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# Assessment of the Alteration of Granitic Rocks and its Influence on Alkalis Release

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**Abstract.** Several concrete structures had shown signs of degradation some years after construction due to internal expansive reactions. Among these reactions there are the alkali-aggregate reactions (AAR) that occur between the aggregates and the concrete interstitial fluids which can be divided in two types: the alkali-silica reaction (ASR) and alkali-carbonate reaction (ACR). The more common is the ASR which occurs when certain types of reactive silica are present in the aggregates. In consequence, an expansive alkali-silica gel is formed leading to the concrete cracking and degradation. Granites are rocks composed essentially of quartz, micas and feldspars, the latter being the minerals which contain more alkalis in their structure and thus, able to release them in conditions of high alkalinity. Although these aggregates are of slow reaction, some structures where they were applied show evidence of deterioration due to ASR some years or decades after the construction. In the present work, the possible contribution of granitic aggregates to the interstitial fluids of concrete by alkalis release was studied by performing chemical attack with NaOH and KOH solutions. Due to the heterogeneity of the quarries in what concerns the degree of alteration and/or fracturing, rock samples with different alteration were analysed. The alteration degree was characterized both under optical microscope and image analysis and compared with the results obtained from the chemical tests. It was concluded that natural alteration reduces dramatically the releasable alkalis available in the rocks.

## 1. Introduction

There are two main types of internal expansive reactions that can affect the durability of concrete: internal sulphate reaction (ISR) and alkali-aggregate reaction (AAR). In AAR two types of reactions are generally considered: alkali-silica reaction (ASR) and alkali-carbonate reaction (ACR) [1]. ASR, the most common AAR, occurs between the alkali hydroxides released during the cement hydration or released from the aggregate particles, with different forms of reactive silica present in aggregates.

Although granitic aggregates are usually considered unlikely to be reactive to alkalis, some studies have been developed which showed that this is not always the case [2, 3, 4]. Granites contain feldspars, quartz and mica as the main components and are classified as alkali slowly reactive, since structures



with these aggregates can present manifestations of ASR decades after being built. The features that can be identified by petrographic analysis and which are supposed to contribute to the potential alkali reactivity of these rocks are the presence of deformed quartz with undulatory extinction, strain lamellae and ribbon texture, bulging and sub-graining. In addition, porosity and microcracks can also play a role by promoting the circulation of fluids in the interior of the aggregate particles. Besides the presence of potentially reactive silica, research developed in Canada [5,6] proved that some aggregates can release alkalis to the concrete interstitial solution therefore increasing the total alkalis content of concrete and eventually contributing to ASR.

Granitic quarries are usually heterogeneous, containing rocks with variable degrees of alteration. Four granitic aggregates were selected for this work which is focused in two interrelated subjects, namely, it aims to determine the content of alkalis released by some Portuguese granites; and to define the influence of the degree of alteration of the rocks on the capacity to release alkalis. The alteration features were evaluated by image analysis in order to establish the classes of alteration. Two alkaline solutions were employed [8], namely NaOH and KOH, in order to evaluate the alkalis release. The results obtained are presented and discussed.

## 2. Materials and methods

Four Portuguese granitic aggregates identified as GR2, GR23, GR24 and GR29 were selected. For each aggregate, two different fractions were hand separated from the batches received from the quarries: one fraction is composed of sound, light grey non-altered rock (GR2-N, GR23-N, GR24-N, GR29-N), and the other of yellowish altered granite (GR2-A, GR23-A, GR24-A, GR29-A). These aggregates were previously tested by the accelerated concrete prism test RILEM AAR-4.1 [1] which had shown reactivity values between 0.03 % (GR23 and GR29) to 0.06 % (GR2 and GR24) at 15 weeks. The expansion values obtained allowed to classify all these granites as Class II, likely to be potentially reactive, according to RILEM [1] and LNEC [7] Recommendations.

The petrographic analysis of the thin sections of the aggregates was performed under polarizing microscope OLYMPUS BX60F5, coupled with a digital camera and Pelcon automatic point-counter. The definition of alkali reactivity was based on the dimensions of the crystals of quartz (<100  $\mu\text{m}$  = microcrystalline quartz). In order to establish classes of alteration, image analysis was carried out on photomicrographs, after converting them to grayscale images. A threshold was applied for selecting a range of certain shades, allowing highlighting the alteration of the feldspars to clay minerals and microcrystalline muscovite and the density of cracks in each sample.

For the determination of the alkali content released by the aggregates the samples were first crushed and grinded, to pass a 150  $\mu\text{m}$  sieve. Then, 100 g of each grinded sample were immersed in 400 mL of alkaline solution at a ratio of 1:4 aggregate/solution and maintained at 38 °C (test conditions of RILEM AAR-3[1]). Two alkaline solutions were employed: NaOH 0.7M (for the evaluation of the K supply) and KOH 0.7M (for the Na supply). Twice a week, the test containers were gently rolled back for about 10 seconds, and at predefined time intervals (e.g.: 2, 7, 13 and 26 weeks) a sample was taken from the test solution (10 mL) to be analysed by Atomic Absorption Spectroscopy (AAS), after filtration and acidification [8, 9].

## 3. Results and discussions

### 3.1. Petrographic Analysis

The petrographic analysis focused on the features of the rocks which are considered to contribute to the occurrence of ASR. Besides the characteristics that refer to the potential reactivity, namely microcrystalline quartz, including myrmekite, sub-graining, deformed quartz exhibiting undulatory extinction, strain lamellae and ribbon quartz, the presence of K-feldspar, Na-plagioclase, perthite, alteration of the feldspars with formation of clay minerals and the process of alteration of biotite were registered. The occurrence of cracks was also considered an important factor as cracking is usually associated with the degree of alteration of the rocks, can improve the progression of the chemical reactions and increase the specific surface exposed to the interstitial fluids.

The aggregates analysed are mainly fine to medium-grained two mica granites, composed of quartz, K-feldspar, plagioclase (essentially albite, An<sub>0-10</sub>, and oligoclase, An<sub>10-30</sub>), muscovite and biotite, with variable contents of chlorite and other accessory minerals (Table 1).

**Table 1.** Mineral composition of the aggregates obtained by point-counting.

	<b>GR2 [3]</b>	<b>GR23</b>	<b>GR24</b>	<b>GR29</b>
<b>K-feldspar</b>	26.3	29.9	31.4	32.9
<b>Plagioclase</b>	26.1	19.5	20.3	24.6
<b>Quartz</b>	34.2	34.4	33.7	32.7
<b>Muscovite</b>	9.7	7.5	10.0	4.0
<b>Biotite + chlorite</b>	3.1	8.1	4.1	5.7
<b>Accessory minerals</b>	0.6	0.6	0.6	0.3

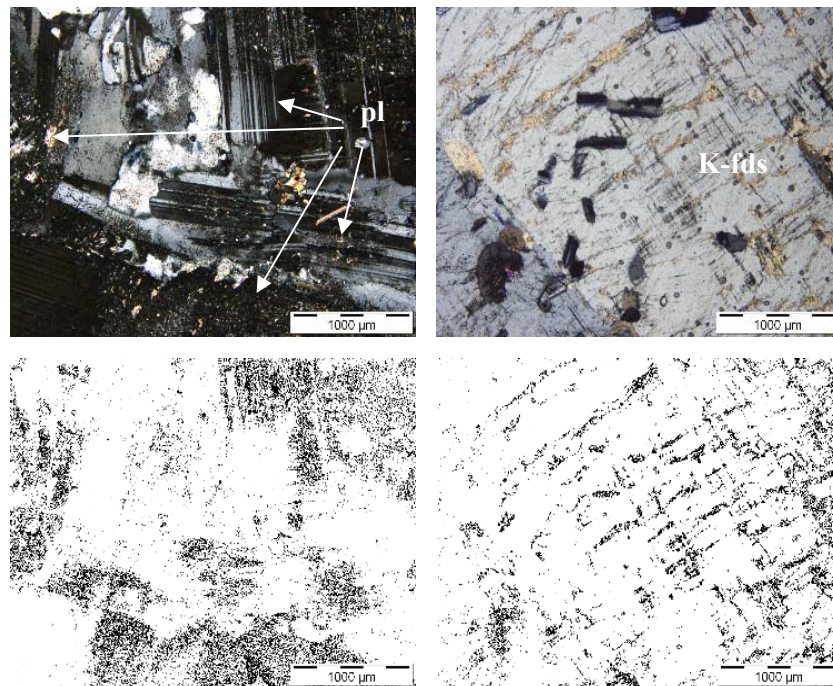
As can be observed from Table 1, quartz is the main component in three of the samples (the exception is GR29) and K-feldspar (26.3 to 32.9 %) is more abundant than plagioclase (19.5 to 26.1 %). The content of muscovite is higher than biotite + chlorite in GR2 and GR24 with a ratio of 3.1 and 2.4, respectively. Biotite + chlorite are slightly more abundant in GR23 and GR29. These results are in agreement with the bulk rock chemical composition (Table 2) which shows that the K<sub>2</sub>O content exceeds by 1.5 the Na<sub>2</sub>O content.

**Table 2.** Chemical composition of the aggregates regarding alkalis.

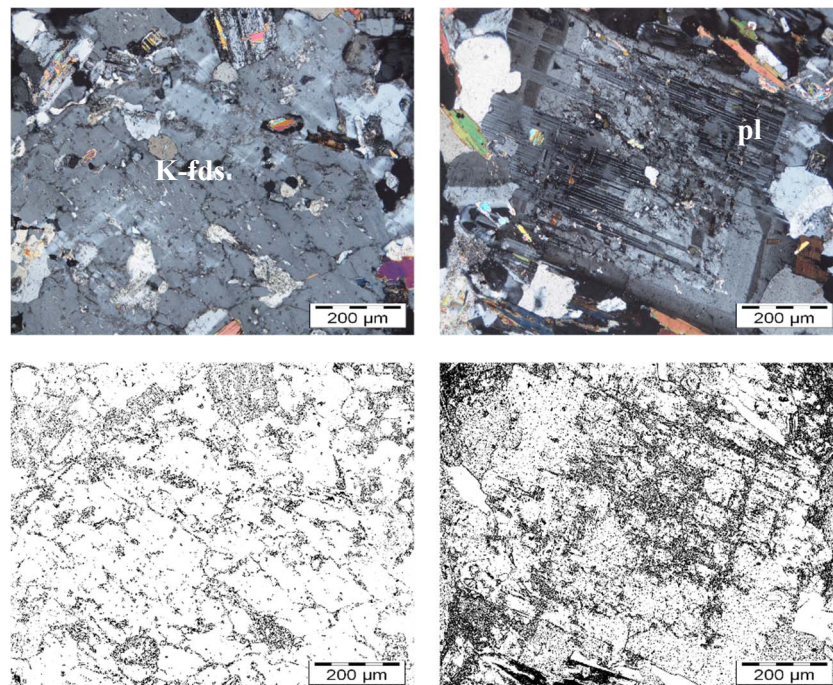
	<b>GR2</b>	<b>GR23</b>	<b>GR24</b>	<b>GR29</b>
<b>Na<sub>2</sub>O</b>	2.81	3.00	2.99	3.26
<b>K<sub>2</sub>O</b>	4.43	4.96	5.19	5.08
<b>K<sub>2</sub>O/ Na<sub>2</sub>O</b>	1.58	1.65	1.74	1.56
<b>Na<sub>2</sub>O<sub>eq</sub></b>	5.72	6.26	6.40	6.58

Most of the selected aggregate samples contain the petrographic deformation features referred above that can imprint potential reactivity to these aggregates. Perthites, myrmekites and either undulation extinction or sub-granulation of quartz crystals are frequent. However, the samples showing more intense deformation are also those exhibiting deeper alteration. On the other hand, the altered fractions (A) are those that present a more intense cracking and a variable content of clay minerals. They are also more porous, therefore presenting a much higher specific surface for both quartz and feldspar to be exposed to the interstitial fluids.

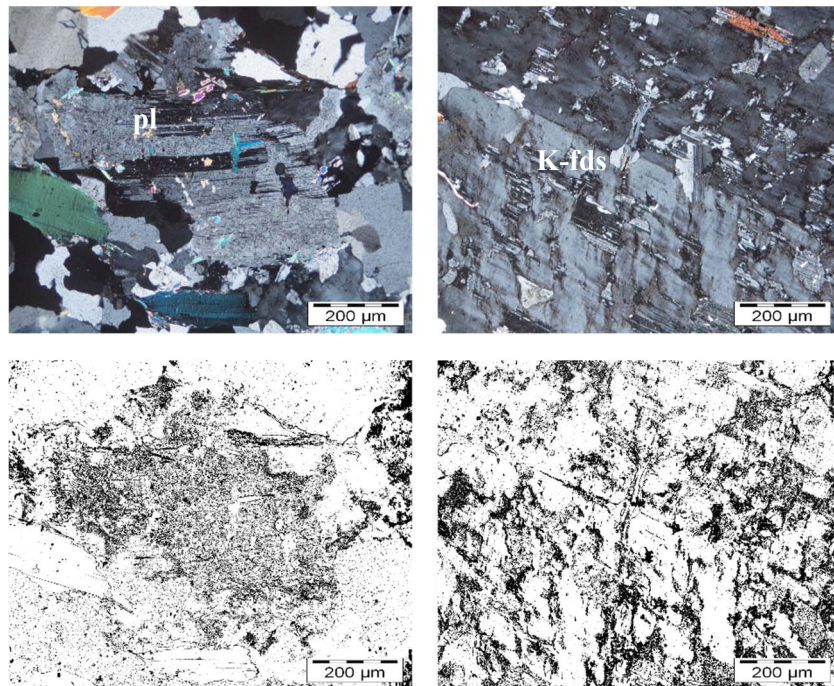
Figures 1 to 4 show some of the petrographic features observed in the granitic samples, namely the alteration of the feldspar crystals. In those figures the results of the image analysis are presented in which the alteration and cracking of the rocks are highlighted in shades of grey. In all the samples, plagioclase crystals show more intense alteration than K-feldspars crystals. In the zoned plagioclase crystals, the nucleus is altered to microcrystalline muscovite and, in some crystals of the samples GR23-A and GR29-A, also calcite is formed. Biotite is partially to completely altered to chlorite. The darker areas of the images obtained by image analysis correspond to the argillization of the feldspars, showing the intensity of alteration of the plagioclase crystals whilst the K-feldspars are mainly crossed by cracks with minor presence of clay minerals. In the non-altered fraction (N) these characteristics are less evident than in the altered fraction (A). However, alteration in the cores of plagioclase crystals is visible even in the non-altered fractions.



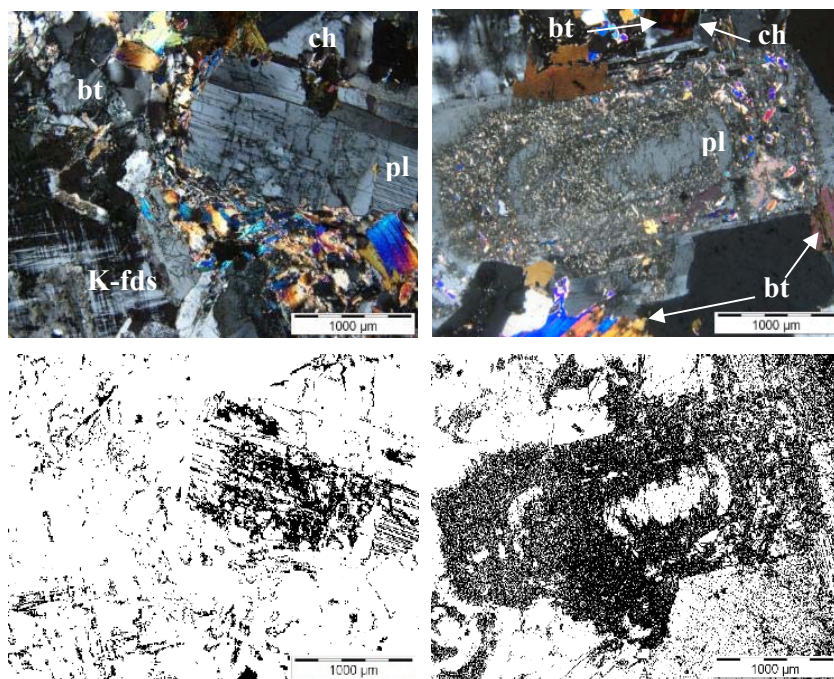
**Figure 1.** Aggregate GR2 containing fractured and altered plagioclase (pl) crystals, slightly altered K-feldspar (K-fds) (photomicrographs obtained in crossed polarized light). Image analysis (below) highlights the differences in the degree of the alteration of plagioclase and K-feldspar crystals by different shades of grey. The darker areas are the ones presenting clay minerals and/or cracks



**Figure 2.** Aggregate GR23 containing slightly altered K-feldspar (K-fds) and zoned, altered plagioclase (pl) crystals (photomicrographs obtained in crossed polarized light). Image analysis (below) highlights the differences in the degree of the alteration of K-feldspar and plagioclase crystals



**Figure 3.** Aggregate GR24 containing altered plagioclase (pl) crystals and K-feldspar (K-fds) (photomicrographs obtained in crossed polarized light). Image analysis (below) highlights the differences in the degree of the alteration of K-feldspar and plagioclase crystals



**Figure 4.** Aggregate GR29, with perthitic K-feldspar (K-fds) and zoned, intensely altered and cracked plagioclase (pl) crystals. Plagioclase shows intense alteration to microcrystalline muscovite, mainly in the nucleus (photomicrographs obtained in crossed polarized light). Image analysis (below) shows that the alteration of plagioclase is more intense than of the K-feldspar crystals

Through the petrographic analysis it can be concluded that the aggregates show variable alteration evidences and grades. Aggregate GR2 is the least altered, even when the altered fraction is considered. There is light alteration in the cores of the plagioclase crystals and alteration is scarce for feldspar-K crystals. In GR2-A, the altered fraction, plagioclase (oligoclase) is more altered in the core and usually is more altered than the K-feldspars.

The other aggregates show stronger manifestations of alteration which might be the deepest for GR29. The zonation of plagioclases is visible mainly in GR23 and GR29. Therefore, the nucleus of plagioclase crystals is marked by intense sericitization for both GR23-A and GR29-A.

Based on the image analysis, the quantification of the areas of the samples showing alteration evidences is presented on Table 3, namely for  $\frac{3}{4}$  of the percentages of altered areas of the crystals, for each type of feldspar considering the aggregate fractions separately.

**Table 3.** Percentage of altered area of the feldspar crystals in the different aggregate's fractions (N - non-altered rock; A - altered rock)

Aggregate	GR2		GR23		GR24		GR29	
	N	A	N	A	N	A	N	A
<b>K-feldspar</b>	10%	13%	21%	27%	31%	35%	33%	39%
<b>Plagioclase</b>	16%	21%	25%	33%	35%	40%	34%	48%

From these results is possible to make a sequence of the aggregates regarding their degree of alteration. Aggregate GR2, as already concluded from the petrographic analysis, is the one with less evidences of alteration. It is followed by the GR23 that showed three out of the four fractions with less than 30% of altered crystals area. Aggregates GR24 and the GR29 are rated as the ones exhibiting the more intense alteration, although the GR29 has the highest values for all the fraction.

### 3.2. Alkalis Release

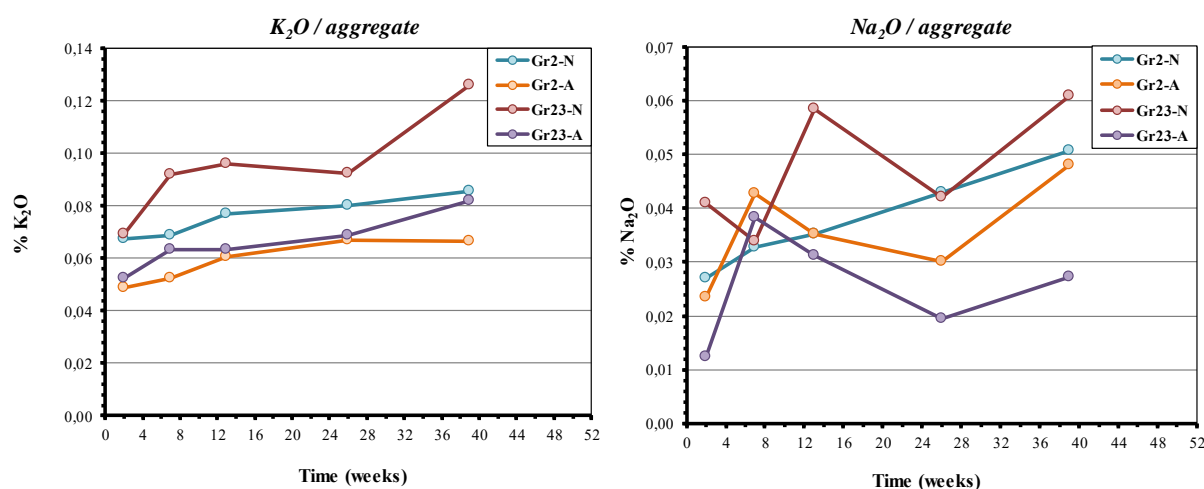
The results for the alkalis released after 26 weeks of testing are plotted in charts in Figures 5 and 6. Curves of the altered (A) and non-altered (N) fractions of the aggregates are presented showing the different behaviour of the aggregates in NaOH and KOH solutions.

The analysis of these charts reveals that the altered fractions (A) release less alkalis than the non-altered ones (N), although in the altered fractions there are more pores and cracks which might allow the access of the interstitial fluids to the interior of the aggregate particles.

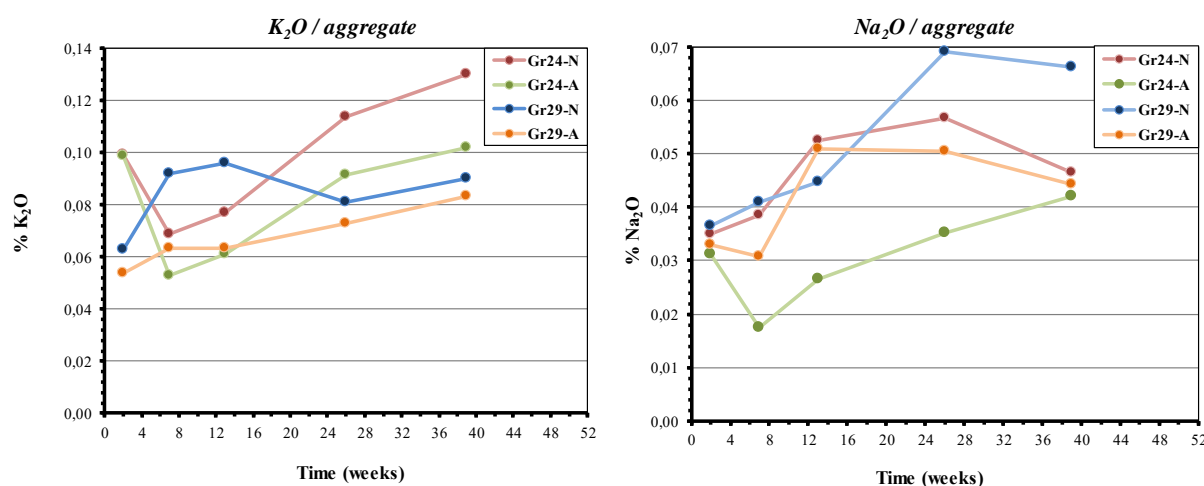
This result is explained by the fact that the minerals in the altered fractions have already lost  $K^+$  and  $Na^+$  during the alteration processes, indicating that the natural mechanisms of alteration have a stronger role in the releasable alkalis than the fact of the facilitated access of the interstitial fluids and the larger specific area available to the alkaline attack due to cracking.

It can also be verified that the content of  $K_2O$  released is higher than  $Na_2O$  in all the samples. This fact can be due to the alteration degree observed in the petrographic analyses, which is more intense for the plagioclase crystals than for the K-feldspars in all the samples. In consequence, less sodium is available to be releasable during the alkaline attack. In addition, the results of alkalis released show a good correlation with the K-feldspar content obtained by petrographic point counting, which is usually higher than the plagioclase (Table 1).

The lines in the charts also show that there is a dominant low positive trend in the evolution of the  $K_2O$  release in the least altered aggregates (GR2 and GR23) whilst the lines are quite irregular for GR24 and GR29, where the values for GR24 decrease from the first to the second assessment, increasing thereafter. GR29-N shows a high increase at the beginning of the test, decreasing in the 26<sup>th</sup> week of test. Only GR29-A follows the tendency shown for GR2 and GR23.



**Figure 5.**  $Na_2O$  (%) and  $K_2O$  (%) contents released with time by GR2 and GR23 aggregates in alkaline solutions (N – non-altered rock; A – altered rock)



**Figure 6.**  $Na_2O$  (%) and  $K_2O$  (%) contents released with time by GR24 and GR29 aggregates in alkaline solutions (N – non-altered rock; A – altered rock)

In what regards the  $Na_2O$ , the least altered aggregates (GR2 and GR23) show variable evolution but it is interesting to notice that the altered fractions (GR2-A and GR23-A) follow parallel tendencies, initially with a clear increase followed by a decrease until the 26<sup>th</sup> week. This variable behaviour is thought to be associated with the compositional zoning of plagioclase, which is more evident for GR23-A. This interpretation does not apply, however, to GR23-N with a very irregular evolution. For the aggregates showing the strongest alteration signs, the lines are also difficult to explain as for the altered fractions (GR24-A and GR29-A) there is a decrease in the  $Na_2O$  content release, with a steady increase of GR29-A after the 2<sup>nd</sup> week, followed by a stabilization, and a positive trend for GR24-A. These tendencies are not reflected in the non-altered fractions of the same aggregates which show irregular evolution along the test period. The analysis of the curves in the four charts shows that the linear tendency observed for  $K_2O$ , with higher release content for the non-altered fractions, cannot be totally extrapolated for the  $Na_2O$  evolution.

As example, the results at the 2<sup>nd</sup> week for GR2-A are higher than for GR2-N. In the analysis of the values of alkalis released it should be taken into account a possible precipitation with time of some



alkalis forming non-soluble alkaline gels. This situation will be confirmed at later ages, when these tests are finished.

#### 4. Conclusions

The results obtained suggest that feldspars in granitic rocks can contribute to the development of ASR in concrete by releasing alkalis to the interstitial fluids, eventually exceeding the maximum content of alkalis considered in the national and international recommendations as the threshold to avoid the occurrence of reactions.

Based on the petrographic examination of the thin sections and the image analysis it is possible to establish a qualitative classification of aggregates regarding the degree of alteration and cracking of the rocks. The features are observed mainly in the feldspar crystals, which can release alkalis to the fluids. Plagioclase is, in all the samples, more altered than the K-feldspar and the rocks are richer in K-feldspar than in plagioclase. Both these facts can explain the different behaviour found in the alkalis release tests carried out.

It has been found that naturally altered aggregates have less capacity to release alkalis than the non-altered aggregates since that the fractions that showed a higher grade of alteration correspond to the particles that release fewer alkalis. The granites tested showed a higher potassium release, which is attributed to the higher natural alteration of plagioclase in comparison to K-feldspars but also to the higher content of K-feldspars than plagioclase in these rocks. The rates of sodium release seem to reflect the compositional zoning of the plagioclase crystals.

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