

ALKALI-SILICA EXPANSION OF HEAVY AGGREGATES USED FOR NUCLEAR SHIELDING CONCRETE

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ABSTRACT

In the present study the potential appearance of the alkali-silica reaction (ASR) in heavy aggregates was studied. ASTM C1260 Standard Test Method for Potential Alkali Reactivity of Aggregates (Mortar-Bar Method) was applied. In order to investigate the effect of the content of alkalis in cement on the expansions due to ASR, three levels of total and soluble alkali content of cement were studied. Three portland cements Type I with different alkali content were selected. Two ordinary portland cements, which are commonly available in the market and one special cement were tested. That cement was specially made for the purpose of the nuclear shielding concrete CEM I NA-SR-LH of low-alkali, increased sulphate resistance and low heat of hydration.

For the tests according to ASTM C 1260 the high-density aggregates, known as absorbing gamma radiation were selected: barite, magnetite and hematite. The expansion test revealed that hematite was highly reactive, regardless of the type of cement. Already after four days of storage in 1 N NaOH and 80°C the mortar bar expansion exceeded the limit of 0.1%, and after next four days was more than 0.2%, which qualifies it extremely reactive aggregate. Other aggregates after 14 days of testing did not exceed 0.1% elongation limit, but the influence of the type of cement was noticed. There was a noticeable tendency for increasing the total expansion with increasing the alkali content of cement.

Keywords

Alkali-Silica Reaction (ASR), high density aggregate, cement composition

INTRODUCTION

Recent observations of premature deterioration of nuclear shielding concrete structures in Seabrook Power Plant (US, NH), including electrical tunnel, containment enclosure building, control building and tank farm area have suggested the development of the alkali-silica reaction (ASR), [1]. The performed petrographic examinations confirmed that ASR was the main reason of concrete degradation, [2]. Expansion produced by alkali-silica reaction has been observed in the turbine generator foundation of the Unit 1, also, Ikata nuclear power station (NPP), in Japan, [3]. ASR has been confirmed at Gentilly Units 1 and 2 in Becancour in Canada, and in Saint-Laurent NPP in Saint-Laurent, France, [4]. The concrete degradation due to ASR on the external face of the containment building of Tihange 2 in Belgium has been observed as well, [5]. These examples demonstrate that the issue of ASR was not considered seriously during the material selection for NPP structures. Regardless of the location of concrete structures in NPP alkali aggregate reaction occurred - ground water (Seabrook) or rain (Tihange 2). The proper

characterization and selection of the aggregate, is crucial in order to prevent such deleterious reaction. There is no generally accepted technology to mitigate the effects of ASR in an existing concrete structure, so the issue of potentially reactive aggregate should be resolved before the construction of the nuclear plant buildings.

Since ASR is a slow process, the harmful chemical and environmental effects of concrete may be seen after years or decades. Therefore, almost all ASR tests involve the acceleration of the reaction by aggravating one or more of the following influential factors, [6-8]:

- Alkali concentration,
- Temperature,
- Humidity.

The ASTM C1260 Standard test method for potential alkali reactivity of aggregates (Mortar-Bar Method) [9] has been intensively used all around the world, [7, 10]. This method was originally designed to assess aggregate reactivity and is recognized as a very severe diagnostic method because of the extreme test conditions, specifically the use of a highly alkaline solution (1N NaOH) and high temperature (+80°C), 6. It was assumed that the alkali content of the cement has negligible or minor effect on the expansion in this test because the specimens are stored in a 1N NaOH solution. ASTM C1260 requires the use of Portland cement that meets ASTM C150 standard specification for Portland cement. However, with regard to the alkali content, a wide variety of cement (Type I÷V) can be used, as they have max. alkali content equal to 0.6%.

It was commonly accepted that expansion due to ASR reaction is unlikely to occur when the alkali content of the cement is below 0.6% $\text{Na}_2\text{O}_{\text{eq}}$, [6, 10], but the great majority of cases of concrete structures reported as showing deterioration due to alkali-silica reaction were made using high-alkali cement, [11, 12].

The Portland cement, beside parameters such as aggregates, chemical admixtures, de-icing salts, or supplementary cementing materials (SCM), is the main contributor of alkalies. The expansion resulting from the ASR increases with an increase alkali content in concrete, [12,13]. Berra et al. [10] tested natural sands with ASTM C1260, and concluded that the original alkali content of the portland cement is a significant factor in affecting mortar-bar expansion. Clinker alkali had no or little effect on moderately reactive aggregates; whereas it had considerable effect on highly reactive aggregate. Highly reactive aggregate tended to be more sensitive to cement alkalinity. It is possible that the alkali content of cement will change the classification for the aggregates near the boundary between reactive and nonreactive aggregates.

The investigation of the possible ASTM C1260 expansion of different heavy aggregates due to ASR is reported in this paper. Tests were focused on heavy aggregates for nuclear shielding concrete (barite, magnetite, hematite) used against gamma radiation and on the influence of the alkali content of cement on the expansion due to ASR.

MATERIALS

The aggregates investigated in this study were acquired from four different quarries. Barite aggregate was imported from two mines localized in Europe that were marked as B1 and B2. Magnetite (M) was also collected from European mine, and hematite (H) was from Africa. Table 1 shows the main components of aggregates after XRF analysis. The aggregates were characterized with following densities: barites 4.2 g/cm³, magnetites 4.8 g/cm³ and hematites 5.1 g/cm³. High density makes them useful for heavyweight concrete, allowing for a designed high density of concretes, as it is specified in shielding elements.

Table 1. Main components of heavy aggregate after XRF method in %, supplier's data

Main mineral constituents	Barite B1	Barite B2	Magnetite	Hematite
BaSO ₄	92	77.5	N/A	N/A
CaF ₂	3.0	N/A	N/A	N/A
SiO ₂	3.0	2.0	3.0	11.5
Fe ₂ O ₃	N/A	N/A	90.8	85.1
FeCO ₃	N/A	12.0	N/A	N/A
Al ₂ O ₃	N/A	N/A	0.4	0.9
CaO	N/A	N/A	2.5	0.05
K ₂ O	N/A	N/A	0.2	0.4
Na ₂ O	N/A	N/A	0.3	0.02

Aggregates B1, B2, M, and H were characterized also using X-ray diffraction to identify their constitutive minerals. The detailed results are described in [8, 14]. The mineral composition of aggregate B1 was barite, quartz and fluorite, for aggregate B2 – barite, siderite and quartz, aggregate M – magnetite, cordierite and fluorapatite; and aggregate H – hematite and quartz. All XRD analysis confirmed the mineral composition of the tested aggregate obtained from the suppliers. Additionally the XRF analysis has been used to verify the suppliers' data and the obtained results confirmed the chemical composition of the heavy aggregates, [15].

In order to investigate the effect of alkali content in cement on the expansions due to ASR, three levels of total and soluble alkali content in cement were investigated, Table. 2. Cements C1 and C2 are commonly used in concrete technology, CEM I according to European Standards and are available at the market. Portland cement C3 was manufactured at semi-technical scale in the Institute of Ceramics and Building Materials. Cement C3 was designed as low-alkali, low heat of hydration and sulphate resistant Portland cement.

Table 2. The main chemical constituents in tested cements, in wt.% by XRF

Main chemical constituents	Cement 1 C1	Cement 2 C2	Cement 3 C3
SiO ₂	22.20	19.03	21.48
Al ₂ O ₃	5.30	4.84	4.80
Fe ₂ O ₃	3.00	3.22	2.62
CaO	66.30	63.64	65.60
MgO	1.30	1.15	0.87
SO ₃	0.60	2.97	2.84
Na ₂ O	0.24	0.21	0.12
K ₂ O	0.82	0.53	0.47
Total Alkalis, Na ₂ O _{eq}	0.78	0.56	0.43
Water-Soluble Alkalis	0.54	0.48	0.37
LOI	3.5	3.34	1.12

TESTING PROCEDURE

Mortar-Bar Method

ASTM C1260 Standard Test Method was used that allowed to detect the potential for deleterious alkali-silica reaction of aggregate in mortar bar exposed to NaOH solution in 80°C.

Mortar bar specimens of dimensions 25×25×285 mm were prepared, the aggregate was crushed and sieved according to standard requirements. Aggregate-to-cement ratio of 2.15 and water-to-cement ratio (by weight) of 0.47 were maintained. Three mortar bars were prepared for each aggregate and cement type. The mortars were mixed and molded within a total elapsed time of less than 2 minutes and 15 seconds. The mortar bars were kept in 23±2°C and RH ≥ 95%, for 24 hours. After 24 hours in mould the mortar bars were stored for the next 24 hours in water in 80±1°C. After that their initial, zero readings were recorded by a digital extensometer before immersing in 1N NaOH at 80°C for 14 days. Subsequent expansion readings were taken at least three times during the test. The final assessment of the potential reactivity of the tested aggregates is carried out on the basis mortar bar expansion, Table 3.

Table 3. Assessment of Reactivity of Aggregate per ASTM C 1260, [9]

Expansion, %	Aggregate reactivity
< 0.1	Innocuous
0.1 ÷ 0.2	Inconclusive
> 0.2	Potentially deleterious

Thin section analysis

The fluorescent epoxy impregnated thin sections were prepared. The aggregate grains were vacuum impregnated and then cut into slices. The aggregate was grinded and polished to 20 µm thickness. Thin section analyses were carried on using the optical polarizing microscope Olympus BX51 connected to a digital camera. Thin sections were examined in plane polarized light (PPL), crossed polarized light (XPL), also with lambda plate. The identification and analysis of minerals was performed in PPL and XPL.

TEST RESULTS AND DISCUSSION

Results of the expansion of the mortar bars as function of the time of their storage in 1N solution of NaOH and at 80°C are shown in Fig. 1-3.

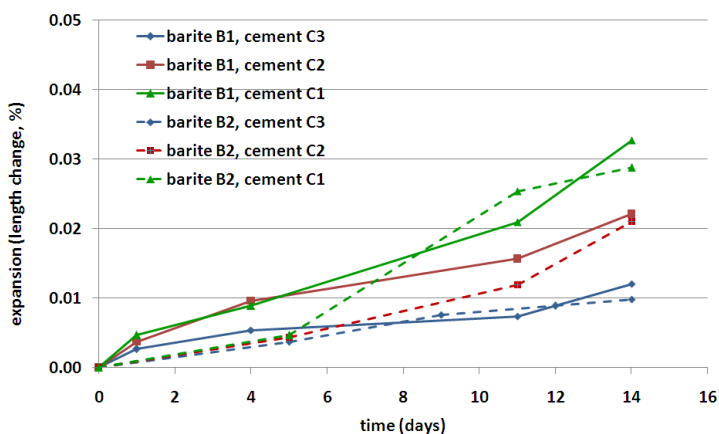


Fig. 1. Expansion of mortar bars (barite aggregate, B1 and B2) stored in 1N NaOH and temperature 80°C during 14 days

Figures 1-3 show the progression of mortar expansion during 14 days of testing. As can be seen, the expansion increased with increasing test duration, and the rate of expansion over the time varied depending on the aggregate and cement type. The expansion development was faster and more extensive for hematite aggregates than that of the other heavy aggregates.

After 14 days of testing two kind of barite, B1 and B2, and magnetite aggregate showed rather slow and small increase of the mortar bar elongation. Barite aggregates B1 and B2 differed in chemical composition but the final results of the elongation after 14 days of testing were similar. The greater difference can be seen in mortars with other type of cement. Regardless of the aggregates, B1 or B2, the degree of expansion increased with increasing alkali content of cement.

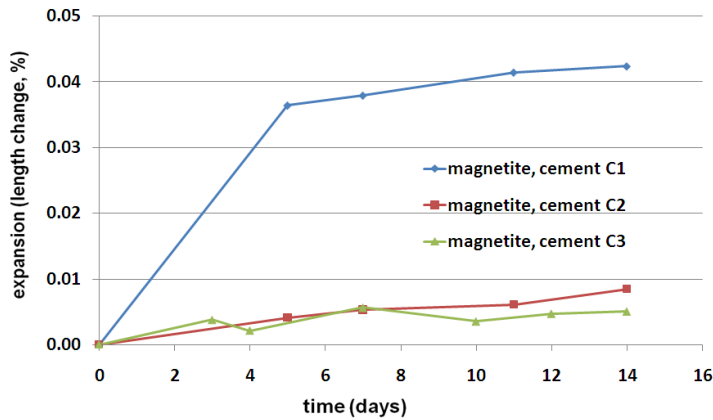


Fig. 2. Expansion of mortar bars (magnetite aggregate, M) stored in 1N NaOH and temperature 80°C during 14 days

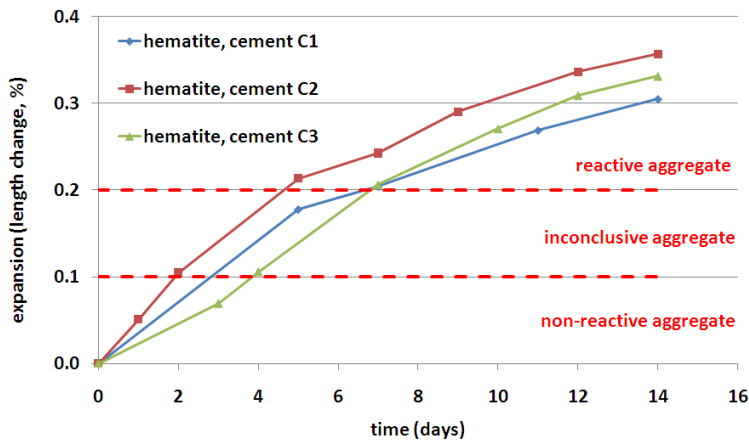


Fig. 3. Expansion of mortar bars (hematite aggregate, H) stored in 1N NaOH and temperature 80°C during 14 days

In magnetite aggregate, the highest elongation achieved mortar bars with cement C1, the highest alkali content, but below the 0.1% limit value. The best results were obtained in mortar bars with both, cement C2 and C3, showing almost no expansion (<0.01%).

The hematite mortar expansion was extremely fast and high, and after max. four days exceeded the 0.1% limit. The mortar bar elongation above 0.20% indicates very reactive aggregates, and all the hematite specimens after 14 days of standard storage showed an expansion of about 0.3-0.35%. In opposite to the barite and magnetite aggregates, in hematite mortars the content of the alkali in cement did not influence the final expansion. The highest elongation was observed in mortars with cement C2 – moderate content of alkalis, and the lowest with cement C1 – the highest content of alkalis.

In the Fig. 4 the characteristic map of cracking due to alkali-silica reaction expansion at the lateral surface of a hematite mortar bar is shown.

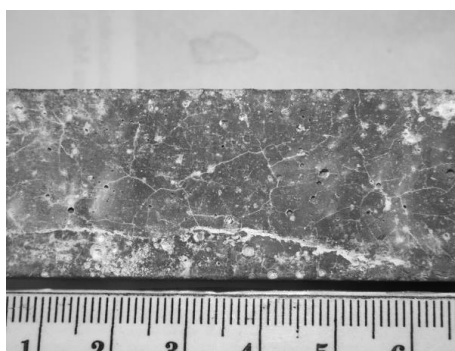


Fig. 4. ASR characteristic map of cracking at the surface of hematite aggregate after 14 days in 1N NaOH and 80°C

The maximum increase in expansion was observed between the first 6 days, and a moderate increase in expansion was noted between 8 and 14 days for hematite mortar bars. The potential for a high reactivity of hematite aggregates was indicated by microscopic test on thin sections. The thin section analysis revealed the presence of the microcrystalline quartz, Fig. 5. In hematite small size crystals of SiO₂ have been found and their size of crystals was comprised between 15 and 60 μm, what makes them microcrystalline and are small enough to be visible in microscope. These kinds of SiO₂ crystals are prone to ASR in concrete, 14.

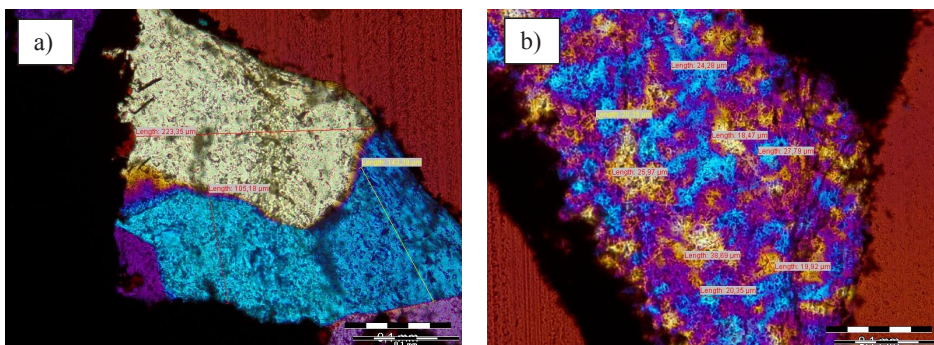


Fig. 5. Grains of hematite aggregate (H) with visible quartz crystals: a) fine-grained crystals, b) microcrystalline quartz; XPL with lambda plate, scale bar = 100 μm

The examples of the evidence of alkali-silica gel in cement matrix, in hematite aggregate and in air-voids are presented in Fig. 6. The characteristic alkali-silica gel was present in small air-voids and partially fulfilled larger voids ($> 500 \mu\text{m}$).

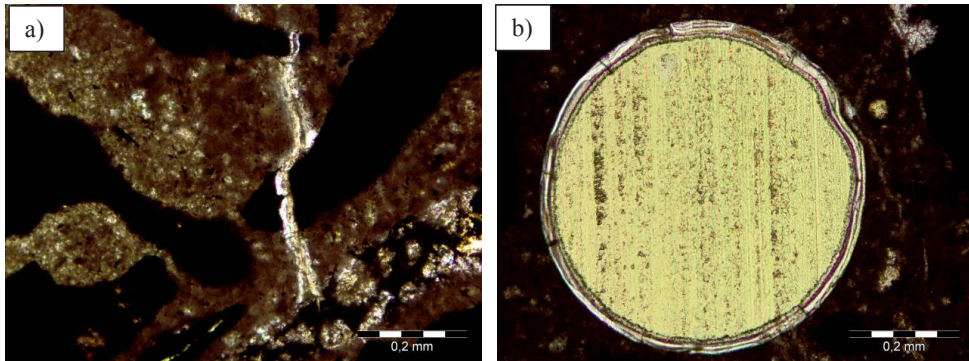


Fig. 6. ASR signs in mortar with hematite aggregate: a) gel in cracked matrix and aggregate, b) gel in air void; PPL (scale bar = $200 \mu\text{m}$)

CONCLUSIONS

From the experimental results, the following conclusions can be drawn:

- The ASTM C 1260 test showed varied results of the expansion of heavy aggregates for nuclear shielding concrete,
- Mortar bars with hematite aggregate showed a very fast and rapid expansion after 4 days, and after next 4 days – the expansion significantly exceeded allowable limit of 0.2%.
- Mortar bars with barite and magnetite aggregate revealed acceptable expansion after 14 days of testing, less than 0.05%.
- As the alkali content in cement increased, the expansion obtained according to ASTM C 1260 also increased, regardless the level of aggregate reactivity.
- Content of alkalis in cement did not significantly affect the expansion of the mortar made with highly-reactive aggregate, but apparently had a significant effect on the expansion of the mortar made with the aggregate treated as non-reactive.
- Alkali content in cement did not affect the decision on potential reactivity of the heavy aggregates, but it evidences that it might suggest the proper choice of the cement for the nuclear shield concrete production, depending of the environmental conditions.

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