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ORIGINAL ARTICLE

Evaluation of glass powder in the mitigation of the alkali-silica reaction (ASR)

Avaliação do pó de vidro na mitigação da reação álcali-sílica (RAS)

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Abstract: The alkali-silica reaction (ASR) is one of the problems that can compromise concrete durability, as the chemical reactions between the reactive aggregate and concrete pore solution, specifically sodium (Na^+), potassium (K^+), calcium (Ca^{2+}) and hydroxyl (OH^-) in the presence of moisture, trigger expansion and cracking of concrete elements. The use of pozzolanic materials, such as finely ground glass powder (particles smaller than 75μm), to partially replace cement has been widely studied and has demonstrated beneficial results. However, because the behavior of pozzolans towards ASR is not completely understood yet, this work evaluates the effect of incorporating ground glass powder into cementitious composites against ASR. To this end, 8% silica fume was used as a control admixture, and four levels of glass powder (10, 20, 30, and 50%) were incorporated into the cementitious mixture to partially replace cement. Expansion tests were carried out in accordance with NBR15577-4 (ABNT, 2018) on mortar bars using the accelerated test method to assess the mitigation of expansions. The results show that the expansions caused by ASR decreased with the increasing content of glass powder in the cement mixtures. Furthermore, only the 50% level reduced the expansions to a safe value according to the Brazilian standard.

Keywords: alkali-aggregate reaction, mitigation, pozzolanic materials, glass powder.

Resumo: A reação álcali-sílica (RAS) é um dos problemas que pode comprometer a durabilidade do concreto, uma vez que as reações químicas entre o agregado reativo e a solução porosa do concreto, em particular, sódio (Na^+), potássio (K^+), cálcio (Ca^{2+}) e hidroxila (OH^-) na presença de umidade, desencadeiam a expansão e fissuração dos elementos de concreto. O uso de materiais pozolânicos, como o pó de vidro finamente moído (com partículas abaixo de 75μm), para substituir parcialmente o cimento, tem sido amplamente estudado e demonstrou resultados benéficos. No entanto, devido ao fato de que o comportamento das pozolanas em relação à RAS ainda não é completamente compreendido, este trabalho avalia o efeito da incorporação de pó de vidro moído em compósitos cimentícios frente a RAS. Para isso, 8% de sílica ativa como controle, e quatro teores de pó de vidro (10, 20, 30 e 50%) foram incorporados à mistura cimentícia em substituição parcial ao cimento. Testes de expansão foram realizados de acordo com a NBR15577-4 (ABNT, 2018) em barras de argamassa usando o método de teste acelerado para avaliar a mitigação das expansões. Os resultados mostram que as expansões causadas pela RAS diminuíram com o aumento do teor de pó de vidro das misturas de cimento. Além disso, apenas o nível de 50% reduziu as expansões a um valor seguro de acordo com a norma brasileira.

Palavras-chave: reação álcali-agregado, mitigação, materiais pozolânicos, pó de vidro.

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1 INTRODUCTION

The civil construction production chain massively consumes cement-based composites. In 2020, the cement industry sector reached sales of 60.8 million tons in Brazil, marking a 10.9% increase compared to 2019 [1]. Despite the strong representativeness of the segment in the economic and social development of the country, the cement production process is responsible for 7% of the global CO_2 emission worldwide [2].

The cement industry is concerned and moving toward sustainable management in the civil construction sector. This movement has led to an effort and interest in the scientific-technical community to conduct several studies aimed at a partially adding or replacing the elements that make up the cementitious matrices, especially cement.

Ground glass is among the alternative supplementary cementitious materials recently explored. Glass is formed by amorphous silica, and when finely ground, it behaves as a pozzolan. In the presence of water, it reacts chemically with the calcium hydroxide, forming compounds with binding properties [3]–[6].

Furthermore, according to Robert et al. [7], in the global market, 130 million tons (Mt) of glass are produced annually. The global volume of recycled glass is estimated to be around 27 Mt, which represents only 21% of the glass produced annually.

According to data released by the Brazilian Technical Association of Automatic Glass Industries (ABRAVIDRO) [1], in 2020 the production of glass packaging in Brazil totalled around 1.4 Mt, placed on the market in various formats and accounting for approximately R$ 120 million. Importantly, this number may fluctuate over time depending on several factors, such as market demand and changes in the industry.

Yet, according to data from ABRAVIDRO [1], in 2020, the recycling rate of glass containers in Brazil was approximately 50%. This means that half of the glass containers used in the country was recycled that year. This value represents a recycling percentage greater than the global average for Brazil, but it still indicates a low rate, as the other half of the glass produced goes to landfills and dumps.

According to the association, initiatives to reuse the material are still insignificant compared to the amount produced annually because of the low cost of production from the raw material and the low added value of this product for collectors and recycling cooperatives, making glass a material with little investment that encourages its recycling.

Another factor pointed out by Mirzahosseini and Riding [8] is that glasses of different colors are often mixed when collected. Mixed-colored glass cannot be recycled because a mixture of different coloring agents results in unpredictable and uncontrollable coloring and properties in the new glass. Some machines can use optical sensors to efficiently classify the color of large pieces of glass. However, sorting small pieces is not feasible nor cost-effective. The concrete industry is one of the possible clients to reuse millions of tons of glass cullet per year as a supplementary cementitious material.

Another pertinent issue is that in some places in Brazil, there are no glass recycling plants. The city of Manaus, for example, had only 1% of its garbage recycled in 2016, according to Nascimento et al. [9], with a significant portion discarded in the Manaus landfill, including glass. According to the authors, the difficulties in processing glass recycling in Manaus are numerous, including the lack of specific public policy for the material classification, cost, collection and separation processes, decontamination, and energy source.

When added to cementitious matrices, the glass powder acts as a filler due to the mechanical effect of filling the voids and as a binder due to the binding effect [4], [5], [10]–[12].

Wilson et al. [13] studied the filler effect of the Ultra-High-Performance Concrete (UHPC) by replacing the initial cement consumption of 556 kg/m^3 with 30% bulk glass powder with particle size d_{50} (12 µm). Subsequently, they analyzed the results using the NI-QEDS technique (NanoIndentation and Quantitative Energy-Dispersive Spectroscopy). The chemical relationships and mechanical properties of the specimens and reference samples were remarkably similar, and the authors concluded that ground glass acted as an inert material.

Despite the proven filler and pozzolanic effects of the glass powder, the high quantity of alkalis (12 to 17% Na_2O) usually present in glass has become a concern for the scientific community, as such concentrations can trigger the Alkali-Aggregate Reaction (AAR) in cementitious matrices. However, Shayan and Xu [14]; Bhandari and Tajne [15]; Kamali and Ghahremaninezhad [10], and Mehta and Ashish [16] showed that the glass powder does not induce the Alkali-Aggregate Reaction (AAR) in cementitious composites, as long as the particle size is equal to or less than 75 µm. Therefore, the performance of glass powder depends especially on particle size [10], [16].

The Alkali Aggregate Reaction (AAR) is the chemical reaction that occurs in cementitious matrices, involving hydroxyl ions (OH^-) associated with the alkaline sodium and potassium components originating from Portland cement or other sources and reactive aggregates. In the presence of water, this reaction causes deleterious expansion. The alkali-aggregate reaction has two main forms: the Alkali-Carbonate Reaction (ACR) and the Alkali-Silica Reaction (ASR). Currently, the alkali-silicate reaction is recognized as a slow type of alkali-silica reaction [17].

The Alkali-Carbonate Reaction (ACR) and Alkali-Silica Reaction (ASR) are distinct chemical processes that can compromise the integrity of concrete structures. ACR involves the interaction between alkaline ions in cement and dolomite in aggregates, resulting in the formation of alkaline carbonates. This reaction can lead to concrete expansion, cracking, and reduced durability. On the other hand, ASR occurs when the alkaline ions in the cement react with reactive silica minerals in the aggregates, producing a gel-like substance. This gel absorbs water, causing swelling, increased pressure, and similar detrimental effects on the structural integrity of the concrete [18], [19].

ASR is considered a common occurrence that develops rapidly [20]. According to Mohammadi et al. [21], ASR affects the concrete compression strength, the tensile strength, and the flexural capacity, as well as the modulus of elasticity. The increasing ASR causes these properties to decrease, depending on different factors, such as the reactivity degree and classification of the aggregates, and the properties of the harmful gel. The authors reported that the degradation of concrete modulus of elasticity is highly dependable on the ASR, even at early ages.

The ABNT NBR 15577 [17] is the Brazilian code that presents robust concepts on this phenomenon. For mitigating or preventing AAR, the standard stipulates five stages as follows: classifying the structure, determining the aggregate reactivity degree, crossing this information with the structure dimensions, and exposure degree.

To evaluate the alkali-silica reaction, Kamali and Ghahremaninezhad [10] investigated concrete specimens with 10% and 20% replacement levels, i.e., glass powder incorporated to replace cement. The two types of ground glass used were an industrial by-product derived from fiberglass (V1) and post-consumption recycled glass (V2), with an average particle size close to 8.4 µm. Following the ASTM C1260 [22] accelerated mortar bar test method, a marked expansion of the control mortar was observed during the 15-day testing period. The ASR decreased in specimens with incorporated glass. Regardless of the ground glass type, the expansion results show that the ASR was mitigated most effectively in specimens with 20% incorporated glass powder.

Shafaatian et al. [23] proved that the ASR mitigation in cementitious composites with supplementary materials results from the reduced concentration of alkalis and hydroxides in solution in the concrete pores due to the pozzolanic reaction and dilution of alkalis resulting from partial cement replacement. The C-S-H with low calcium content produced by the pozzolanic reaction binds to the alkalis in the interstitial solution while reducing the alkalis available for the ASR. The authors also stated that the microstructure densification generated from the pozzolanic reaction reduces ion transport, contributing to the reduction of alkalis in the pore solution and mitigating the ASR.

This paper reports on an experimental campaign aimed at evaluating the effect of incorporated glass powder on ASR mitigation. Glass powder passing through the #200 sieve (75µm opening) was added to the cementitious mixture at four replacement levels (10, 20, 30, and 50%) to compare the expansions caused by an ASR reactive aggregate. An additional mortar mixture with 8% silica fume was evaluated as a control test.

2 MATERIALS

The materials used to assess the mitigation of the expansion caused by the ASR were Portland cement, silica fume, glass powder, and reactive aggregate.

The Portland cement used was Type III according to ASTM C 150 (Standard specification for Portland cement) [24], with a density of 3.12 g/cm^3, a specific surface area of (490 ± 20) m^2/kg, 0,85%Na_2O_{eq}, and autoclave expansion according to ASTM C 151 (Standard Test Method for Autoclave Expansion of Hydraulic Cement) [25] of 0.10%.

The silica fume has a density of 3.50 g/cm^3 and a specific surface area of (2470 ± 10) m^2/kg.

The glass powder used was the #200 (75 µm opening) with a density of 2.55 g/cm^3 and a specific surface area of (393 ± 10) m^2/kg. The soda-lime glass powder used was obtained from processing recycled, amber-colored beverage bottles. After washing to remove the labels and glue, the bottles were processed in a concrete mixer with steel balls. Subsequently, the broken glass was ground in a ball mill, covered with flint and silicate sedimentary rock, and grinding balls of the same material. This process is shown in Figure 1. The final screening was performed in the Materials and Components Laboratory (LMC) of the Federal University of São Carlos, in São Carlos, SP, Brazil. Initially, the glass powder was dried in an oven at 110 ± 5 °C for 24 h, then sieved through a Ro-Tap W. S. TYLER vibrating screen.

Figure 1. Process for obtaining the glass powder. Source: Freitas et al. [26].

The particle sizes of the glass powder and the silica fume were determined by laser granulometer, and the curves obtained by the test can be seen in Figure 2.

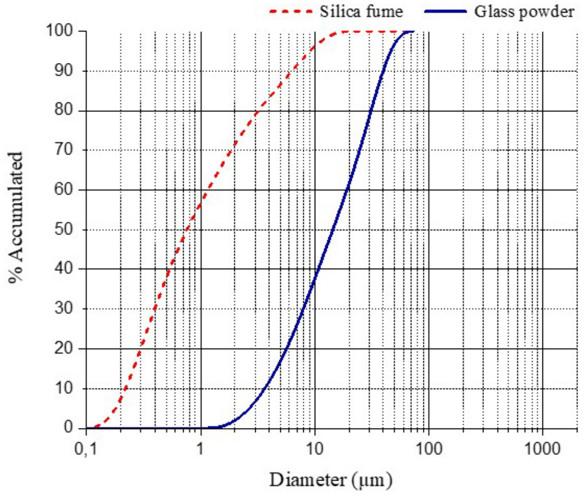

Figure 2. Particle size-distribution of the glass powder and silica fume.

Analyzing the particle size-distribution curves of the glass powder and the silica fume, it is observed that the average diameter (d_{50}) of the glass powder is 14 µm, and the average diameter (d_{50}) of the silica fume is around 0.8 µm.

A highly ASR aggregate was used in this study to induce ASR, and the glass powder served as the mitigating agent to analyze the mitigation of expansion caused by these aggregates. The X-ray diffractometry-based mineralogical characterization studies unveiled the predominant composition of the aggregate sample, which primarily comprises calcite ($CaCO_3$), dolomite ($CaMg(CO_3)_2$), quartz (SiO_2) and secondarily by mica ($KAl_2(AlSi_3O_{10})(OH)_8$) and feldspar ((K, Na)[AlSi3O_8]- $CaAlSi_2O_8$). Based on the results of the chemical analyses, it is estimated that the limestone rock consists of about 60% calcite, 22% dolomite, and 18% other minerals (quartz, mica, and feldspar). In Figure 3, the diffractogram of the reactive aggregate is depicted.

Arranged in a clastic texture, these minerals collectively categorize the rock as limestone. Through the petrographic study, the sample was considered potentially reactive due to the presence of micro-granular quartz (< 1 mm), comprising 1 to 5% of the sample's total constituents.

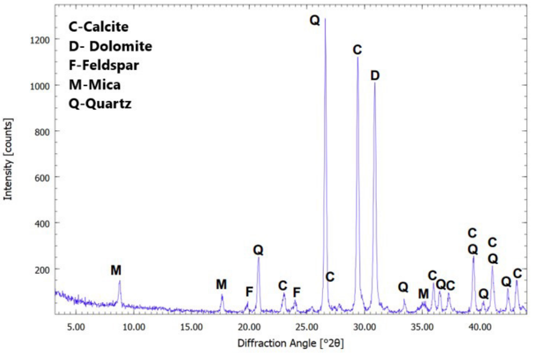

Figure 3. Diffractogram of the reactive aggregate.

The chemical composition of these materials was determined by X-ray fluorescence spectrometry (XRF) and is shown in Table 1.

Table 1 - Chemical composition of the used glass powder and silica fume.

Components	Values (% mass)			
	Portland cement	Glass powder	Silica fume	Reactive aggregate
SiO_2	18.76	74.00	94.10	10.94
Al_2O_3	4.15	3.70	< 0.20	1.67
Fe_2O_3	3.56	0.42	< 0.50	1.31
CaO	63.00	9.10	< 0.20	42.54
MgO	0.51	0.74	-	4.70
SO_3	2.79	-	-	0.09
CO_2	4.72	-	-	36.64
Na_2O	0.32	11.00	< 0.20	0.01
K_2O	0.81	0.56	1.28	0.33
Others	1.38	<0.50	1.72	1.77
LOI*	5.89	0.58	3.60	38.16
IR**	1.34	0.02	-	14.54

*Loss on ignition (LOI) and **Insoluble residue (IR)

3 METHOD

Tests for determining the expansion of mortar bars and tests for determining the mitigation of expansion in mortar bars were carried out in the laboratory of the Brazilian Portland Cement Association (ABCP) in the city of São Paulo, Brazil, and are described below.

3.1 Pozzolanic Activity of Glass Powder

The pozzolanic activity was determined by the modified Chapelle Method following the ABNT NBR 15895 [27] standard method, which allows quantitatively evaluating the pozzolanic activity of a given material by determining the fixed calcium oxide content.

The method consists of keeping a solution containing 2.0 g calcium oxide, 1.0 g pozzolanic material, and 250.0 g water at 90 °C, stirring for 16 h. After cooling, a sucrose solution is added and stirred for another 15 min.

After filtering approximately 100 mL, a 50 mL aliquot is titrated with 0.1 N HCl using a phenolphthalein solution. The amount of CaO fixed by the pozzolanic material is calculated. The result is expressed in $Ca(OH)_2$mg fixed per g of pozzolanic material.

A pozzolanic material must have a minimum consumption index of 330 mg CaO/g sample (436 mg $Ca(OH)_2$/g sample).

Additional tests were carried out to determine the pozzolanic activity of the glass powder. Performance was determined with Portland cement at 28 days, according to ABNT NBR 5752 [28] and ASTM C618-05 [29].

Portland CP II-F-32 cement, sand, water, and glass powder were mixed to produce two mortars mixes: a reference mix (without the addition of pozzolan) and the studied mix, which include glass powder as a cement replacement.

For each mortar type, six cylindrical specimens of 50 mm x 100 mm were molded and subjected to the compressive strength test after 28 days. After curing inside the molds for 24 hours in a moist chamber, the specimens were demolded cured in water saturated with lime according to ABNT NBR 7215: 2019 [30].

3.2 Accelerated mortar bar test

The expansion of mortar bars using reactive aggregate samples was determined by the accelerated method, following the ABNT NBR 15577-4 [31] test method. The dimensional variation of the bars was determined using a dimensional variation analysis frame for 28 days, as established in the standard, Figure 4.

Figure 4. Measuring the dimensional variation of the specimens.

The ABNT NBR 15577 [31] establishes that mortar expansion greater than or equal to ≥ 0.19% indicates that the aggregate is potentially reactive, as shown in Table 2.

Table 2. Aggregate reactivity according to NBR 15577 (ABNT, 2018).

Potential reactivity class of the aggregate	Expansion of mortar bars at 30 days - %
Potentially harmless grade R0	Less than 0.19%
Potentially reactive grade R1	Between 0.19 and 0.40%
Potentially reactive grade R2	Between 0.41 and 0.60%
Potentially reactive grade R3	Greater than 0.60%

All bars' specimens were molded within 2 minutes and 15 seconds, from the moment the mortar was mixed until the molding of the third specimen. After 24 hours in the mold, the bars were demolded, identified, and kept in a container with deionized water in a ventilated oven at 80 ± 2 ºC for another 24 hours. After that, the initial zero reading of the specimens was performed, followed by another immersion in a tank with 1N NaOH solution, at 80 ± 2 ºC.

Measurements were carried out at 16 and 28 days and at least six intermediate ages in between; all readings were performed at approximately the same time of the day. The ABNT NBR 15577 [31] categorizes the aggregate reactivity into four classes according to the expansion percentage of the mortar bars at 28 days (30 days since molding).

3.3 Determining the expansion mitigation in mortar bars

The ASR mitigation was measured by the test of expansion mitigation of mortar bars using the accelerated method, following the ABNT NBR 15577-5 [31]. The test procedures followed the ABNT NBR 15577-4 [31] standard described in item 3.2. The standard used for this testing step has the same principle as part 4, but it is indicated to evaluate how efficiently cement, with added pozzolanic materials or blast furnace slag, inhibits the expansion resulting from the ASR.

The specimens tested were produced with reactive aggregate, Portland cement type III, glass powder, and silica fume. The replacement of cement with glass powder was done by mass in the proportions of 10, 20, 30, and 50%, and another with added 8% silica fume instead of the glass powder.

4 RESULTS AND DISCUSSION

4.1 Pozzolanic Activity of Glass Powder

Fine glass powder particles (less than 75 µm) have pozzolanic properties and can be used as a cementitious material to replace Portland cement [32]–[35]. This statement was confirmed in this study since the material evaluated by the modified Chapelle methodology had a minimum consumption index of CaO/g sample ($Ca(OH)_2$/g sample) higher than the NBR 15895 [27] requirement and a higher value also due to the methodology of the Performance Index with Portland Cement according to ASTM C 618-05 [29]. Table 3 shows the results.

Table 3. Result of the glass powder analysis by the modified Chapelle test.

Methodology	Values	Standard limit
Modified Chapelle NBR 15895 [27]	1319	> 436 mg $Ca(OH)_2$/g sample
Performance Index with Portland cement ABNT NBR 5752 [28]	84%	> 90%
Performance Index with Portland cement ASTM C 618-05 [29]	84%	> 75%

Evaluating the performance index of the specimens at 28 days, molded with Portland cement, the samples did not reach the performance index of 90%, a value required by ABNT NBR 5752 [28] for pozzolanic materials.

However, the American standard of ASTM C618-05 [29] requires a minimum performance index of 75% to classify a material as pozzolana, with the material in this study reaching this index.

Liu et al. [36] reported that adding glass powder decreased the compressive strength of mortar samples compared to the control mortar sample (only with Portland cement), but, at the same time, the added glass powder accelerated the hydration process, forming more CSH gel, which, in turn, plays a fundamental role in the pozzolanic behavior of glass powder.

Similarly, Patel et al. [37] analyzed the pozzolanic activity index of the cementitious material containing cement and glass powder with 100% particles passing the #200 sieve (75 µm opening) by obtaining a relationship between the mortar compressive strength, the cementitious material to be analyzed, and the compressive strength of the reference mortar. These authors reported that values ranged from 76 to 96%, above the 75% limit required by the ASTM C618-05 [29] for pozzolanic materials.

4.2 Determining the expansion in mortar bars

The potential reactivity of the aggregate was determined by the expansion of mortar bars using the accelerated method. The aggregate was mixed with cement only, as determined by the standard test method, and this control sample is referred to as the control mix of the study.

The obtained curve was correlated with the classification of aggregates determined by the ABNT NBR 15577 standard code [17]. According to the standard, the result allows to conclude that the aggregate can be classified as potentially Reactive grade R2 due to a dimensional variation of 0.59% observed at 28 days (30 days since the specimens were molded).

4.3 Determining the expansion mitigation of mortar bars

The efficiency of cementitious materials to inhibit the expansion caused by the alkali-silica reaction was determined by the accelerated method in mortar bars, according to ABNT NBR 15577-6 [17]. The mitigation of the expansion reaction is considered efficient when the expansion of mortar bars is less than 0.19% at 28 days (30 days after molding). Figure 5 shows the dimensional variation values for the samples analyzed in this study.

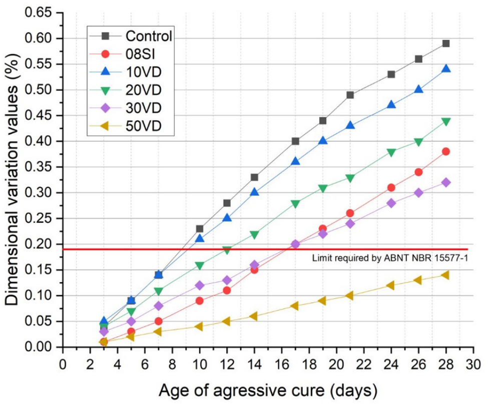

Figure 5. Assessment of ASR mitigation with glass powder and silica fume replacing Portland cement.

In all cases, the dimensional variations of the specimens prepared with reactive aggregate and added glass powder and with silica fume are smaller when compared to the control specimen. The results show that mortar expansion decreased with the increasing content of added minerals. At 28 days, the expansion values of mortars with 8% silica (08SI) and added 10, 20 and 30% glass powder (10VD, 20VD, and 30VD) reduced by 36%, 8%, 25% and 46%, approximately, compared to the control samples, respectively. However, despite the decreasing expansions, it is highlighted that only the 50%-added glass powder (50VD) mitigated the alkali-silica reaction to the level required by the standard, with a dimensional variation value of 0.14% expansion, decreased by 76% compared to the control samples.

Among the several existing ways to mitigate the expansions caused by the alkali-aggregate reaction, the use of supplementary cementitious materials is effective as long as quality materials and adequate dosages are used [38].

Afshinnia and Rangaraju [38] reported that the dilution effect and the pozzolanic reactivity of supplementary cementitious materials are responsible for reducing the expansion of mortar bars.

Monteiro et al. [39] stated that pozzolanic materials form additional hydrated calcium silicate (C-S-H) during the pozzolanic reaction. Due to the negative charge, C-S-H attracts the alkaline ions present in the concrete pores, reducing the concentration of free alkalis while mitigating the alkali-aggregate reaction.

Hooton et al. [40] conducted a study to analyze measures for preventing the alkali-silica reaction in concrete, correlating tests performed in the laboratory in the short term with long-term results. The tests were conducted to evaluate added supplemental cementitious materials and their effects on the alkali-silica reaction by comparing concrete exposed to adverse weather conditions outdoors and over a long-term exposure of 20 years. Two types of cement were studied: one with a high alkaline content, which cracked in 5 years, and another with a low alkaline content, which cracked after 12 years. These authors also studied two more types of concrete, one containing 50% ground granulated blast-furnace slag and another ternary mixture containing 25% slag plus 3.8% silica fume, reporting no cracks in both cases. They concluded that when sufficient levels of pozzolanic material were used, cracks due to the alkali-aggregate reaction did not develop, even after 20 years.

5 CONCLUSION

Considering only and exclusively the materials used in the reported experimental program, the results allow the following conclusions:

1. Glass powder used as a partial replacement for Portland cement showed pozzolanic reactivity since the modified Chapelle test yielded a minimum consumption index of CaO/g sample ($Ca(OH)_2$/g sample) greater than that established by ABNT NBR 15895 [27]. The result indicates portlandite consumption by the pozzolanic reaction.
2. The aggregate potential reactivity was determined by the expansion of mortar bars using the accelerated method, resulting in the dimensional variation of 0.59% at 28 days, defining a potentially reactive grade R2 according to ABNT NBR 15577 [17].
3. Analyzing the mitigation of the ASR, the silica fume content reduced the expansion caused by ASR by 36%. However, it was not enough to mitigate to adequate performance levels according to ABNT NBR 15577 [17].
4. The expansion caused by the ASR is reduced with increasing content of added glass powder. However, only for 50%-added glass powder (50VD), the reaction mitigated to a performance level considered adequate, i.e., the 0.14%-dimensional variation decreased the expansion by 76% compared to the control samples. This fact can be explained by the high alkali content of the glass powder.
5. The pozzolanic reaction plays a crucial role in diminishing the presence of alkalis within the pore solution of concrete. By reacting with calcium hydroxide, pozzolanic materials give rise to compounds of limited solubility. These compounds display reduced susceptibility to alkali reactions, leading to a decline in alkali concentration within the concrete's pore solution. As a result, incorporating pozzolanic admixtures into concrete formulations proves advantageous in alleviating the detrimental consequences of alkali-silica reactions, thereby enhancing concrete's resilience and longevity.
6. The ASR mitigation depends on both the content and the pozzolanic reactivity of the material incorporated to replace the Portland cement.

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