The behavior of some Brazilian cement types in mitigation on Alkali-Aggregate Reaction

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Abstract

Alkali-aggregate reaction (AAR) is a common concern in projects subjected to aggressive environments and to successive humidity conditions such as bridges, foundation blocks or dams. One way to mitigate this reaction is use cement with additions. Therefore, this work aims to analyse the reduction of the average expansion from the AAR due to the use of seven cements with different contents of pozzolan and blast furnace slag available in the market of Fortaleza/CE. Borosilicate glass was crushed, sieved and used as a reactive material in the production of mortar bars. Accelerated tests were performed with the bars according to Brazilian standardized test ABNT NBR 15577/18, which is similar to ASTM C 1260. Results indicate that there is a tendency of reduction of the average expansion with the increase of pozzolan and slag content on cement, highlighting cements CP IV 32 RS (Portland Cement with more than 15% of pozzolan and sulfate resistant) and CP III 40 RS (Portland Cement with more than 35% of blast furnace slag and sulfate resistant) as the ones with the best performance against AAR.

Keywords: alkali-aggregate reaction, cement with addition of blast furnace slag, cement with pozzolanic addition, mitigation.

1. INTRODUCTION

Concrete durability is related to its capacity to resist chemical, physical, mechanical and biological aggressions from the environment for which it was designed or to harmful internal interactions into concrete itself. Among the chemical aggressions, attacks by chloride and sulfate ions, carbon dioxide, acid attacks and the alkali-aggregate reactions (AAR) stands out [1].

AAR is a chemical reaction that takes place in a mortar or concrete, between hydroxyl ions (OH^-) associated with alkalis, sodium oxide (Na_2O) and potassium oxide (K_2O), from cement or other sources, and potentially reactive aggregates, under different temperature, pressure and pH conditions, in the presence of water [2-3]. Figure 1.1 illustrates this process.
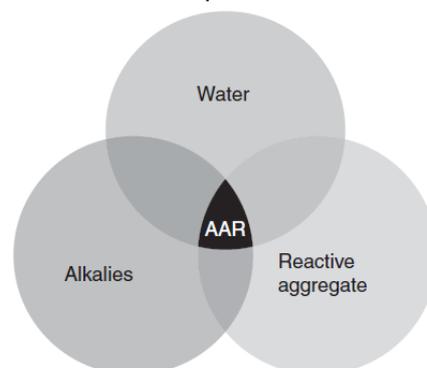

Figure 1.1: AAR conditions [4]

Rolim [4] points out that alkalis are released in the cement hydration process, where a portion of these is available in the solution of the concrete pores. In addition to cement, according to the author, other

materials used in concrete production, such as water, additives and aggregates containing alkaline minerals can be considered as sources of alkali in the mixture.

This reaction may or may not result in an expansive gel (depending on the concentration of lime or alkali) which, when disposed in the voids of the concrete and on the surface of the aggregate in contact with the cement paste, may result in generalized cracks and thus compromise the quality of the structure [5].

Hasparyk [6] also points out that the crack system generated by the expansion of the gel can allow the appearance of other deterioration mechanisms, accelerating the degradation process and affecting the durability of the concrete more significantly, since it exposes it to environmental conditions and the action of aggressive agents.

Valduga [7] mentions some of the symptoms that occurs in concrete structures as a result of AAR: micro-cracks, mainly in the mortar that fills the space between the coarse aggregates; cracks close to the surface of coarse aggregates, which can also occur at the paste / aggregate interface; detachment (loss of adhesion) of the mortar close to the surface of the coarse aggregates; possibility of occurrence of reaction frontier around the aggregates that reacted with the alkalis; presence of gel exuding or filling voids in the concrete; movement (opening and / or relative displacement) of contraction and concreting joints; characteristic cracking on the surface, with a polygonal panorama, with predominance in the direction of greater dimension.

Malvar et al. [8] argue that the most efficient measure to prevent AAR is the use of non-reactive aggregate, a solution that is not always practical or economically viable. To control the reaction, Silva [9] says that there are several materials available, and a way to minimize its risk is using cement with a high content of mineral additions. According to Mehta [10], about 50% to 70% of the clinker presented in Ordinary Portland Cement (OPC) can be replaced by several complementary materials. Thomas et al. [11] state that products such as pozzolans, slag, fly ash and mixtures based on lithium salts can be used to prevent the reaction, as long as they are used in the correct quantity.

Cachepa [12] highlights the decrease in the content of alkalis in cement and the use of pozzolans as a mineral addition, both in cement and in concrete, as methods of mitigating AAR. However, according to Hasparyk [13], depending on characteristics such as quantity of reactive phases, alkali content and fineness of the addition, the opposite effect can happen if they are used without control.

Munir et al. [14] mention some materials that can be used to mitigate AAR: granulated blast furnace slag, calcined clay, fly ash, rice husk ash, sugarcane bagasse ash and silica fume. The minimum levels of these mineral additions to OPC may vary depending on the degree of reactivity of the aggregate, the content of alkalis in the cement, the quality of these materials, as well as the expansion reduction mechanisms [8].

It is very important to determine the reactivity of the aggregate before its use in concrete and, from this assessment, select the most appropriate Portland Cement to mitigate AAR. Cements, beyond clinker and calcium sulfate, may contain additions of limestone filler, blast furnace slag and pozzolanic materials in different levels. When the aggregate is reactive and cement with slag or pozzolanic materials is available, the question is whether the content those mineral admixtures is sufficient to mitigate the alkali-aggregate reaction [1].

Ceará State, in the Northeast of Brazil, is the 3rd state in the number of eolic wind farms, producing 2,2 GW. Eolic towers usually has concrete foundations and AAR is a concern. In that region of Brazil, seven types of Portland Cement with pozzolans and/or blast furnace slag are marketed and there's no idea about they performance in AAR mitigation.

Thus, the aim of this article is to evaluate the AAR mitigation of seven Portland Cements with different contents of pozzolan and blast furnace slag commonly used in civil construction works in the city of Fortaleza/CE.

2.   LITERATURE REVIEW

In the early days of the knowledge of the alkali-aggregate reaction (AAR) it was believed that limiting the alkali content of the cement was sufficient to prevent the reaction from developing. The alkaline equivalent was thus limited to 0.60%. Subsequently, it was observed that this restriction was not enough itself, as some works carried out with less than 0.60% alkaline equivalent cements showed signs of AAR [4]. Research has shown, then, this limitation in the alkaline equivalent should be associated with the use of mineral additions to achieve the necessary efficacy against AAR. Cunha and Cincontto [1]

state that the efficiency of active additions in mitigating AAR varies according to their chemical and mineralogical composition, proportion in cement, degree of reactivity of the aggregate and the type of reaction.

Mineral additions, based on their physical-chemical action, are generally classified into three groups: pozzolanic material, cementing material and filler. The additions contribute to the production of cementitious materials with better technical characteristics, as they modify the internal structure of the hydrated cement paste, contributing to the reduction of the hydration heat [17]. About the groups mentioned, pozzolans are the preferred additions for the production of concretes of greater durability and in the mitigation of AAR since it consists of alumina, silica and iron oxide, according to their origin [12].

The performance of pozzolanic activity is influenced by the quality and quantity of amorphous phases present in pozzolan structure, also depends on its chemical and granulometric composition, specific surface area, texture (shape and size of the grain), density and w/c ratio [18].

Munir et al. [14] state that the use of fly ash in 30% of replacement to OPC is effective in reducing expansions due to AAR. Trindade [19] studied the addition of rice husk ash to concrete, without any type of processing. When using OPC plus 20%, 30% and 40% of fly ash, the mixtures proved to be efficient to mitigate AAR and their effect increased proportionally to the increase of addition of fly ash, being classified as harmless at 30 days. The mixture using Brazilian Portland Cement CP IV (it has more than 15% of pozzolan in its composition) showed excellence in mitigating the expansive reaction, being classified as harmless for the age evaluated.

When present in the composition of Portland Cement in contents of 25% to 35%, fly ash has is efficient in minimizing AAR. Blast furnace slag at levels above 50%, silica fume at levels from 10% to 12% and metakaolin at levels close to 10% have also been reported to be effective in reducing the reaction [8].

Unlike pozzolans, slag forms hydration products similar to cement, even without the presence of calcium hydroxide, however, the speed of blast furnace slag hydration reactions is very slow [19]. According to Cachepa [12], the main compounds of granulated blast furnace slag are silica, calcium oxide, alumina, magnesium oxide and iron oxide. For Wapda [20], the use of slag between 30% and 40% in the replacement of clinker reduces the expansion of AAR and the heat of hydration of the cement generated in the concrete mass.

In a study developed by Mizumoto and Salles [21], all ground slag samples indicated a minimum content of 40% of addition to inhibit the alkali-aggregate reaction. According to Munhoz [22], as the content of blast furnace slag, fly ash, metakaolin and silica fume increases, the expansion due to AAR is reduced. The minimum content necessary to mitigate expansions is lower for silica fume (around 10%), while for slag it would be around 50%. For these previous authors, among the types of pozzolans studied, silica fume would be the most efficient in minimizing the effects of AAR, followed by metakaolin and by fly ash.

The minimum contents of mineral admixtures to mitigate the expansion of aggregates analyzed in the research by Cunha and Cincontto [1] were: 10% of silica fume, 15% of metakaolin, 20% of fly ash and 60% of blast furnace slag. Authors explain that this performance is probably related to the chemical composition of the materials and the particles diameter of them, which confer high pozzolanic reactivity, markedly reduce the calcium hydroxide content of the mortar and favour the mitigation of the AAR. In the case of blast furnace slag, according to the these authors, higher levels were needed to reduce the CaO / SiO_2 ratio and the $Ca(OH)_2$ content after hydration reactions, attributing the lower efficiency of this addition to its chemical composition, whose calcium content is higher than the other additions, requiring levels of addition up to six times higher than silica fume to obtain the same degree of efficiency.

3. MATERIALS AND METHODS

3.1 Materials

Eight types of cement were used: the first was a Standard Cement, with a lot of clinker, called here by cement A; two types of cement with up to 14% of pozzolan, in Brazil called CP II Z, where in cement B, the pozzolan was fly ash, and in cement C, the pozzolan was calcined clay; three types of cement with more than 34% of pozzolan, in Brazil called CP IV, where in cement D, the pozzolan was fly ash, in cement E are calcined clay and in cement F are natural pozzolan; one type of cement with up to 34% of blast furnace slag, in Brazil called CP II E, here called cement G; and one type of cement with more than 35% of blast furnace slag, in Brazil called CP III, here called cement G. Cements B to G are

commercial in Fortaleza's market and cements B, D, G and H are from the same brand and cements C, E, F are from three other different brands. Cement A is a special one and there's out of Fortaleza's market. Table 3.1 brings cement composition according to Brazilian Standard ABNT NBR 16697:2018.

Table 3.1: Types of Portland Cement, according to Brazilian Standard ABNT NBR 16697:2018

Name	Initials	Content of clinker + gypsum	Blast furnace slag	Pozzolanic material	Limestone
Ordinary Portland Cement	CP I	95% - 100%	0% - 5%		
	CP I-S	90% - 94%	0%	0%	6% - 10%
Compost Portland Cement with blast furnace slag	CP II E	51% - 94%	6% - 34%	0%	0% - 15%
Compost Portland Cement with pozzolanic material	CP II Z	71% - 94%	0%	6% - 14%	0% - 15%
Compost Portland Cement with carbonatic material	CP II F	75% - 89%	0%	0%	11% - 25%
Blast Furnace Slag Cement Portland	CP III	25% - 65%	35% - 75%	0%	0% - 10%
Pozzolanic Portland Cement	CP IV	45% - 85%	0%	15% - 50%	0% - 10%
High Initial Strength Portland Cement	CP V	90% - 100%	0%	0%	0% - 10%

According to cement manufactures, the percentage of pozzolan and blast furnace slag of each used cement are exposed in Table 3.2.

Table 3.2: Content of blast furnace slag and pozzolanic material in used cement at this manuscript

Cement	Initials	Blast furnace slag	Pozzolanic material
A	CP I	-	-
B	CP II Z	-	14% of fly ash
C	CP II Z	-	10% of calcined clay
D	CP IV	-	30% of fly ash
E	CP IV	-	20% of calcined clay
F	CP IV	-	35% of natural pozzolan
G	CP II E	34%	-
H	CP III	50%	-

In order to ensure the occurrence of the alkali-aggregate reaction, as indicated in the American Standard ASTM C441 / C441M-17 [23], the natural aggregate was replaced by borosilicate glass, since this material is mostly composed by silica. To obtain the granulometry defined in standard ABNT NBR

15577-4 [15], the glass was placed in a Los Ángeles abrasion machine, with subsequent separation of the normative fractions with the aid of sieves with 4.75mm; 2.36mm; 1.18mm; 0.60mm; 0.30mm and 0.15 mm opening (Figure 3.1).
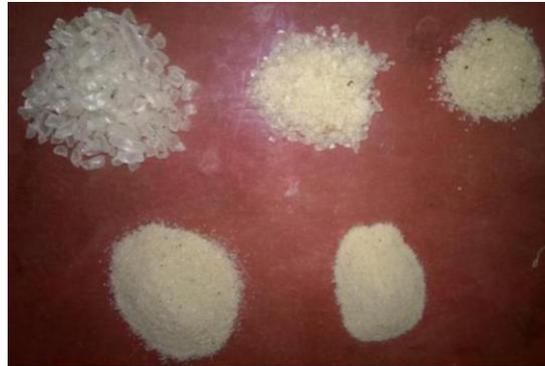

Figure 3.1: Borosilicate glass fractions used in the tests

3.2 Methods

The bulk density of borosilicate glass sand was obtained following the procedures of DNER ME 194/98 [24] standard, obtaining the value of 2.50 g/cm³. So, eight types of mortar were made, one for each cement of this paper, using the mixture specified by the ABNT NBR 15577-4 [15], which is 1: 2.25: 0.47 (cement: aggregate: distilled water) by mass.

The test followed the procedures recommended in the aforementioned standard for dosing, mixing, casting, curing and measuring three specimens per mix of mortar, as well as for the preparation of a solution, composed of sodium hydroxide and distilled water kept at 80 ºC in water bath equipment (Figure 3.2). Measurements were performed every 4 days for greater control of expansions, the last being obtained at 30 days. The apparatus used for the measurements was a metallic frame with a dial indicator with an accuracy of 0.001 mm (Figure 3.3).

Figure 3.2: Equipment used

Figure 3.3: Apparatus used for measurement

At the end of each test, the average expansion values were compared with the prescribed limits for verifying the mitigation of the aggregate reactivity presented at part 1 of NBR 15577 [25], as follows:

- For expansions equal to or greater than 0.19% at 30 days, reactivity is potentially harmful and mitigation with the materials used is not proven.

- For expansions of less than 0.19% at 30 days, reactivity is not harmful and mitigation with the materials used is proven.

In addition, the reduction (R) of the average expansion (in percentage) was calculated due to the use of mitigating cementitious materials according to Equation 1, present in ABNT NBR 15577-5 [16], which lists the values of the average expansions obtained by the method of part 5 of the standard (E5) with the values obtained by the method with standard cement of part 4 (E4) to verify how much mitigation there was by using cement with the addition of pozzolan.

$$R = 100 \cdot \frac{E4 - E5}{E4}$$ (1)

4. RESULTS AND DISCUSSION

Average expansion of specimens produced with each cement type are shown in Table 4.1.

Table 4.1: Results of expansions measured in mortar bars produced

Mortars made with cement	Expansion in% at the age of								
	* value obtained with the aid of a calliper or regression								
	1 day	4 days	8 days	12 days	16 days	20 days	24 days	28 days	30 days
A	0,000	0,071	1,066*	1,353*	2,109*	2,570*	2,797*	2,971*	3,145*
B	0,000	0,003	0,011	0,043	0,204	0,436	0,525	0,809	0,935
C	0,000	0,008	0,110	0,319	0,860	0,900*	1,117*	1,334*	1,443*
D	0,000	0,001	0,004	0,011	0,030	0,042	0,053	0,069	0,078
E	0,000	0,014	0,023	0,050	0,150	0,325	0,461	0,750	0,862
F	0,000	0,003	0,022	0,047	0,251	0,590	0,708	1,210*	1,331*
G	0,000	0,017	0,029	0,086	0,254	0,478	0,642	1,068*	1,235*
H	0,000	0,002	0,007	0,020	0,039	0,063	0,072	0,126	0,160

From Table 4.1 data, Figure 4.1 illustrates specimens' behaviour in the age, comparing with the standard limits at 30 days of age. As can be seen, only cements D and H mitigate the expansive reaction which means a expansion less than 0.19% ate 30 days of age.
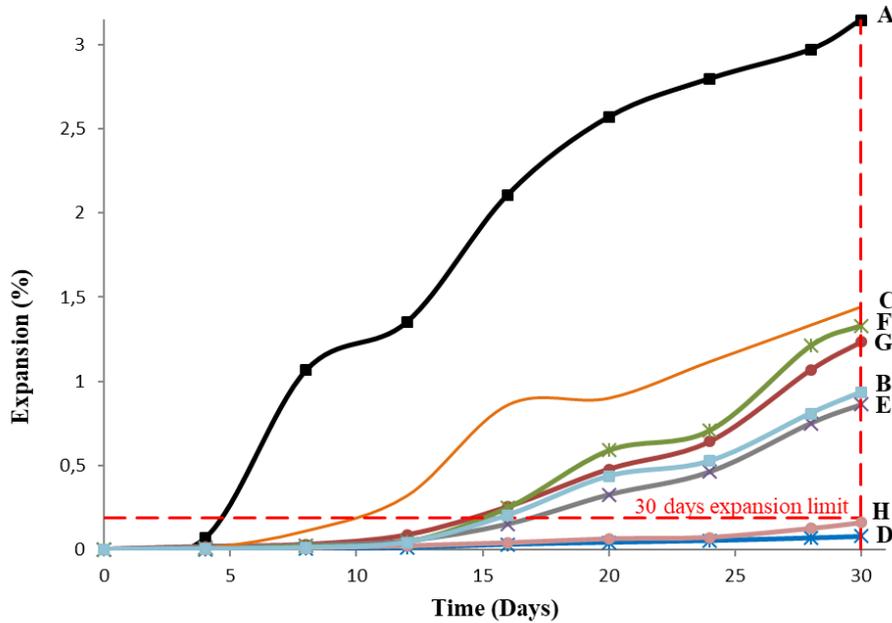

Figure 4.1: Bars expansions over time with pozzolanic and slag Brazilian cements

Some considerations about the tests with cement A, C, F e G are pertinent. After a certain age, the specimens expanded so intensely that they exceeded the measurement limits of the apparatus shown in Figure 3.3. Then, to complete all measurements by 30 days, the difference between measurements was made with the aid of a calliper with a precision of 0.05 mm. When it was not possible to use the calliper, the remaining expansions were obtained by regression. However, once at these dates, the average expansion values were already very high, it can be said that reactivity is potentially harmful and mitigation with glass as an aggregate is not proven for such samples.

Figures 4.2 and 4.3 show the specimens made with cement A, that have a great content of equivalent alkalis once this cement is type CP I, according to Tables 3.1 and 3.2. As can be seen by these figures, specimens made with this type of cement had a lot of expansion (in Figure 4.2 it is possible to seen bent specimens due to a lot of expansion) and a lot of cracks (see Figure 4.3).

Figure 4.2: Specimens made with cement A after 30 days of testing (Standard Cement)

Figure 4.3: Cracks formation in mortar specimens made with cement A after 30 days of testing (Standard Cement)

As can be seen at Table 4.1 and Figure 4.1, among cements tested, only specimens from Cement H and Cement D presented expansions below the normative limits, at 30 days, and the latter being the one that least expanded. According to Table 3.2, cement D has 30% of fly ash in composition although cement H has 50% of blast furnace slag. These results are coherent with Shayan [26] and Malvar et al. [8] that report the presence of pozzolan and blast furnace slag, respectively, mitigates in some degree the expansion due to AAR.

Figures 4.4 and 4.5 refer to specimens made with cement D (according to Table 3.2 this cement has 30% of fly ash). As already mentioned, this cement mitigated the expansion, however, it is possible to see in Figures 4.4 and 4.5 that the map crack pattern can still be verified in those specimens.

Figure 4.4: Cement D specimens after 30 days of testing

Figure 4.5: Formation of cracks in Cement D specimens after 30 days of testing

Important highlight the mitigation performance of fly ash against to calcined clay once specimens made by cement B, with 14% of fly ash, mitigates more the expansions than specimens made by cement F, that contains 35% of natural pozzolan, and mitigates almost the same than specimens made by cement E, with 20% of calcined clay. Another comparison can be specimens made by cement B with specimens made with cement C. Cement C has only 10% of calcined clay and has the second worst behaviour although cement B has 4% more pozzolan (which means 14% of fly ash) resulting in 40% less expansion. According to Ghafoori and Kian [29], the efficiency of mitigation by fly ash or natural pozzolans varies according to the type of reactive aggregates.

Using Equation 1, the average expansion reduction related to cement A was calculated for all cements at 30 days. Results can be seen in Table 4.2.

Table 4.2: Reduction in average expansion compared to standard cement (30 days)

	Cement						
	C	F	G	B	E	H	D
Reduction (%)	54,12	57,68	60,73	70,27	72,59	94,91	97,52

Such differences can be explained by the content of pozzolan and of blast furnace slag added to the cement, whose values were provided by the manufacturers and already shown in Table 3.2.

When applying Pearson's correlation coefficient to the variables "reduction of expansion" and "pozzolan content", it can be find the value of 0.721, which, according to Hinkle, Wiersma and Jurs cited by Mukaka [28], if this value is between 0.7 and 0.9, it shows a high positive correlation, which means, as one increases, the other has a high tendency to increase.

Applying the same coefficient for the blast furnace slag content, the value is 0.999, which would indicate, according to the same authors [28], a very high positive correlation. However, in order to better assess the existence of this correlation, these tests should be carried out with more cements with different blast furnace slag contents.

5. FINAL CONSIDERATIONS

According to the results obtained, it can be concluded that cements with pozzolanic additions and blast furnace slag have the capacity to reduce to some degree the average expansion due to the alkali-aggregate reaction that occurs with glass. Such mitigation increase as the content increases in the cement composition. Furthermore, there is a high positive correlation between the reduction of expansion and the pozzolan content in the cement. More tests must be done with the cements with blast furnace slag for a more reliable correlation.

Another finding was that the results show evidence that the type of pozzolan has an influence on the mitigation of AAR, once cements with fly ash mitigate more than cements with calcined clay and cement with natural pozzolan.

The best performance in the mitigation of AAR occurred with cements D and H, respectively, that are a cement with 34% of fly ash and a 50% of blast furnace slag, respectively. Such cements remained below the normative limits for bars expansion on accelerated test, even using a highly reactive aggregate, after 30 days.

The work limitations were related to the exacerbated growth of some samples, which prevented the measurement of the expansion over the entire course of the 30 days using the normative apparatus, having to be measured with the aid of a calliper or using regression.

As future work, it is suggested to repeat the cement tests with slag additions (CP II E and CP III) including more cements with other levels of addition. Besides, it is proposed that tests that last longer be carried out, such as part 6 of NBR 15577 [30], so that one can have a better evaluation of the behaviour of such cements in larger volumes of concrete.

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