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Alkali–silica reactivity of some frequently used lightweight aggregates

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Abstract

Lightweight aggregates (LWAs) are frequently used in concrete as well as in thermally insulating mortars and grouts, so that information on their alkali–silica reactivity (ASR) is very important. Four LWAs—expanded vermiculite, expanded clay, expanded glass and perlite—were studied regarding their ASR, using the following test methods: the accelerated mortar bar test (ASTM C 1260), the rapid chemical test (ASTM C 289) and the combined scanning electron microscopy–energy dispersive X-ray technique (SEM–EDX). According to these methods, neither the expanded vermiculite nor the expanded clay exhibited any potential ASR. On the other hand, in the case of the aggregates containing a glassy phase (expanded glass and perlite), the results of SEM–EDX analysis showed serious decomposition of aggregate texture due to ASR, although no deleterious expansion was observed in the accelerated mortar bar test. Therefore, suitable test criteria for ASR need to be defined for LWAs of this type when the AMBT method is used, as has already been suggested for slowly reactive aggregates in Australia.

Keywords: Lightweight aggregate; Alkali–aggregate reaction; SEM; EDX

1. Introduction

Different types of lightweight aggregate (LWA) suitable for construction purposes can be found on the market, varying in their composition, density, surface texture, porosity and water absorption capacity. The most frequently used LWAs are expanded clay, expanded glass, perlite, expanded vermiculite and sintered ash. Most LWAs contain a glassy phase and/or other alkali-sensitive minerals, and should therefore always be treated as potentially reactive. They may react expansively with quite low levels of cement alkalis, and the possibility of alkali–silica reaction (ASR) must always be taken into account.

The available literature from this field is somewhat contradictory. Some researchers who have studied ASR both in natural and in artificially prepared LWAs report that there are practically no signs of this reaction in concretes and mortars made from such aggregates [1,2]. Others [3,4], however, have demonstrated significant damage to these cement composites due to ASR, which was such that the structures affected had to be either demolished or extensively repaired. Many researchers have found that ASR occurs in some LWAs (perlite, expanded glass, expanded clay), but shows no harmful effects, such as expansion and/or efflorescence [5]. It is supposed that the pores in the glassy phase of LWAs accumulate the gel produced, which is less prolific, more "sticky" and more readily carbonated [6], and thus indirectly prevents expansion. Concrete additives such as air-entraining agents may, on the one hand, allow the gel to withdraw into the pores and cause no expansion, but on the other hand they may reduce the freezing and thawing resistance of the concrete [7]. A reduction of expansion due to accommodation of the alkali–silica gel in the air-void system has also been reported [8]. About 6% air voids reduced the expansion of mortar bar prisms by about 50%.

The objective of the investigations described in the paper was to ascertain which of the widely available and frequently used artificial LWAs would exhibit an ASR, and could therefore inflict damage if used. If such an aggregate was used for structural concrete, the consequences could be serious. If used for thermally insulating mortar or plaster, damage in the form of cracks would not be so dangerous but would still be very unacceptable because it would affect the durability and aesthetic properties of structures.

It is true that the new European standard for the use of LWAs in concrete, mortar and grout (EN 13055-1), which admits the possibility of this reaction, prescribes preventive

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Table 1
Water absorption, specific surface areas, and main mineralogical components of the investigated LWAs

Type of lightweight aggregate	Water absorption (% by mass)	Specific surface area (m^2/g)	Mineralogical components
Expanded vermiculite	162	3.73	Dehydrated vermiculite
Expanded clay	56	0.30	Quartz, feldspar, enstatite, glass
Expanded glass	48	0.37	Glass
Perlite	230	0.97	Glass

Table 2
Results of semiquantitative chemical analysis of the investigated LWAs

	Expanded vermiculite	Expanded clay	Expanded glass	Perlite
SiO_2	44.4	47.0	71.2	76.5
CaO	–	8.0	10.1	1.3
MgO	23.4	4.7	2.0	–
Al_2O_3	13.1	20.2	1.8	13.4
FeO	11.1	14.9	0.4	1.4
Na_2O	–	0.7	13.5	3.0
K_2O	6.6	3.7	0.9	4.3
TiO_2	1.4	1.3	–	–

measures, such as the incorporation into the concrete of materials with a low alkali content and combinations of nonreactive aggregates, and the setting of limits to the degree of saturation of concrete/mortar with water, but it does not prescribe any specific test methods nor any specific permitted limits. For this reason, the authors wished to verify to what extent some of the existing methods for the determination of the alkali–aggregate reaction of natural aggregates are suitable for the evaluation of light aggregates.

2. Experimental

2.1. Materials

Four of the most common types of LWA on the Slovenian market were investigated: vermiculite, expanded clay, expanded glass and perlite.

Vermiculite is a naturally occurring sheet-silicate mineral, resembling mica in appearance, which exfoliates when heated to 750–1100 °C as a result of the mechanical separation of the layers due to the rapid conversion of interlayer water to steam. Expanded clay is produced by firing natural clay, which swells at 1000–1200 °C due to the action of the gases generated inside the mass. Expanded glass is produced by mixing together finely ground glass and foaming agents, and firing the mixture at a temperature where foaming takes place (generally above 800 °C). Perlite is obtained from pumice, which is a glassy form of rhyolitic or dacitic magma. It contains a few percent of water, which causes expansion due to the expelling of this water on heating at 900–1200 °C.

2.2. Characterization of the investigated LWAs

The water absorption, specific surface areas and main mineralogical components of the investigated LWAs are

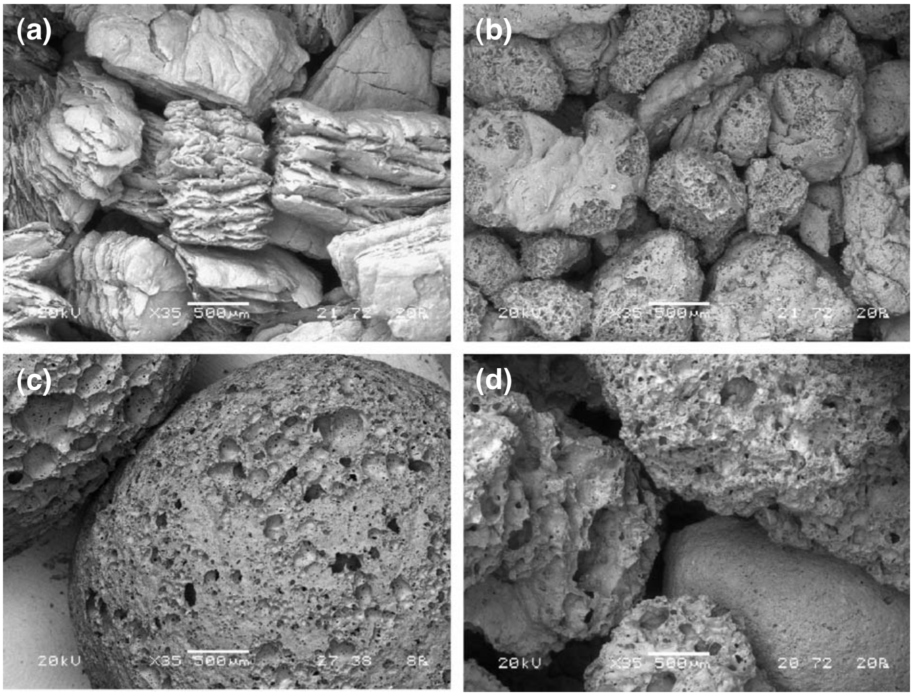

Fig. 1. SEM micrographs of (a) expanded vermiculite, (b) expanded clay, (c) expanded glass and (d) perlite.

Table 3
The Rc and Sc factors of the investigated LWAs

	Expanded vermiculite	Expanded clay	Expanded glass	Perlite
Rc (mmol/l)	671.9	82.5	− 917.9	198.1
Sc (mmol/l)	17.3	101.9	2890.0	1420.4

presented in Table 1, and the results of semiquantitative chemical analysis using the energy-dispersive X-ray technique (EDX) in Table 2.

Scanning electron microscopy (SEM) micrographs of expanded vermiculite (a), expanded clay (b), expanded glass (c) and perlite (d) are presented in Fig. 1, in which the open-flaked texture of the expanded vermiculite and the irregular to regular, rounded, highly porous granules of the other three LWAs can be clearly seen.

2.3. Test methods

Standard water absorption tests (ÖNORM 3233) were carried out on the LWAs, and their specific surface areas were determined using the BET method.

The mineral composition of the LWAs was determined by XRD analysis. Their pore texture and morphology, as well as their semiquantitative chemical composition, were determined by SEM–EDX analysis.

The ASR of the LWAs was studied using the chemical test method described in ASTM C 289, and the expansion of mortar bars according to ASTM C 1260.

Because of the lightweight nature of the aggregates, the ASTM C 1260 method was modified with regard to the aggregate particle size distribution and the aggregate-to-cement ratio. The cement used contained 0.85% Na_2O equivalent. The cement-to-aggregate ratio and the w/c factor were 4.5:1 and 0.47, respectively. The mortar bars were prepared from room-dry samples of the bulk LWAs.

The reactions between the LWAs and the cement matrix during the accelerated mortar bar test were observed and evaluated by low vacuum scanning electron microscopy (LV-SEM) and by the EDX method.

3. Results

3.1. Alkali–silica reactivity according to ASTM C 289

The values of the Rc and Sc factors for the LWAs according to ASTM C 289 are shown in Table 3, and also in Fig. 2, in the diagram prescribed by this standard. The Rc value, which represents the reduction of alkalinity of the NaOH solution due to ASR, is usually above zero (when testing natural dense aggregate). However, in the case of the expanded glass, the obtained Rc value of − 917.9 mmol/l was significantly below zero, indicating an increase in the alkalinity of the original solution due to the release of alkalis from the aggregate. The calculated initial alkalinity of the solution before the reaction was 999.3 mmol/l, and after the reaction, 1917.2 mmol/l. This Rc value was determined by titration to phenolphthalein, as prescribed in ASTM C 289. However, Dent Glasser and Kataoka [9] have shown that, in the case of a high concentration of silica in the solution, the use of phenolphthalein as an indicator makes the end point indefinite, and lower than it should be. From these findings, it follows that the true value of Rc should, in the case of the investigated expanded glass and the perlite, be higher than the value obtained. The difference between these two values is the result of the high concentration of silica in the test solutions.

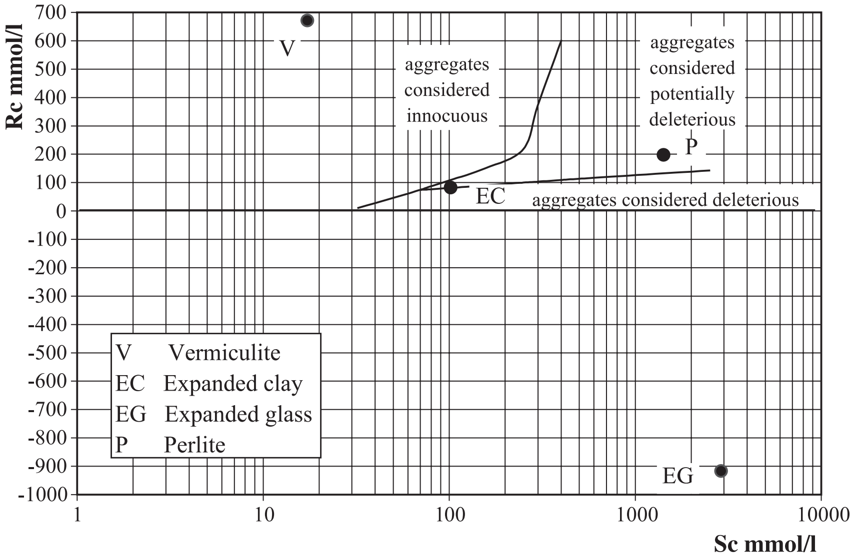

Fig. 2. Location of the samples in the Rc–Sc diagram.

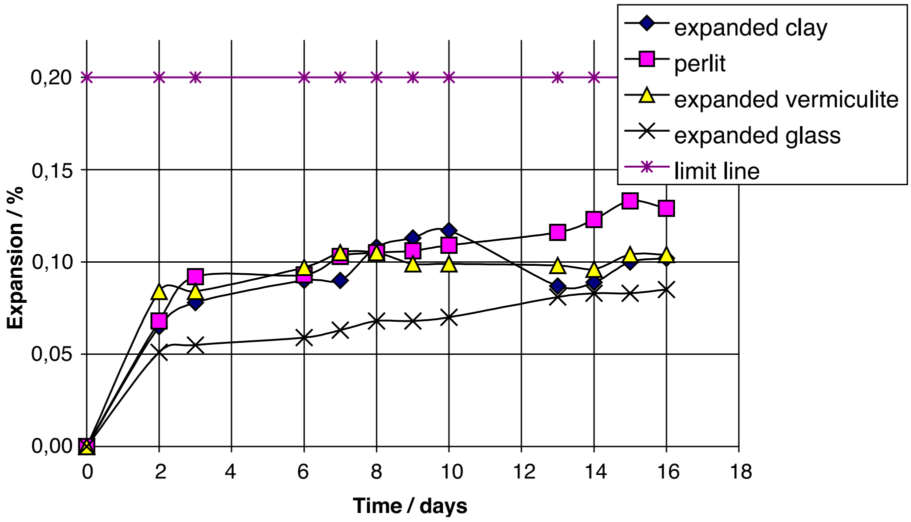

Fig. 3. Average linear expansion of the mortar bar specimens depending upon the number of days of exposure.

From Fig. 2 it can be clearly seen that only the expanded vermiculite had a low Sc value. This value was considerably higher in the case of the expanded clay, and much higher in the case of the expanded glass and perlite, indicating the high sensitivity to alkalis of these two LWAs.

3.2. Expansion of mortar bars, as determined by the ASTM C 1260 method

The results of measurements of linear expansion according to the ASTM C 1260 method are presented in Fig. 3.

After the 3-week exposure period, no cracks, gel spots or wet patches were observed on the surface of the mortar bars. The smallest expansion was observed in the case of the prisms made from expanded glass (0.09%). In the case of the expanded vermiculite and the expanded clay, the measured expansion reached a value of 0.10%, whereas in the case of the perlite, it amounted to 0.13%. None of the specimens exceeded the limit for potentially deleterious expansion, i.e. 0.2% according to ASTM C 1260.

3.3. SEM–EDX analysis of the mortar bars after the accelerated mortar bar test (ASTM C 1260)

The morphology and chemical composition of the reactive products, after the accelerated mortar bar test according to ASTM C 1260 had been performed, were determined on both thin-sectioned and fractured samples of the mortar bars. The back-scattered electrons (BSE) image mode of SEM and the EDX were used.

Fig. 4 is an SEM micrograph of a thin section taken from a mortar bar made using expanded vermiculite, showing the latter embedded in the cement matrix. No reaction can be seen, and the physical bond between the granule and the cement matrix is intimate.

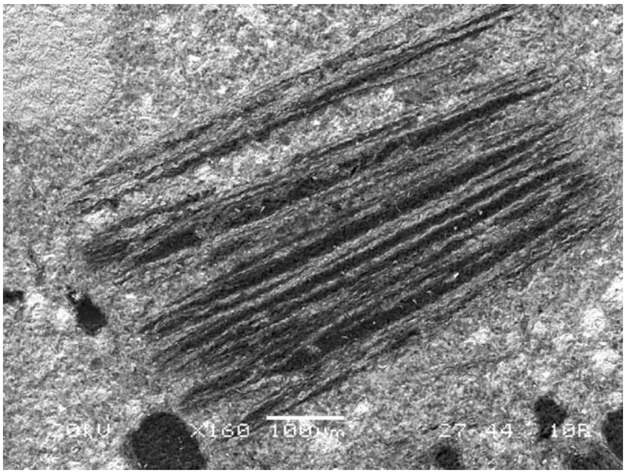

Fig. 4. SEM micrograph of a thin section of a mortar bar with vermiculite.

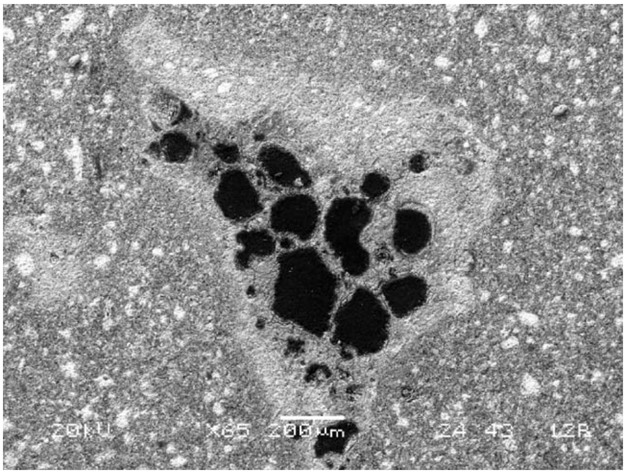

Fig. 5. SEM micrograph of a thin section of a mortar bar with expanded clay.

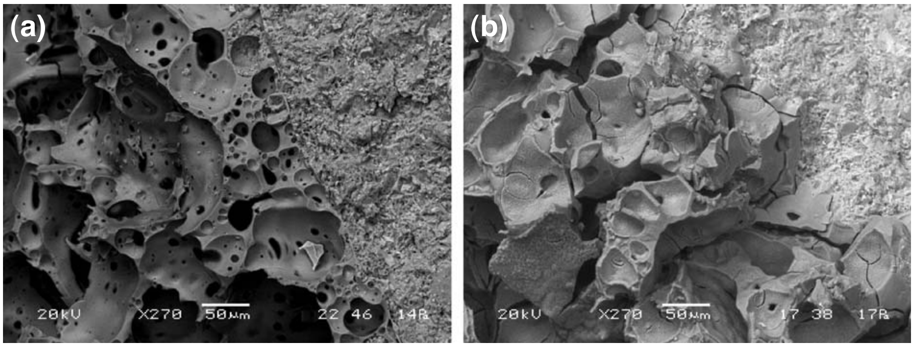

Fig. 6. SEM micrographs of mortar bars with expanded glass (a) not exposed to the alkaline solution and (b) after the accelerated test.

Fig. 5 is a SEM micrograph of a thin section of mortar made using expanded clay. The reaction zone around the granules can be clearly seen, but the contact with the cement matrix is still good, in spite of some fine cracks visible in individual granules, which probably occurred during the preparation of the specimen.

Fig. 6 shows SEM micrographs of the fractured surfaces of two mortar bars made using expanded glass. The fracture shown on the left side, in Fig. 6a, corresponds to an expanded glass granule of the mortar bar which was not exposed to the alkaline solution, whereas the fracture shown on the right side, in Fig. 6b, corresponds to a similar granule, the difference being that, in this case, the mortar bar had undergone the accelerated test. A comparison between the two micrographs shows that, whereas the interior walls of the expanded glass granule belonging to the nonexposed prism are clean and smooth, without any visible reaction products or traces of ASR, the interior walls of the expanded glass granule belonging to the exposed prism are highly deteriorated and fissured.

SEM analysis at a higher magnification confirmed the assumption that destructive reactions had taken place on the interior walls of the granules. Typical reaction products with varying morphology and composition are shown in Fig. 7. Fig. 7a is a SEM image of a thin section of the expanded glass mortar, showing a detail of a granule. In this figure, it can be clearly seen that the expanded glass granule is deeply cracked, and that there are cracks along the boundary between the cement mortar and the granule, some of which spread into the cement matrix. Contact between the granule and the cement matrix has been lost. The texture of the interior surface of the expanded glass granule is no longer smooth, but has changed into a textural gel. The EDX spectrum also indicates a change in the chemical composi-

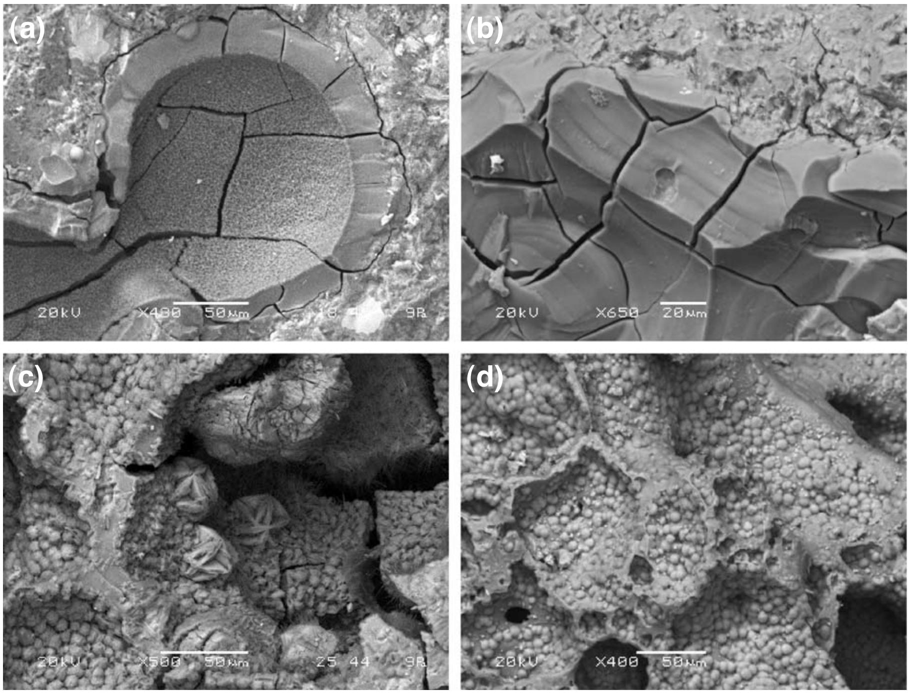

Fig. 7. Typical reaction products found in samples of the mortar bars made using expanded glass: (a) changing of expanded glass into a cracked textural gel, (b) a massive alkali–calcium–silica gel in cracks, (c) crystallized products having a rossete morphology and (d) round agglomerates.

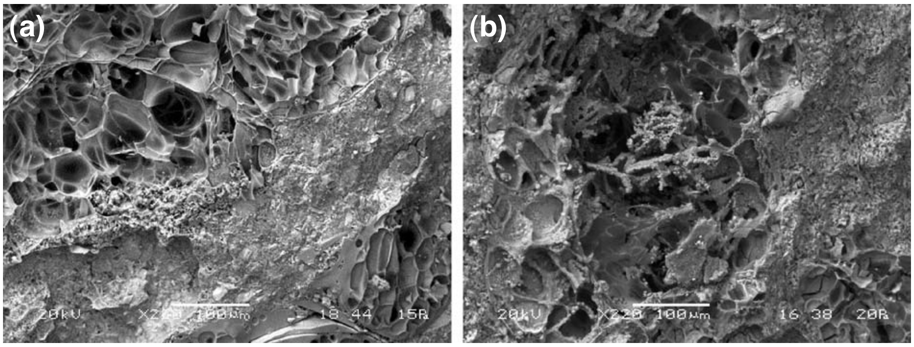

Fig. 8. SEM micrographs of mortar bars with perlite (a) not exposed to the alkaline solution and (b) after the accelerated test.

tion of the expanded glass, for which strong enrichment with calcium at the expense of silicon is typical when compared to the primary material composition. Fig. 7b shows massive alkali–calcium–silica gel, which has been found in the cracks of the mortar bar. Fig. 7c shows crystallized products having a rosette morphology, in which the aluminium content is strongly increased (by a factor of 3 to 4) and the silicon content is reduced (by about a quarter) with regard to the primary composition. Fig. 7d shows round agglomerates, whose calcium content has increased by a factor of up to 3 at the expense of silicon and alkalis.

A comparison of the SEM micrographs of two perlite granules is presented in Fig. 8. Fig. 8a shows parts of two granules from a cement prism which was not exposed to the mortar bar test, whereas Fig. 8b shows part of a granule from a cement prism which had been exposed to the mortar bar test. The perlite granule from the non-exposed prism has clean and smooth internal walls, without any visible reaction products or traces, whereas the internal walls of the perlite from the cement prism which had been exposed to the mortar bar test are partly dissolved, or cracked and partly replaced, and partly deposited with reaction products.

SEM–EDX analysis of the perlite granules, after the latter had been exposed to the mortar bar test, proved the occurrence of several kinds of reaction products, which differed with regard to their morphology and location (Fig. 9). In Fig. 9a, numerous needle-like and a few millimeters long crystals can be seen on the granule's interior walls. Some of these crystals appear to be interlocked with one other, and to be growing in the plane of the interior walls (see marker A), whereas others appear to be growing with their longer axis perpendicular to the perlite

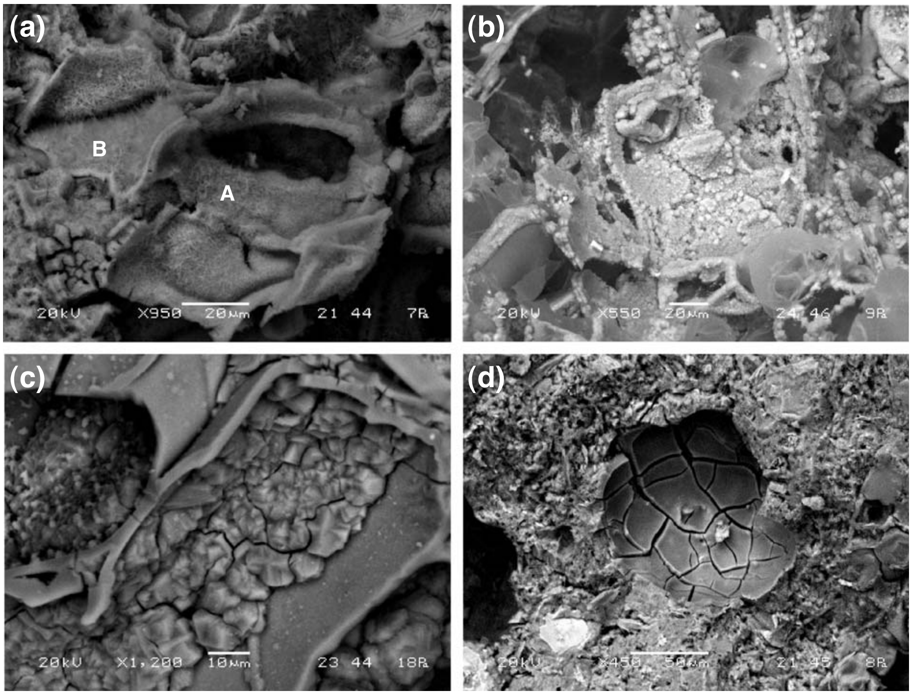

Fig. 9. Typical reaction products found in samples of the mortar bars made using perlite: (a) needle-like crystals appear to be growing in the plane of the interior walls—A or to be growing with their longer axis perpendicular to the perlite wall—B, (b) round agglomerates, (c) crystallized calcium-rich products and (d) a massive alkali–calcium–silica gel.

wall (see marker B). EDX analysis of the chemical composition of these crystals indicated a decrease in the content of silicon (by about 50%), and in the content of aluminium (by about two-thirds), as well as in the content of alkalis, all at the expense of an immensely increased amount of calcium (by a factor of 6 to 7). These needles can be a type of spongy gel, or more probably, products which have developed on the surface of the perlite during the reaction. Unfortunately, no final definition can be made, because the products were very fine and grown out of the primary material, making semiquantitative analysis unreliable.

Fig. 9b shows round agglomerates, up to 10 mm in size, partly in the shape of strips, bands and deposits on the interior walls, and partly as crusts or layers. The results of the EDX analysis indicate a significant reduction in the silicon content and an increase in the calcium and alkali (potassium) content.

Fig. 9c presents reaction products, sparse in quantity and crystallized within the perlite pores. Chemical analysis of their composition showed an increase in calcium content at the expense of silicon and aluminium. Sulphur is also present.

Fig. 9d shows the presence of massive alkali–calcium–silica gel, which has penetrated into an air void. However, the frequency of such occurrences is relatively low.

Similar as in the case of expanded glass, enrichment with calcium and alkalis at the expense of silicon has been found in perlite after the accelerated mortar bar test.

4. Discussion

The results of tests of the ASR of the LWAs according to the rapid chemical method defined in ASTM C 289 showed that the expanded glass and perlite were highly reactive. Although both of these types of glassy aggregate have a similar SiO_2 content, the expanded glass exhibited a much higher reactivity according to ASTM C 289. The most likely reason for this is its alkali content, which is about three times greater than that of the perlite. It may therefore be assumed that this increased alkali content provides an additional inner source of alkali which promotes ASR (although the results of mortar bar tests according to ASTM C 1260 showed that none of the aggregates used caused either expansion or cracks in mortar bars).

The results of SEM–EDX analysis of the LWAs, after the accelerated mortar bar test, confirmed that there was no evidence of reaction between the aggregate and the cement matrix in the case of expanded vermiculite, whereas some evidence of chemical reaction, but not of the ASR type, was found in the case of the expanded clay. However, in the case of the two glassy LWAs (the expanded glass and the perlite), the situation is alarming. A strong reaction has undoubtedly taken place between the aggregate and the alkali, which is manifested in the generation of secondary reaction products, partly in the form of massive calcium–alkali–silica gel and partly in the form of various crystalline products. In general, according to their chemistry, the products appear to be a replica of the primary material, except for the significant enrichment in calcium at the expense of silicon. These products are generated on the original glassy material, and also partly replace it, bringing about the rupture or decay of the texture of the aggregate particles. They certainly could have a deleterious effect on the mechanical properties of concrete made from such LWAs, and might also affect the concrete’s thermoinsulating properties.

The fact that there was no visual manifestation of ASR during the accelerated mortar bar test can be ascribed to the porous texture of the aggregate, which can accommodate the gel and other reaction products. Surprisingly, the expansion of prisms made using expanded glass was comparable to that of prisms using perlite, although we had predicted that, due to its chemical composition and the very high Sc value obtained according to ASTM C 289, the expanded glass would be much more reactive than the perlite. The reason for this probably lies in the difference in the pore structure of the two aggregates. Perlite has a more uniform, fine open capillary system, whereas the pore system of the expanded glass is, by contrast, rougher and a little less connected (a comparison of these two pore systems can be seen in Figs. 6a and 8a). The pore system of the expanded glass can therefore accumulate more gel and other reaction products than that of the perlite.

The results of the investigations of the LWAs showed that, although their mineral composition had a very important, and thus critical, influence on the course and extent of the ASR, other parameters, such as water absorption and specific surface area (as indirect indicators of porosity), could play some less important role in the resistance to ASR of cement composites made from LWAs.

5. Conclusions

The results of the study of the possible danger of ASR in cement composites made using LWAs have shown that the investigated expanded vermiculite and expanded clay did not exhibit any potential ASR. On the other hand, the two glassy aggregates, expanded glass and perlite, showed high reactivity according to the ASTM C 289 test. The results of SEM–EDX analysis of mortar bars made using perlite and expanded glass showed serious decomposition of the aggregate’s texture after the accelerated test, and thus indicated the likelihood of long-term negative effects on cement composites containing these LWAs. In the authors’ opinion, SEM–EDX analysis is, at present, the most suitable for determining the extent and consequences of ASR in the case of such aggregates, although it is not possible to quantify the reaction. The results of the accelerated mortar bar test did not, in this case, show any deleterious expansion, which indicates that the ASTM C 1260 expansion limits may not be appropriate for assessing the risk of ASR in cement composites containing LWAs. For aggregates containing glass, it would be necessary to determine suitable expansion limits on the basis of their known service performance, as has already been similarly proposed for slowly reactive aggregates by Shayan and Morris [10].

A thorough assessment of both the composition of LWA concrete composites and of the environmental factors likely to affect them during their anticipated service life is needed in order to assure the long-term durability and performance of these materials.

Acknowledgements

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