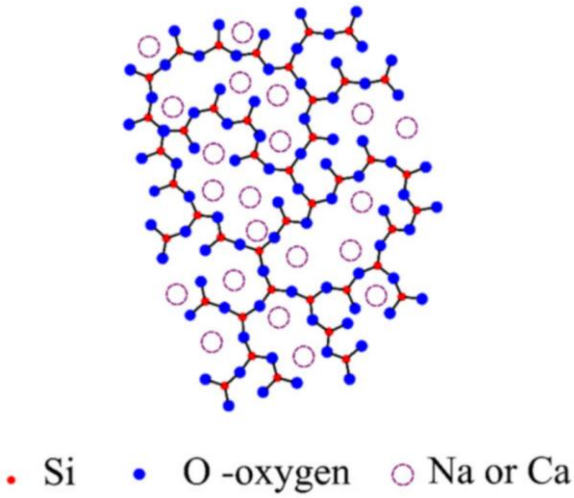

Fig. 1. Structure of Na-Ca silicate glass (Adapted from [18]).

To be used as supplementary cementitious material (SCM) the glass residue must be used in the particle size of the order of magnitude of the cement particles, in the form of microparticles (<45μm) [2]. In Fig. 2, there is a SEM image of glass microparticles, in which the irregular shape of the particles can be highlighted.

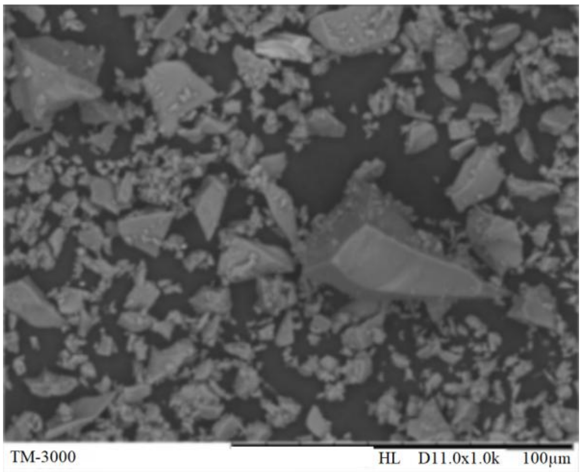

Fig. 2. SEM image of glass powder microparticles [19].

When glass is sprayed to microparticle size, due to its chemical composition rich in silica and the degree of disorder of its atomic structure, it can be used as Supplementary Cement Material, a substitute for cement in cement systems, the most common phenomenon. important in this substitution is the pozzolanic reaction of glass, which leads to the formation of a large amount of extra C–S–H in cement mixtures [20]. The amorphous structure of the glass allows it to be easily dissolved in a high alkalinity environment and to function as a pozzolanic material in cement systems [11]. The replacement of 20% of cement by microparticles of glass powder, generate a positive effect on hydration and microstructure of cement pastes [21].

The basic physical characteristics of GP particles, such as water absorption, particle size and morphology can greatly influence the properties of mortars and concretes. for example, the use of smaller glass particles improves the fluidity of Portland cement systems in the fresh state, as the particle dimensions tend to approximate each other, whereas large GP particles are irregularly shaped [11].

III. SUPPLEMENTARY CEMENTITIOUS MATERIALS (SCM) AND POZZOLANIC REACTION

i. SCM

Portland cement does not completely satisfy the needs of the concrete construction industry, SCM enable a wider range of applications for cement systems. Although natural pozzolans in the raw state or after thermal activation are still being used in some parts of the world, due to economic and environmental considerations, many industrial by-products have become the main source of mineral additives in concrete [22-23].

The use of urban, industrial and agricultural waste such as SCM can have an impact on a number of properties of fresh and cured concrete, depending on the quantity and properties of the materials used. One or more goals can be achieved with the use of mineral additives in fresh concrete, such as improving workability, reducing exudation and segregation, decreasing the hydration temperature, reducing the alkali-silica reaction, the significant pore refinement increases its electrical resistivity and resistance to chlorides and sulfates, reduces corrosion rates, improves the strength of the aggregate and concrete paste interfaces, increases electrical resistivity, produces low-cost and ecologically sustainable concrete [8,24].

The replacement of cement by SCM in concrete and mortar has an important role in today's engineering activities with regard to its impact on the environment and in the manufacture of low-cost concrete. The use of such SCM reduces carbon dioxide (CO_2) emissions and energy consumption, which in turn reduces the adverse impact of cement production on nature, while lowering concrete costs. However, studies should focus on improving the durability of concrete and exploring residual materials that can be used as mineral additives [8].

One of the main characteristics to be considered when assessing the potential application of a residue for the addition or replacement of a cement portion for the production of concrete refers to its ability to promote pozzolanic reactions [25].

ii. Pozzolanic Reaction

The term pozzolana refers to the city of Pozzuoli, in Italy, pozzolana was discovered by the Romans, the ancient Romans realized that those natural pozzolans had cementing properties when mixed with lime and water, and started using them to make building blocks . The cement characteristics are known as pozzolanic properties that depend on the amount of reactive SiO_2 and Al_2O_3 available in the aluminosilicate [26]. The products of the pozzolanic reaction are similar to those of the Portland Cement reaction including calcium silicate hydrates (C-S-H) and calcium aluminate silicate (C-A-S-H) hydrates [27].

A pozzolana is defined as a siliceous or siliceous and aluminous material, which by itself has little or no cementing properties, but when presented in a finely divided form and in the presence of moisture, they react with calcium hydroxide (Ca(OH)2), CH, produced during the hydration of Portland cement, as a result, there is the production of chemical phases with greater stability and binding power, being the main responsible for the resistance of hydrated cement pastes. The reaction between SCM with pozzolanic activity and the phases formed after cement hydration generally occurs according to the mechanism of Eq. (1) and Eq. (2) [[22,25,28].

- Cement hydration (fast reaction):

Portland Cement + Water ⇒ C−S−H + $Ca(OH)_2$ (1)

- Pozzolanic reaction (slow reaction):

$Ca(OH)_2$ + Pozzolanic material + Water ⇒ C−S−H (2)

The pozzolanic activity originates from the thermodynamic instability that exists when these materials come into contact with CH saturated water, given that the silica and alumina found in pozzolana belong to easily displaceable structures (amorphous or disordered structures) [29]. Pozzolanic reactions continue to occur as long as calcium hydroxide is present and in contact with amorphous silica, there is enough water for the reaction to occur and there is space available for the reaction products to reside [30].

Calcium hydroxide crystals (also called portalndite) make up 20 to 25% of the volume of solids in the hydrated cement paste, and are formed by the hydration of C_3S and C_2S. Unlike C-S-H, calcium hydroxide is a compound with a defined stoichiometry, $Ca(OH)_2$, which tends to cause the formation of large crystals with a distinct hexagonal morphology. The extra C-S-H formed by the consumption of portlandite in the pozzolanic reaction, is technologically advantageous because it increases the amount of binder phase present in the paste, using the same amount, or even less cement [23].

This reactivity enables cementitious materials to perform well. In addition to the presence of silica, the pozzolanic properties are also related to the specific surface area of the material, particle size, degree of dehydroxylation and quality of the raw materials. Also, the greater reactivity is due to the presence of considerable amounts of amorphous minerals, while materials with a high content of crystalline minerals have low pozzolanic reactivity [25].

The addition of high levels of pozzolans such as glass is disadvantageous with regard to carbonation. The consumption of Calcium Hydroxide in the pozzolanic reaction is quite high, this high consumption of CH decreases the amount of carbonable products and reduces the pH of the pore water, increasing the carbonation rates [11,24].

IV. MICROFILER EFFECT, POZZOLANIC REACTION AND ARS MITIGATION OF GP

The use of glass residue powder in cement systems affects fresh and hardened properties, hydration products, microstructures and durability, compared to traditional raw materials, however, there are still some doubts about durability and long-term performance of these materials [18].

i. Microfiler Effect of GP

The use of glass microparticles in cement systems brings an initial benefit to the system by filling the gaps in the hydrated cement matrix, which ends up leaving the microstructure of the Cement-Microparticle glass system dense and thick. The smaller the size of the glass microparticles, the greater their microfiller effect, and the more noticeable the physical filling effect, which makes pores less numerous and smaller, reduces connectivity between pores and as a consequence, greater densification occurs. matrix, increased mechanical properties, reduced permeability and transport of harmful agents, which directly impacts the system's durability, microparticles with microfiller effect are fundamental for the manufacture of Ultra High-Performance Concretes [10-11, 31-34].

The penetration of sodium chloride and sulfate ions can be reduced with the inclusion of GP as a cement replacement. This can be attributed to the refinement of the pores by the microfiler effect and the pozzolanic reaction, which decreased the porosity and the connectivity of the pores for the transport of chlorides and sulfides [11].

ii. Pozzolanic Reaction of GP

Recent studies have shown promising potential for the use of Glass Pozzolan (GP) as an alternative to supplementary cementitious material (SCM) due to the scarcity of fly ash and slag in the United States [3]. The pozzolanic reaction is drastically increased by reducing the size of glass particles, allowing higher levels of incorporation, by maintaining the fluidity of cement mixtures in their fresh state [11].

The smaller the average diameter of the glass particles and the greater their specific surface, the better the mechanical results of the samples that replaced the cement with the glass, due to the microfiller effect at earlier ages and the greater reactivity of the glass to react with the portlandite. forming (C-S-H) and (C-A-S-H) gels at later ages. However, the use of glass particles with a smaller average diameter causes pozzolanic activity to manifest at an early age [31-35]. The increase in pozzolanic reactivity is directly proportional to the reduction in the size of the glass particle, the use of smaller particles promotes greater formation of C-S-H and greater consumption of portlandite. The use of glass also promotes an improvement in cement hydration, as there is more water left for the cement particles [11,31].

Most of the finer particles of glass react to form C-S-H, whereas, in the case of larger particles, the reaction is slowed down gradually, in which the inner part of the grain ends up not reacting, and the unreacted grain is left with a ring of C-S-H at the end. around you [36]. The amorphous phases can be inferred from chemical and mineralogical characterization techniques, as is the case with X-ray Fluorescence (XRF), which identifies oxides of the elements that constitute the samples, associated with the X-ray diffraction technique (XRD), which identifies the crystalline phases of the material. The particle size distribution can be given from the granulometric analysis via sieving and sedimentation, or via laser granulometry, which is able to provide more reliable results for finer particles [25].

Microparticles can also play the role of “nucleation induction” and promote better cement hydration as can be seen in Fig 3 [21, 34, 37]. SEM image of a mortar that replaced 30% of the cement with glass microparticles, where the germination of C-S-H on the glass particles is observed. It is also possible to observe the absence of portlandite phase and pores/voids in relation to the reference sample. The better mechanical properties of samples containing glass were explained by the pozzolanic reaction and the microfiller effect that the glass particles generated in the system [37].

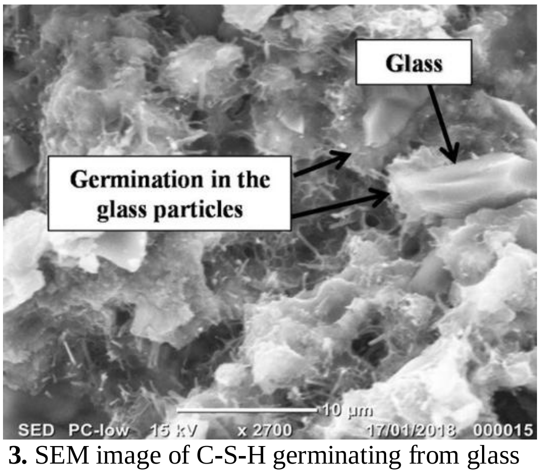

Fig. 3. SEM image of C-S-H germinating from glass microparticles in a mortar containing 30% replacement of cement by glass after 90 days of curing [37].

The reaction of C-S-H formed by the pozzolanic reaction of glass microparticles with portlandite happens slowly, to better understand this reaction. Mejdi et al. [36] made a composite containing only portlandite and glass powder microparticles, this composite glass/portlandite system was designed to reduce the complexity of the Portland Cement system that has several compounds. Through XRD analysis, as can be seen in Fig. 4, it can be seen that in a system containing 75% glass microparticles and 25% portlandite, portlandite was almost completely consumed by the pozzolanic reaction in the 91-day period, due to the expressive reduction of the peaks referring to portlandita and the appearance and growth of the peaks referring to C-S-H.

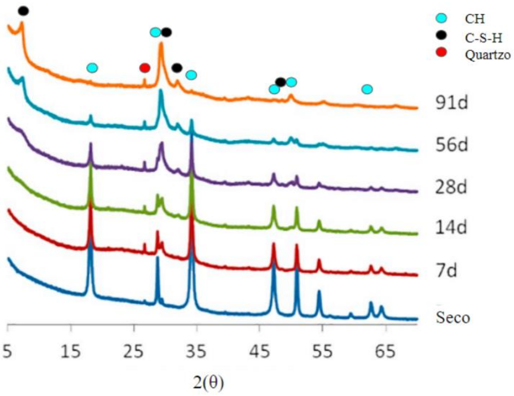

Fig. 4. Crystalline phase evolution of the system containing 75% glass powder with 25% portalndite over time after its hydration (Adapted from [36]).

The formation of C-S-H around the glass particles can be seen in the SEM image with EDS graphs in Fig. 5, where it can be seen that between the C-S-H and the glass particle there is a compound of intermediate composition [36]. The microstructure of silica-rich C-S-H, formed by pozzolanic reactions, is more resistant and less porous, the glass microparticles generate a dense edge of C-S-H gel around the larger glass particles and densifies the cementitious matrix, which generates greater resistance of matrix in the aggregate paste transition zone, this effect is more noticeable in later ages [38].

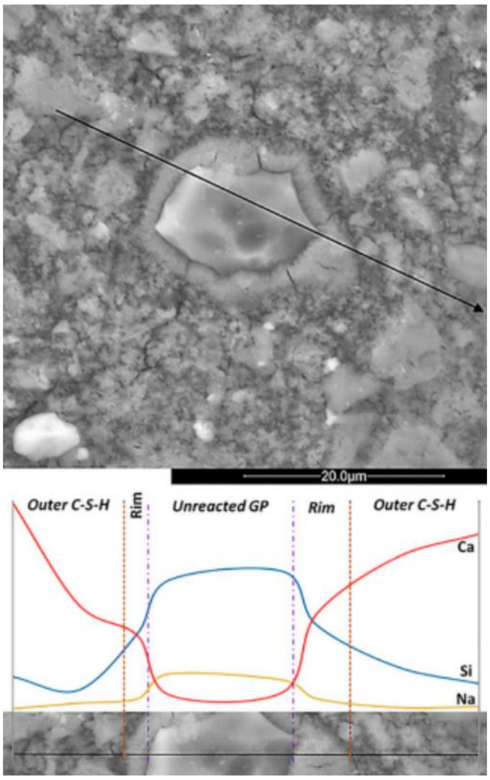

Fig. 5. EDS lines showing the distribution of the elements Ca, Si and Na around the glass particle, reaction of the glass with the portlandite (Adapted from [36]).

Refined pore structure for greater replacement of cement by 40% GP, indicating greater gel formation (C-S-H). Following the criterion of consumption of portalndite to the glass residue can be considered as a more pozzolanic addition than blast furnace slag and a fly ash [3].

iii. Use of Glass Microparticles for ARS mitigation

The alkali-silica reaction (ASR) is a problem of concrete durability, the reaction products generate swelling in the concrete by varying the volume, which is a deleterious behavior, as it ends up causing expansion, cracking and, consequently, shortening the service life of the concrete. concrete. ARS products are formed by chemical reactions between the alkaline solution of the pores of cement systems and amorphous or metastable forms of silica in aggregates. Based on the chemical composition of the ASR products reported in a series of studies, it is clear that the presence of reactive silica, alkalis and some calcium are essential conditions for ASR. Due to the slow formation of ASR products, damage to concrete structures caused by ASR is generally observed after decades and is difficult to predict. Most ASR mitigation approaches are based on the design of initial mixtures, such as by the appropriate use of low alkaline cements and/or incorporation of SCMs with pozzolanic activity, during the manufacture of concrete [39].

The high alkali content in soda-lime glass suggests that glass may increase the total alkali in cement systems, leading to the development of significant ASR, which actually occurs when used as an aggregate replacement in concrete or mortar, but when glass is used as SCM in the form of glass microparticles (<90µm), ASR is reduced or eliminated due to the development of pozzolanic hydrates. The smaller the size of the glass microparticle used, the greater the mitigation effect of ASR [4,18,38-41].

Some studies have shown that glass particles larger than 1mm make concrete more prone to damage induced by ASR [17], but finely ground glass particles did not contribute to the harmful expansion due to ASR, and help mitigate the ASR. The mass substitution of 30% of cement with glass powder microparticles (with an average dimension of 1µm and Blain fineness of 800m2/kg), led to a 52% mitigation in ASR in the research of Hendi et al. [41], as can be seen in the Fig 6. The use of glass particles is more effective to mitigate ASR in cement systems.

One explanation was that the secondary C-S-H generated by the pozzolanic reaction had a lower Ca/Si ratio, exhibiting a better ability to incorporate alkalis and inhibit ASR expansion. Another explanation is that the dissolution of GP could increase the concentrations of Si and Na in the pore solutions that led to the decomposition of the monosulfate, resulting in the continuous release of Ca, Al and Si [11].

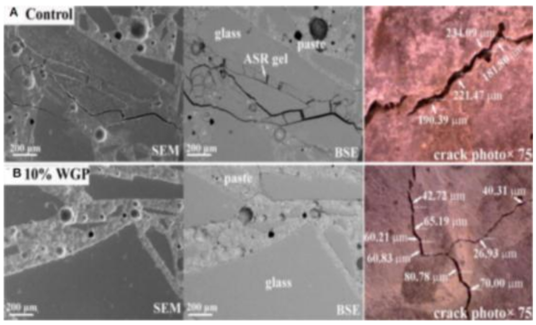

Fig. 6. SEM images (left), BSE images (middle) and photos of cracks (right) of mortars after being subjected to the accelerated ASR test for (1d + 28d) ((a): control; (b): 10% WGP [39].

Monitoring the composition of the pore fluid revealed that the GP, over time, acts as a net contributor to the alkalinity of the pore solution, but these alkalis are released slowly. Overall, SLGP can reduce ASR, albeit insufficiently and temporarily. This reduction in ASR is likely through a combination of reduced pH of the pore solution at early ages (due to the dilution of cement) and reduced portlandite content and transport properties at later ages (due to the pozzolanic reaction).

It was found that the reaction products formed with high Na/Si and Ca/Si ratios were favorable to mitigate the expansion of ASR. For the sample prepared with WGP, increasing the Ca/Si ratio would increase the stiffness of the ASR gel, and a higher Na/Si ratio would help to reduce the osmotic pressure. The findings of this study would be useful for the selection of pozzolans to mitigate the effect of ASR when using crushed glass shards as aggregate in cement mortars [39].

V. CONCLUSION

The growing environmental and economic pressures on the cement industry and the increasingly stringent performance requirements for concrete/mortars/cement will continue to drive the need for binders with additions in their formulations, making them more complex in their variety, chemical and mineralogical composition. and structural, so there is a need to better understand and predict the behavior that such additions will have when they are incorporated on a large scale in the future, as in the case of the use of glass microparticles as SCM.

The particle size of the glass is an important parameter for its incorporation in cement systems, several recent works have demonstrated a direct correlation between the reduction of the used particle size and the increase of the surface area of the particles, with the increase of the pozzolanic activity and its manifestation at earlier ages, improvement of the microfiler effect and the refinement of pores, densification of the cementitious matrix, improvement of the aggregate/paste interface, by filling empty spaces with C-S-H, improvements in the resistance to compression and flexion, mitigation and neutralization of expansion by ARS. The use of smaller particles of glass still allows higher levels of substitution of cement for glass, bringing better results. Due to the consumption of portlandite, Portland cement systems that use glass as SCM, carbonation is unprotected by the lack of calcium reserve, which can be compensated by the increase in density, reduced connectivity between the pores and the impermeability that the glass itself generates.

The use of glass as a pozzolana brings environmental benefits by reducing the consumption of cement and the high levels of CO_2 emissions, by giving a new destination to the high volume of glass waste generated by urban and industrial centers, and technology. The use of glass or any new SCM requires extensive testing to assess the parameters that the glass must have in order to maintain good long-term performance in a Portland cement system.

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