Alkali–silica reactivity of expanded glass granules in structure of lightweight concrete

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Abstract. Main component in the lightweight concrete, which provides its properties, is aggregate. A lot of investigations on alkali silica reaction (ASR) between cement and lightweight aggregates have been done with their results published in the academic literature. Whereas expanded glass granules, which is relatively new product in the market of building materials, has not been a frequent research object. Therefore lightweight granules made from waste glass and eight types of cement with different chemical and mineralogical composition were examined in this research. Expanded glass granules used in this research is commercially available material produced by Penostek. Lightweight concrete mixtures were prepared by using commercial chemical additives to improve workability of concrete. The aim of the study is to identify effect of cement composition to the ASR reaction which occurs between expanded glass granules and binder. Expanded glass granules mechanical and physical properties were determined. In addition, properties of fresh and hardened concrete were determined. The ASR test was processed according to RILEM AAR-2 testing recommendation. Tests with scanning electron microscope and microstructural investigations were performed for expanded glass granules and hardened concrete specimens before and after exposing them in alkali solution.

1. Introduction
Lightweight aggregate concrete (LWAC) is a modern building material with advanced properties such as low density and improved thermal conductivity properties. Expanded glass (porous glass) granules could be used in development of LWAC. Expanded glass is a finely porous (85-90% porosity), clean, light, flame retardant, easily handled, efficient and environmentally friendly building material with excellent properties of thermal insulation. Expanded glass granules contain a glassy phase and may react expansively with cement alkalis, and the possibility of alkali-silica reaction (ASR) must always be taken into account in the production of LWAC [1]. Previous investigations of lightweight aggregate reactivity provided that in some cases lightweight aggregates (perlite, expanded glass, expanded clay) may provoke ASR but with no harmful effects such as expansion and/or efflorescence [2].

The fundamental processes in alkali silica reactions are swelling and/or dissolution of soluble silica and formation of alkali silicates gel by their reaction with calcium ions supplied by cement hydration reactions [3;4]. The chemistry of silica dissolution to form alkali silicate solutions is complex even in the relatively simple industrial processes in common use [5;6].

The accelerated method of aggregate alkaline reactivity testing is usually applied as a fast indicator of the sensitivity of aggregates used in concrete, to test the potential destructive expansion caused by the
alkali-silica reactions. It is based on the papers published by Oberholster and relies on expansion testing of mortar bars immersed in a normal sodium hydroxide solution at the temperature of 80ºC for 14 days [7]. The combination of the alkaline medium and high temperature causes fast, measurable expansion, even in case of low reactive aggregates. In this method the source of alkali is sodium ions, so the standard does not specify the alkali content in the cement used for mortar making.

The goal of this investigation was to evaluate experimentally interaction between expanded glass aggregates and different types of cement. The present paper includes test results of eight LWAC compositions and alkali reactivity assessment of the selected silica aggregates made of waste glass with eight different cement types available at the local market. ASR test was carried out according to RILEM TC 106 testing method. The expansion test results have been compared. The microstructure of LWAC was observed by the scanning microscope before and after the alkali-silica reaction has been conducted.

2. Materials and methods

2.1. Materials

2.1.1. Cement. The materials used in this study were commercially available cements, fillers and admixtures. Eight different cement types were used to determine potential deleterious reactions between expanded glass granules and cement matrix. The chosen cements were available in local markets and produced in six different countries: Latvia, Sweden, Denmark, Estonia, Lithuania and Turkey. Three cement types were produced in Cemex plant in Latvia: CEM I 42.5N, CEM I 42.5R, CEM II A-M (S,L) 42.5 N. Cement CEM II/A-LL 42.5 N was produced at the Lithuanian cement production plant in Akmene. One of the binders, CEM I 42.5N, was produced in Estonia by Kunda-Heidelberg cement, but CEM II/A-LL 42.5R was produced in Sweden. White cements were produced by Danish Aalborg: CEM I 52.5R and Turkish Cimsa – CEM I 52.5N.

The mineral composition and Na equivalent of cements were determined according to LVS EN 196-2:2005 standard and are given in Table 1. As it is seen in Table 1, highest Na equivalent is for cement Kunda – 1.68. Aalborg cement and Cimsa cement provided lowest Na equivalent – 0.13 and 0.31 respectively. Limestone factor (LSF), Silica ratio (SR) and Alumina-iron ratio (AR) are presented in Table 1.

| Cement producer, type | Mineralogical composition, % |
|-----------------------|-----------------------------|
| Cemex CEM I 42.5R    | Naeq 0.96 LSF 105.01 SR 2.52 AR 1.41 C3S 68.22 C2S 1.50 C3A 6.22 C4AF 9.22 |
| Kunda CEM I 42.5N    | 1.68 78.67 2.46 1.82 60.23 6.21 10.20 9.92 |
| Rocket CEM II/A-LL 42.5R | 0.91 111.67 2.65 1.37 76.89 8.90 5.53 8.65 |
| Cemex CEM II        | 1.00 102.11 2.47 1.39 61.88 7.04 6.32 9.66 |
| Cemex CEM I 42.5N   | 0.99 101.32 2.65 1.39 63.23 7.75 6.12 9.28 |
| Akmene CEM II/A-LL 42.5 N | 0.87 110.55 2.19 1.31 74.56 8.90 5.98 10.26 |
| Cimsa CEM I 52.5N   | 0.31 101.86 4.97 20.05 69.77 10.32 10.80 0.64 |
| Aalborg CEM I 52.5R | 0.12 96.24 10.16 5.92 73.74 15.56 4.94 1.07 |

2.1.2. Expanded glass granules. Expanded glass granules are multifunctional lightweight material with improved insulation properties and closed pore structure. Expanded glass granules were produced by Penostek and fraction 0/5 mm was used to design LWAC. Physical and mechanical properties of expanded glass granules were determined and compared to declared by producers. Bulk density, thermal conductivity, compressive strength and water absorption of expanded glass granules were determined. As it is seen in Table 2, declared and obtained properties differ. Thermal conductivity is
higher for granule fraction 0/5 mm, which was declared according to Standard ТУ 5914-001-15068529-2006 and bulk density is from 187-230 kg/m$^3$ compared to declared 100-190 kg/m$^3$.

**Table 2** Mechanical and physical properties of expanded glass granules

| Property                        | Obtained     | Declared     |
|---------------------------------|--------------|--------------|
| Thermal conductivity, W/m$^2$C  | 0.069-0.071  | 0.059-0.061  |
| Compressive strength, MPa       | 0.75-0.80    | 0.45-0.55 MPa|
| Bulk density, kg/m3             | 187-230 kg/m3| 100-190 kg/m3|

Micro structural analysis was done by scanning electron microscope TESCAN MiraLMU Field-Emission-Gun. Pore size of expanded glass granules was determined and granule internal and external wall thickness was measured. Overall view of expanded glass granules with fraction 2/4mm is given in Figure 1.

2.2. **Lightweight concrete mixture composition.** Lightweight concrete with expanded glass granules was designed to obtain new generation lightweight concrete with improved thermal and mechanical properties. Mixture design consisted of commercially available materials and mixture was created to provide optimal relation between fresh concrete properties and hardened concrete properties. Chemical admixtures were used to control concrete setting time, workability and to avoid water bleeding. The mixing procedure of concrete was as follows: all dry materials were mixed with hand mixer till a homogenous mixture formed (approximately for 1.5 min). Then water was added in two steps. During the first step approximately 70% of water was added and batch was mixed for 1.5 minutes. During the second step the rest of the water was added. Total mixing time was approximately 5 minutes. Mixture with the cone slump of class S5 has been obtained.

2.3. **Testing methods of concrete**

Fresh and hardened concrete properties were obtained for all eight concrete mixtures with different types of cement. Fresh concrete properties like air content (LVS EN 1015-7), cone slump (according to LVS EN 12350-2), consistence of fresh mortar (by flow table) according to LVS EN 1015-3 and fresh concrete density (LVS EN 12350-6) were determined. Mechanical, physical and microstructural investigations were carried out for specimens at the age of 7 and 28 days. Flexural and compressive strength were obtained according to LVS EN 1015-11. Remoulded samples were kept in an ambient temperature until testing day. 28 days old samples were investigated by EDX. Alkali-aggregate reaction test was performed according to RILEM AAR-2 (RILEM TC 106-AAR) for all eight concrete samples with different cement types and SEM investigations were performed.
3. Results and discussions

3.1. Fresh concrete properties

Cone slump test and consistence of fresh mortar by flow table method were performed for all eight concrete mixtures. Several cement types reacted differently with chemical admixtures and fresh concrete fluidity varied and led to improved or worsened concrete casting properties which were tested by cone slump (Abrams cone) and with flow table. As it is seen in Table 8, flow table results vary from 167-185 mm depending from chosen cement type. Cone slump results indicated similar results and cone slump was in range from 22-25 cm, which corresponds to cone slump class S5 (≥22 cm).

Density of fresh concrete was in the range from 734 to 781 kg/m³ and air content was in the range from 7.0 to 8.7%. Results of fresh concrete properties is given in table 4.

Table 4. Fresh concrete properties of expanded glass lightweight concrete with different types of cements.

| Cement type used in LC | Flow table, mm | Cone slump, cm | Fresh concrete density, kg/m³ | Air content, % |
|------------------------|----------------|----------------|------------------------------|---------------|
| 1 Cemex CEM I 42.5R    | 180            | 23.5           | 763                          | 7.5           |
| 2 Cemex CEM I 42.5N    | 179            | 24             | 756                          | 7.1           |
| 3 Cemex CEM II A-M (S-L) 42.5N | 175          | 23.5           | 766                          | 8.1           |
| 4 Aalborg white CEM I 52.5R | 181          | 25             | 768                          | 7.0           |
| 5 Kunda CEM I 42.5N    | 185            | 24.5           | 755                          | 7.7           |
| 6 Cimsa white CEM I 52.5N | 167          | 22             | 781                          | 7.9           |
| 7 Akmene CEM II/A-LL 42.5N | 175          | 23             | 734                          | 8.7           |
| 8 Rocket M-500 CEM II/A-LL 42.5R | 168         | 23             | 758                          | 8.1           |

3.2. Hardened concrete properties

Density, flexural and compressive strength of LWAC are given in Table 9. Density of LWAC was in the range from 490 to 555 kg/m³, flexural strength from 1.0 to 1.4 MPa for 7 days old samples and 1.0 to 1.5 MPa for 28 days old samples. Compressive strength of lightweight concrete was in the range from 2.7 to 4.3 MPa after 7 days hardening. Concrete with cement Akmene provided lowest compressive strength as previously. Highest compressive strength was for concrete with Cemex CEM I 42.5R. Compressive strength of 28 days old samples was in the range from 2.8-4.0 MPa. Strength reduction was observed for concrete with Cemex CEM I 42.5R, Kunda CEM I 42.5N and Rocket M-500 cements.

Table 9. Fresh concrete properties of expanded glass lightweight concrete with different types of cements

| Cement type used in LC | Density, kg/m³ | f<sub>flex</sub>, 7d, MPa | f<sub>flex</sub>, 28d, MPa | f<sub>comp</sub>, 7d, MPa | f<sub>comp</sub>, 28d, MPa |
|------------------------|----------------|----------------|----------------|----------------|----------------|
| 1 Cemex CEM I 42.5R    | 545            | 1.4            | 1.5            | 4.3            | 4.0            |
| 2 Cemex CEM I 42.5N    | 490            | 1.1            | 1.1            | 3.1            | 3.4            |
| 3 Cemex CEM II A-M (S-L) 42.5N | 530          | 1.3            | 1.4            | 3.8            | 3.8            |
| 4 Aalborg white CEM I 52.5R | 520          | 1.1            | 1.1            | 3.3            | 3.6            |
| 5 Kunda CEM I 42.5N    | 555            | 1.1            | 1.4            | 4.0            | 3.8            |
| 6 Cimsa white CEM I 52.5N | 520          | 1.1            | 1.2            | 3.7            | 3.7            |
| 7 Akmene CEM II/A-LL 42.5N | 500          | 1.0            | 1.0            | 2.7            | 2.8            |
| 8 Rocket M-500 CEM II/A-LL 42.5R | 520          | 1.3            | 1.3            | 3.7            | 3.6            |
3.3. Microstructural analysis with SEM
SEM analysis was performed for lightweight concrete specimens after 28 days hardening period in room conditions. Figure 2 a demonstrate microstructures of lightweight concrete specimens made from Rocket M-500 CEM II/A-LL 42.5R cement. Cement matrix between expanded glass granules is uncompact and rich with pores. Hydrated cement paste (HCP) is rich with ettringite needles, which are present in the whole volume of HCP. It is well known that decrease of the pH-value by leaching of alkalis or by other pH-reducing reactions such as ASR promotes a recrystallization of ettringite in the hardened concrete. All specimens investigated in this research have a great potential expansion space for the formation of new phases due to porous cement matrix as shown in figure 2a.

![Figure 2. Lightweight concrete microstructure analysis with SEM. Normal sample (2a) and after ASR test (2b). Sample with cement Rocket M-500 CEM II/A-LL 42.5R.](image)

3.4. Alkali-Silica reactivity
ASR was tested according to RILEM TC 106-2 - Detection of potential alkali-reactivity of aggregates -the ultra-accelerated mortar bar test. ASR tests results are given in Figure 3. Final relative expansion was in range from 0.160 to 0.205%. The lightweight concrete with Aalborg cement showed the lowest expansion – 0.160%, it was followed by Cimsa cement – 0.185%. Cemex CEM I 42.5N provided relative expansion 0.195%, Cemex CEM II, Rocket M-500 and Kunda cement provided relative expansion 0.200%, but the highest relative expansion was for lightweight concrete with Akmene cement and Cemex CEM I 42,5R – 0.205%.

![Figure 3. ASR test results. Relative expansion of mortar bars](image)
3.5. **SEM observations after ASR**

According to microphotographs, expanded glass granules were seriously damaged after ASR test in all specimens no matter what the type of cement was used. As it is seen in Figure 2b, the specimens after exposure to the accelerated alkali-aggregate reaction conditions showed a slightly different picture. ASR gel with thickness up to 10µm appears between the aggregate and paste and on pore walls of expanded glass granule. 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. Ettringite was not found in the microstructure of specimens.

4. **Conclusions**

By integrating expanded glass granules into the structure of cement paste the lightweight concrete could be produced. During mixing procedure expanded glass granules remain uncrushed and constantly fill the volume of lightweight concrete due to the incorporation of chemical additives in the lightweight concrete mixture composition.

During lightweight concrete aging mechanical properties increased up to 7th day of hardening. Then it stops and only negligible strength increase was observed at the age of 28 days. By observing lightweight concrete samples with SEM after 28 days curing period corrosion marks could be detected. Corrosion occurred between expanded glass granule and hydrated cement paste.

Relative expansion for prismatic samples was in range from 0.160-0.205%. Lowest expansion provided Aalborg white cement, but even this result did not reach limits of applied standard which is 0.05%. SEM observations of samples after expansion test revealed destroyed structure of expanded glass granule and ASR products were detected on the contact layer between expanded glass granule and cement paste. Cements with lower Na equivalent, higher silica ratio (SR) and higher C2S provided lower expansion value. Advantages of limiting expansion was observed for white cements.

Since corrosion resistance of obtained lightweight concrete did not reach standard limitation, further research must be provided. By choosing cement, which provides less expansions (reduced Na equivalent and increased silica ratio (SR)), and adding puzzolanic materials to lightweight concrete mixture design, potential reactions could be reduced.

5. **Acknowledgement**

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6. **References**

Examples taken from published papers:

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