011912-MEDE

ASSESSMENT OF THE POTENTIAL ALKALI-REACTIVITY OF VOLCANIC AGGREGATES FROM AZORES ISLANDS

Sara Medeiros[1*], Tetsuya Katayama[2], Vittorio Zanon[1], Isabel Fernandes[3*], António Santos Silva[4], João Carlos Nunes[1], Vidália Miranda[5], Violeta Ramos[3], Dora Soares[4]

[1]Universidade dos Açores, Departamento de Geociências, Ponta Delgada, Azores, Portugal

[2]Kawasaki Geological Engineering Co.Ltd, Tokyo, Japan

[3]Universidade do Porto, DGAOT/Centro de Geologia, Porto, Portugal

[4]Laboratório Nacional de Engenharia Civil, Lisboa, Portugal

[5]Laboratório Regional de Engenharia Civil, Ponta Delgada, Azores, Portugal

Abstract

Volcanic rocks have been found to be potentially alkali-reactive in a number of countries, including Japan, Iceland and Turkey. To characterize the potential reactivity of the volcanic rocks used as aggregates for concrete in the Azores islands, an experimental program is being carried out which includes petrographic examination, chemical analysis and lab accelerated tests.

The geochemical composition of the rocks of the Azores islands varies considerably. The rocks used as aggregates are mainly silica-undersaturated basalts and trachytes. The petrographic examination and the chemical analyses of the rocks confirmed that one of the five analysed samples contains free silica and another sample presents volcanic glass. The occurrence of microcrystalline quartz as a secondary product filling the interstices of the trachyte was confirmed by SEM/EDS.

This paper presents the preliminary results obtained on the characterization of the first five samples of volcanic rocks from Azores regarding the potential reactivity to alkalis using different methods.

Keywords: Azores islands, volcanic aggregates, petrography, geochemistry, expansion tests

1 INTRODUCTION

One of the main issues concerning alkali-silica reactions (ASR) is the identification of the aggregates that are susceptible to ASR before their use in concrete, aiming to prevent possible future damages.

Several studies on ASR have been carried out on volcanic aggregates used in different countries such as Japan, Iceland, Australia, New Zealand, Turkey and Brazil [1-8]. In Portugal, the only information about the performance of volcanic rocks is related to the study of the pavement of the airport at Santa Maria Island in the Azores archipelago. This study was developed some years ago [9] and it was concluded that the deterioration of the concrete was due to a complex process, which involved expansive reactions associated with altered volcanic aggregates. Presently, no systematic studies exist on the characterization of the aggregates used in concrete in the Azores islands. In order to overcome this lack of information, a

* Correspondence to: saramedeiros@uac.pt
* Correspondence to: ifernand@fc.up.pt

comprehensive study was initiated in 2010 with the joint cooperation between the University of Azores, the University of Porto, the LNEC and the Regional Laboratory of Civil Engineering of Azores. The Azores archipelago consists of nine volcanic islands in the North Atlantic Ocean, at the triple junction of the North American, Eurasian and African tectonic plates. The tectonic framework of the Azores involves the presence of the Mid-Atlantic Ridge (MAR), which marks the boundary between the North American and the other two plates. The boundary between the Eurasian and the African plates is somewhat controversy, but in general terms, it corresponds to the Azores-Gibraltar Fault. According to [10], this boundary displays three segments with different natures: (1) a compressive regime sector, close to the Iberian margin, (2) the Gloria Fault, as a dextral transform fault and (3) a transtensional regime sector, between the MAR and Santa Maria Island. The archipelago magmatism belongs to the alkaline series with a predominant sodic character with compositional characteristics that range from basalts to trachytes, and includes more evolved (silicious) rocks such as comendites and pantelerites. Since the settlement in the 15th century, several eruptions occurred in São Miguel, Terceira, São Jorge, Pico and Faial islands, and in the surrounding seafloor. Santa Maria Island is the oldest of the archipelago, with an estimated age of 8.12 Ma. [11].

This paper presents the first results regarding the assessment of the potential reactivity of some volcanic Azorean rocks. Five rock samples from four islands were selected. Instructions in the Portuguese recommendation [12] were followed and the procedures complemented with specific test methods published in the literature about volcanic rocks. For some of the samples, the tests performed were not conclusive and further studies have to be developed.

2 MATERIALS AND METHODS

2.1 General

The five rock samples came from the islands of Santa Maria (SMA-SM1), São Miguel (SMG-SM1), Terceira (TER-SM1 and TER-SM2) and Graciosa (GRA-SM1). They belong to different geologic formations with different geochemistry that characterize the eruptive history of the islands. The rocks from Santa Maria Island belong to the Facho-Pico Alto complex composed of basalts [13]; the aggregates from São Miguel Island belong to the Picos Volcanic Complex and are predominantly basalts [14]; the lavas from Terceira Island belong to Guilherme Moniz and are alkaline trachytes [15] and to the Fissure Zone that consists of basalts [16]; the rocks from Graciosa Island belong to Praia complex or Vitoria Unit and are basalts [17].

The study started with the petrographic examination in accordance with [12]. Complementary evaluation included: (1) bulk chemical analysis, (3) chemical test for alkali-reactivity, and (4) phosphoric acid extraction method. At the present stage, accelerated mortar and concrete expansion tests were performed just on the sample collected in a quarry of basaltic rocks in São Miguel Island (SMG-SM1).

2.2 Methods for assessment and analysis

Mineralogical and textural characteristics of the samples were examined in conventional thin-sections (Nikon Eclipse E200 Pol, plus Zeiss MRc5 camera). Selected polished thin-sections were carbon-coated and examined by scanning electron microscope (SEM) equipped with an energy dispersive spectrometer (EDS)(JEOL JSM-6301F, NORAN-VOYAGER: 15 kV, working distance 15 mm, collection time 60 seconds, dead-time 30%). SEM/EDS were used only in the SMA-SM1 and TER-SM1 samples to observe or confirm some minerals that were not easily identified by optical microscopy.

Chemical analyses were performed on the five rock samples to calculate the normative quartz. The major oxides were determined by fusion-inductively coupled plasma (FUS-ICP: Thermo Jarrell-Ash ENVIRO II ICP) at Activations Labs, Canada. Free-silica content by phosphoric acid attack method was used to detect reactive silica minerals in volcanic aggregates, following the instructions in [18].

The chemical alkali-reactivity test was performed on all five samples according to ASTM C 289-07 [19] after crushing and sieving (150-300 μm). Potential reactivity of the sample (SMG-SM1) was evaluated according to three different expansion test methods: 1) the accelerated mortar-bar test following ASTM C 1260-07 [20], with high-alkali Portland cement (CEM I 42.5 R, 0.82% Na_2Oeq), graded aggregates (<4.75 mm), cement/aggregate (0.44) and water/cement ratio (0.47), extended up to 28 days to confirm the results obtained at 14 days; 2) the accelerated concrete prism method in accordance with RILEM AAR-4.1 [21], using the same high-alkali Portland cement as above, and graded aggregates (<20 mm), cement/aggregate (0.23) and water/cement (0.50); 3) concrete prisms according to RILEM AAR-3 [22], with which measurements are extended at regular intervals until 1 year.

3 RESULTS

Based on the petrography, the five samples (SMA-SM1, SMG-SM1, TER-SM1, TER-SM2, GRA-SM1) were classified according to their mineralogical composition and textural features [23]. Table 1 presents the main characteristics of the rocks (Figure 1) which had two generations of phenocrysts.

Petrography

Basanite (SMA-SM1) is composed of olivine with iddingsitized rims, clinopyroxene, plagioclase phenocrysts and opaques (ilmenite and magnetite) with a matrix formed by the same minerals but with a lower amount of olivine. Anhedral intergranular analcite was identified in several areas by SEM/EDS analysis.

Trachybasalt (SMG-SM1) is composed of olivine, clinopyroxene, plagioclase and opaques in a matrix formed by the same minerals plus apatite and a small amount of intergranular groundmass. Olivine crystals show typically a granular morphology, occasionally with skeletal morphology. Glomeroporphyritic crystal clots with clinopyroxene, plagioclase and opaques are also present. Some clinopyroxenes show reaction rims and embayments. A xenocryst of quartz surrounded by pyroxenes and some opaque minerals was identified by optical microscope. Brownish and isotropic volcanic glass, with SiO_2 58% according to EDS analysis performed in Japan, fills the interstices in the groundmass. The dark colour of the rock is due to the abundance in glass, which can be readily recognized in thin-section with thickness of 15μm. The amount of volcanic glass varies with the sampling area in the various sectors of the quarry.

Trachyte (TER-SM1) has a mineral assemblage of large crystals of sanidine and nepheline, and smaller sanidine, nepheline, biotite and opaques. The pseudo-fluidal groundmass consists of feldspars (plagioclase, anorthoclase), pyroxenes (aegirine, augite), aenigmatite, apatite and opaque minerals (mainly magnetite). Interstitial silica, presently microcrystalline quartz, was confirmed by SEM/EDS (Figure 2) and in vesicles. This trachyte shows evidence of alteration containing an interstitial iron silicate gel (SiO_2 ca 50%, Fe_2O_3 26%), which is pseudomorphic after aegirine and more siliceous than hisingerite but its alkali-reactivity is unknown.

Basalt (TER-SM2) consists of phenocrysts of olivine, clinopyroxene and plagioclase (labradorite) set in fine-grained groundmass composed of the same phases and opaque minerals dominated by ilmenite. Olivine is slightly iddingsitized. Plagioclase crystals show reaction rims and embayments and are occasionally present as monomineralic and polymineralic clots. The same is observed in the olivine crystals, featuring glomeroporphyritic aggregates. Interstitial space is filled with anorthoclase. By SEM/EDS performed in Japan andesitic glass inclusions within the plagioclase were also identified.

Trachybasalt (GRA-SM1) is composed of plagioclase, olivine and opaque minerals, set in an intergranular groundmass where the same minerals are present but with lower olivine and higher pyroxenes content. Olivines sometimes occur in monomineralic clots.

Petrographic examination showed that, with the recommendations in [24], most samples do not contain reactive minerals with the exception of TER-SM1 (interstitial silica) and SMG-SM1 (volcanic glass) samples. According to Portuguese standard [12], these aggregates would be classified as potentially reactive if the reactive forms of silica exceeded 2% of the bulk rocks. Due to the small sizes of the crystals, no point counting was done under the transmitted light, but other tests were applied to assess the content of free silica.

Rock chemistry

Table 2 summarises the chemical composition of the samples tested. A classification according to the TAS diagram [25], which slightly differs from the classical one [26], shows that most of the rocks are basalts with a limited degree of evolution, and that all the samples belong to the alkaline series. This is consistent with the works [27] published on the geochemistry of Azorean rocks. Basanite (SMA-SM1) has a relatively high value of ignition loss (LOI), suggestive of an alteration that formed iddingsite and analcite. Calculation of CIPW norm indicates that all the basalts are undersaturated with silica yielding a variable content of nepheline and/or olivine in the normative mineralogy (Table 3). Trachyte (TER-SM1) is oversaturated with silica and has normative quartz (Table 3), which was confirmed by the phosphoric acid test.

ASR tests

The chemical test (Figure 3) [18] revealed that two (TER-SM1, TER-SM2) of the five samples were deleterious, while the others were innocuous. The accelerated mortar bar test of trachybasalt (SMG-SM1) showed an average of 0.01% expansion, indicative of non-potential reactivity, according to the expansion limit of 0.10% at 14 days [20], but was extended up to 28 days to confirm the results (Figure 4). The accelerated concrete prism test (AAR-4) indicated that the SMG-SM1 basalt is considered non-potentially reactive, based on the recommended limit in AAR-4.1 [21] that aggregates with expansions less than 0.02%, or 0.03% [28] at 20 weeks can be classified as non-reactive (Figure 4). The concrete prism test (AAR-3) [22] showed an interim result with an increase in the expansion rate after 140 days, but definitive conclusions can only be taken at the end of 1 year of testing.

4 DISCUSSION

In general, conventional thin-sections (thickness 30 μm) used by the geologists are too thick to identify or quantify potentially reactive silica by point counting in optical microscope without the use of reflected light, due to the small size of crystals in the groundmass of volcanic rocks. Therefore, other supplementary methods have to be used to verify the occurrence of deleterious forms of silica (e.g. [1,4]).

Volcanic glass

The petrographic study showed that most of the samples do not contain potentially reactive silica forms. However, the samples of trachybasalt (SMG-SM1) and basalt (TER-SM2) contained some amount of volcanic glass of andesitic composition (SiO_2 58% and 55%, respectively) in the groundmass interstices. The amount of the glass varies depending on the sample sites in the same quarry. According to Katayama [1,29], andesitic glass is not deleterious, but volcanic glass becomes deleterious with increasing the SiO_2 content (>62-65%) in the course of crystallization of the magma, e.g. dacitic glass and rhyolitic glass: with fresh Icelandic basalt (SiO_2 50%), this limit corresponds to the content of interstitial glass <30-35 vol%. The presence of dacitic or rhyolitic glass has been described by many authors to be associated with ASR [1,2,4,6,8,29,30]. A wide range of volcanic rocks are considered reactive when rhyolitic interstitial glass is present in the groundmass of basalt, andesite and rhyolite [1], but basaltic rocks are non-reactive when they are fresh and entirely composed of basaltic glass sideromelane [4]. Aggregates will be highly reactive when altered to contain opal, besides cristobalite and rhyolitic glass. All the studied samples are recent, except for the basanite (SMA-SM1) of Pliocene age, which was slightly altered. The trachybasalt (SMG-SM1) is generally fresh and it is the only one containing interstitial glass.

Rock chemistry and silica minerals

The basaltic rocks did not exceed 50% of SiO_2, while the trachyte (TER-SM1) had more than 60% SiO_2 with oversaturated silica (normative quartz) in the CIPW norm. According to [1,4], fresh volcanic rocks must contain more than 50% SiO_2 to show potential reactivity, and oversaturated silica generally appears as reactive quartz polymorphs cristobalite and tridymite. In well crystallized volcanic rocks, the amount of the normative quartz nearly corresponds to that of the free silica as extracted by the phosphoric acid method [29]. With the trachyte sample (TER-SM1), the presence of the colourless irregular-shaped microcrystalline silica in the interstices was confirmed by SEM/EDS and elemental mapping of Si, and this silica was extracted by the phosphoric acid treatment. For other rock types, none of the silica form has been identified or extracted. The form of the silica polymorph can be identified by X-ray diffraction (XRD), as stated in [4, 24].

ASR tests

Two samples, trachyte (TER-SM1) and basalt (TER-SM2), tested as deleterious according to the chemical test [18]. In this test, rocks with low dissolved silica (Sc) and low reduction in alkalinity (Rc) are classified as innocuous, whereas rocks with silica minerals cristobalite and tridymite show higher Sc and Rc being considered potentially deleterious indicative of pessimum proportions [1,4]. However, some altered aggregates are often misjudged as innocuous in this test and reactive in the mortar bar test, while others are identified incorrectly as reactive [3]. In such case, Rc is raised by smectite by the cation exchange, which occurs abundantly in altered basalt of Neogene age like the Hvalfjordur sand in Iceland [4]. The potential reactivity of the trachyte can be interpreted by the presence of crypto- to microcrystalline quartz [4,28]. For the basalt (TER-SM2), there seems to be no clear explanation for the results obtained, hence further tests are necessary to clarify and understand the potential reactivity of this aggregate. However, the andesitic glass inclusions identified by EDS analysis within the plagioclase phenocrysts could explain the potential reactivity in the chemical test (dissolved silica). The chemical method as well the mortar bar test is widely used in Japan and New Zealand. According to [2], some studied Japanese rocks showed that in both methods the same rock types were considered reactive. Experiences show that basaltic aggregates of the smectite alteration are often misjudged as innocuous in the chemical method, due to high values of Rc [4].

The trachybasalt (SMG-SM1) tested as non-reactive by [18], but it was thought necessary to run the expansion tests due to the presence of andesitic interstitial glass. The accelerated mortar bar test [20] gave expansion less than 0.10%, even at the end of 28 days. In this test, Turkish andesitic aggregates (SiO_2 54-61%) with interstitial volcanic glass showed potentially deleterious expansion (> 0.10%) [6], although they were described as basalt, and composition of the glass was not determined. In our study, only a small amount of andesitic glass (SiO_2 58%), not a sideromelane, was present in the basalt. Wigum *et al.* [3] studied several Icelandic basaltic aggregates mainly of basaltic sand and gravel, and showed that the majority have deleterious expansions at 14 days. This indicates that this test is adequate to these rocks. The accelerated concrete prism method also indicated that SMG-SM1 aggregate is considered non reactive. The results of the concrete prism method AAR-3 [22] will be crucial to understand the behaviour of this aggregate.

5 CONCLUSIONS

In order to assess the potential alkali reactivity in the lavas of the Azores archipelago, several test methods were performed on selected volcanic aggregates:

- Petrography identified a silica mineral in one of the samples trachyte (TER-SM1), which is in agreement with the silica oversaturated character, as confirmed by the CIPW norm.
- In the trachybasalt (SMG-SM1), small amounts of andesitic glass were identified by petrography and by EDS analysis. CIPW norm did not detect any free silica and, according to the chemical test, this aggregate was classified as non reactive.
- The accelerated laboratory tests performed to evaluate the performance of the aggregate, so far have not revealed potential reactivity of the trachybasalt (SMG-SM1). However, different sectors of the quarry show variable amounts of volcanic glass that should be taken into account in future studies.
- Basalt sample (TER-SM2) was free of reactive silica based on the petrography and CIPW norm. However, SEM/EDS lead to the identification of andesitic glass and as the chemical test indicated deleterious reactivity of this aggregate, further tests should be carried out to verify the presence of reactive silica.
- As for the basanite (SMA-SM1) and trachybasalt (GRA-SM1), none of the petrographic analysis, CIPW norm and the chemical test suggested the presence of reactive constituents.

Following the interim results above presented, a research project will start in 2012 in which a new and wider sampling campaign will be carried out and further tests performed in order to obtain detailed information on Azorean aggregates.

ACKNOWLEDGEMENTS

The analyses by SEM/EDS were performed at CEMUP which equipment was funded by the projects REEQ/1062/CTM/2005 and REDE/1512/RME/2005 of Portuguese Foundation for Science and Technology (FCT). The authors also acknowledge the financial support of the FCT under the projects PTDC/CTM/65243/2006 and PDTC/ECM/115486/2009.

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Table 1: Petrographic characteristics of the samples (based on [23]).					
Sample	SMA-SM1	SMG-SM1	TER-SM1	TER-SM2	GRA-SM1
Island	Santa Maria	São Miguel	Terceira		Graciosa
Classification*	Basanite	Trachybasalt	Trachyte	Basalt	Trachybasalt
Paragenesis[1]	ol+cpx+pl+op	ol+cpx+pl+op	f+ne+ae	ol+cpx+pl	ol+pl+op
Age [26]	4.13 Ma	1652 A.D. (?)	-	2115 BP	2000 BP
Notes	Iddingsite rim to olivine	Quartz xenocryst; volcanic glass	Interstitial silica, as quartz	Andesitic glass	

*According to Le Maitre *et al.* (1989)
[1] ol: olivine; cpx clinopyroxene; ae: aegirine; pl: plagioclase; op: opaques; f: feldspars sanidine and anorthoclase; ne: nepheline.

Table 2: Geochemical composition of the samples [23].					
Major Oxides (%)	SMA-SM1	SMG-SM1	TER-SM1	TER-SM2	GRA-SM1
SiO_2	43.31	47.25	64.79	48.54	47.7
Al_2O_3	12.98	14.09	13.92	14.65	16.76
Fe_2O_3-total	11.88	12.2	6.1	12.12	11.11
MgO	11.05	8.06	0.65	6.82	7.15
CaO	11.52	9.16	0.71	10.88	9.58
Na_2O	3.62	3.25	6.35	3.09	3.82
K_2O	0.76	2.04	4.97	0.97	1.36
TiO_2	2.29	3.48	0.61	3.11	2.74
P_2O_5	0.42	0.61	0.07	0.59	0.56
MnO	0.17	0.16	0.22	0.17	0.17
LOI	2.15	-0.2	1.13	-0.1	-0.73
SUM total	100.2	100.1	99.51	100.8	100.2

Table 3: Calculation of CIPW* norm [23].					
Normative minerals	SMA-SM1	SMG-SM1	TER-SM1	TER-SM2	GRA-SM1
q (quartz)	0.0	0.0	9.07	0.00	0.0
or (orthoclase)	4.5	12.1	29.37	5.73	8.0
ab (albite)	6.6	21.0	43.93	26.15	21.9
an (anorthite)	16.9	17.8	0.00	23.24	24.6
lc (leucite)	0.0	0.0	0.00	0.00	0.0
ne (nepheline)	13.0	3.5	0.00	0.00	5.6
kp (kaliofilite)	0.0	0.0	0.00	0.00	0.0
ns (sodium metasilicate)	0.0	0.0	1.70	0.00	0.0
ks (potassium metasilicate)	0.0	0.0	0.00	0.00	0.0
ac (acmite)	0.0	0.0	2.19	0.00	0.0
di (diopside)	30.2	19.1	2.65	21.91	15.8
hy (hypersthene)	0.0	0.0	6.61	2.23	0.0
ol (olivine)	17.8	14.7	0.00	10.69	16.5
il (ilmenite)	4.4	6.6	1.15	5.90	5.2
mt (magnetite)	2.6	2.6	0.00	2.68	2.9
ap (apatite)	0.99	1.4	0.17	1.40	1.3

* CIPW norm: method of normative mineral calculation first devised in 1903 by four American petrologists: Cross, Iddings, Pirsson and Washington.

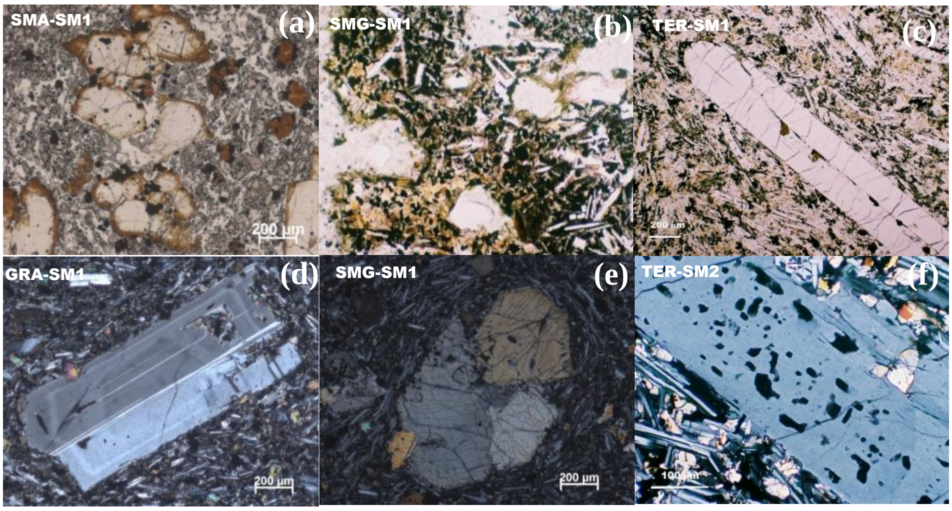

FIGURE 1: Photomicrographs of the volcanic aggregates studied: (a) Olivine with iddingsitized rims in SMA-SM1 sample (PPL); (b) Volcanic glass as an intergranular filler in SMG-SM1 sample (PPL), (c) Sanidine phenocrysts in TER-SM1 sample (PPL), (d) Zoned plagioclase in an intergranular groundmass in GRA-SM1 sample (XPL); (e) Clinopyroxene crystals in SMG-SM1 samples (XPL) and (f) Plagioclase phenocryst containing inclusions of andesitic glass in TER-SM2 sample (XPL).

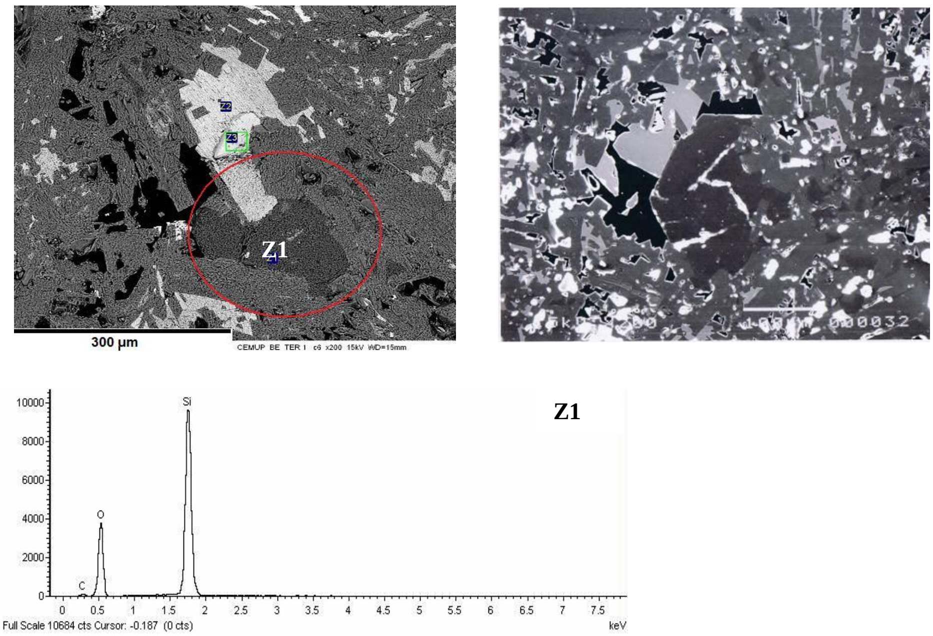

FIGURE 2: Silica in trachyte, presently microcrystalline quartz (TER-SM1): Irregular-shaped (left), and euhedral (right). SEM image and EDS spectrum [left, 23].

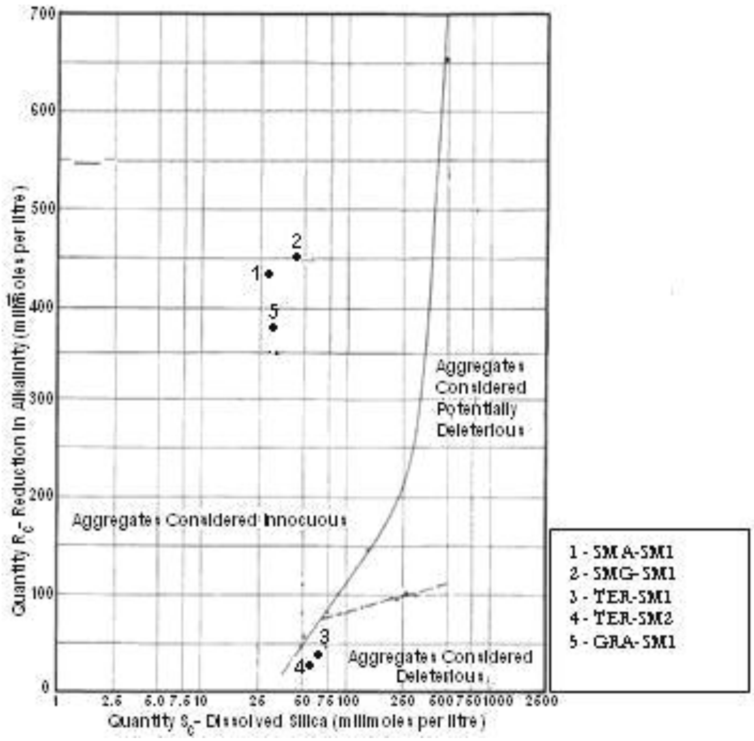

FIGURE 3: ASTM C 289-07 chemical test of basaltic rocks (1,2,4,5) and trachyte (3).

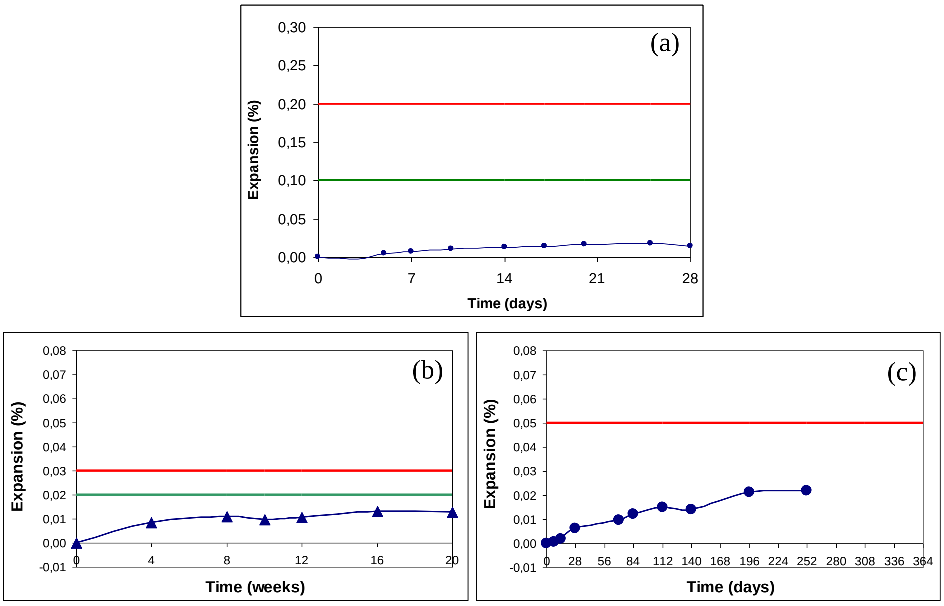

FIGURE 4: Expansivity test results of trachybasalt (SMG-SM1) sample [23]: a) ASTM C 1260-07; b) AAR-4; c) AAR-3.