



# Mechanism of preventing the alkali–aggregate reaction in alkali activated cement concretes



Pavel Krivenko<sup>a,\*</sup>, Rostislav Drochytka<sup>b</sup>, Aleksandr Gelevera<sup>a</sup>, Elena Kavalerova<sup>a</sup>

<sup>a</sup> Kiev National University of Civil Engineering and Architecture, Scientific Research Institute for Binders and Materials, Kiev, Ukraine

<sup>b</sup> Brno University of Technology, Institute of Technology of Building Materials and Components, Brno, Czech Republic

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## ABSTRACT

Processes of structure formation taking place in the interfacial transition zone “cement paste – aggregate” have been studied on a variety of cement model systems. The results of this study suggest that, depending upon the contents of components capable of actively interacting with alkalis in the presence of reactive SiO<sub>2</sub> in the cement and aggregate, the processes taking place during an alkali–aggregate reaction could be constructive or destructive in character. So-called “constructive processes” are attributed to binding the corrosion reaction products with the formation of the alkaline aluminosilicate hydrates. The results of this study have been taken as a base in developing a mechanism of preventing the alkali–aggregate reaction in the alkali activated cement concretes through the introduction of additional quantities of Al<sub>2</sub>O<sub>3</sub>-containing substances.

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## 1. Introduction

First observations of reactivity of the alkalis contained in some mineral constituents go back to 1916 when a geologist E.A. Stephenson has reported about a reaction between feldspar and sodium carbonate which resulted in the formation of a gel. Degradation of concrete as a result of this reaction between the alkalis contained in cement and some ground rocks was first observed in the USA. In 1922 similar deteriorations took place in the New River Hydropower Station (Virginia, USA) 10 years after it had been erected. In 1940 Stanton [1] has reported about an “alkaline reaction” (alkali–aggregate reaction, AAR) due to opal-containing fractions of the rocks used for the diverting dam in California. These deteriorations have initiated a number of extensive studies to be held in the USA to reveal causes for taking appropriate measures.

In 1947 a reaction between the alkali and silicic acid (alkali–silica reaction, ASR) was described by Bogue [2]. A few years later, in

1952 this reaction was described by Kuehl in his book, in which he made references to the data obtained in the USA [3].

Starting from 1950s this reaction is also known in Australia and since the mid of 1950s more and more countries (Canada, Iceland, South Africa and other countries) report about deteriorations of bridges, sleepers, dams, roads due to the ASR. Very often these deteriorations are attributed to other causes such as low frost resistance or other aggressive exposure. The losses were huge, which is why the problem became urgent and required prompt actions to be taken.

As a result of extensive studies and experimental works [3–6], basic fundamentals of the ASR mechanism have been formulated. These are:

- sources of alkalis are cement, concrete admixtures, outdoor aggressive environment;
- limit values of the alkalis (Na<sub>2</sub>O + 0.658 K<sub>2</sub>O) contained in portland cement (expressed in terms of Na<sub>2</sub>O-equivalent content) should be less than 0.6% by mass;
- expansion due to the ASR is associated with osmotic pressure from the aluminosilicate gel- a product of reaction between the active silica and alkali. This gel acts as a

\* Corresponding author.

E-mail address: [pavlo.krivenko@gmail.com](mailto:pavlo.krivenko@gmail.com) (P. Krivenko).

semi-permeable membrane, through which the  $\text{OH}^-$ ,  $\text{K}^+$ ,  $\text{Na}^+$ -ions can move in the direction towards the aggregate surface. The alkali metal silicates start to fill a surrounding pore space resulting in the expansion pressure;

- a free  $\text{Ca}(\text{OH})_2$  of the cement paste plays an important role. The alkali metal silicate gel itself is not able to produce great expansion pressure. However, the presence of calcium creates conditions promoting synthesis of a dense gel-like calcium silicate hydrate (CSH), which is present in the outer zone of reaction and acts as a semi-permeable membrane allowing an access of new portions of water, the  $\text{Na}^+$  and  $\text{OH}^-$ -ions to a place of reaction, but restricting a removal of the reaction products from the interfacial transition zone (ITZ), thus increasing pressure in the ITZ “cement paste – alkali-susceptible aggregate”. This pressure may achieve 20 MPa [7] and the attained strength of the concrete in tension may reach only 3...5 MPa, thus resulting in microcracks and progressive degradation of concrete. Besides, the alkali which is released in the end of this reaction takes part in deterioration of the deeper layers of the alkali-susceptible aggregate grains. That is why, finally, an available content of the  $\text{Ca}(\text{OH})_2$  can serve to some extent as a restricting factor not allowing this reaction to damp. This can explain the reasons for more severe destructive effect in case of excessive quantities of the  $\text{Na}^+$ - and  $\text{Ca}^{2+}$ -ions compared to solely  $\text{Na}^+$ -ions;
- reduction of risk due to expansion is reached by introduction of active mineral additives of sedimentary and igneous origin (gaizes, zeolites, perlite, tuff, pumice, etc.) and those of man-made origin (fuel ashes, metallurgical slags, silica fume, etc.) in order to bind excessive quantities of calcium.

This problem becomes more and more serious since contents of alkalis in contemporary cements and concretes tend to increase due to the wider use of the cements with the higher quantities of sodium and potassium oxides as a result of high degree of cement kiln exhaust gas cleaning and return of the cement kiln dust back into the process. As a result, contents of alkalis in cements (in  $\text{Na}_2\text{O}_{\text{eq}}$ ) tend to increase up to 1...2% by mass (against  $\leq 0.6\%$  by mass permitted under the standards). Also, of wider and wider use is a class of alkali activated cementitious materials (alkali activated slag cement, alkali activated portland cement, alkali activated slag portland cement, alkali activated pozzolana cement, geocement-based adhesives, etc.), the contents of alkalis in which (in  $\text{Na}_2\text{O}_{\text{eq}}$ ) vary between 1.5% and 5% by mass [8–10]. And, at last, a quantity of alkalis in concrete tends to increase as a result of the wider use of chemical, mineral and organo-mineral additives containing alkalis.

For a long time the above views on the mechanism of ASR have been taken as a base for developing recommendations on how to prevent dangerous consequences of this reaction and to apply restrictions on a wider use of alkali activated cements which contain alkalis in much greater quantities, as is shown earlier, compared to those of traditional cements [11,12].

At the same time, some researchers have reported on the results of attempts to explain the processes of corrosion of alkali-susceptible aggregates in the presence of alkalis not only from the point of view of quantitative content of alkalis and free  $\text{Ca}(\text{OH})_2$ . So, Malek and Roy [13] studied a role played by  $\text{Al}_2\text{O}_3$  which is released from the feldspathoid stone through dissolution and found that with increase in the  $\text{Al}_2\text{O}_3$  to  $\text{SiO}_2$  ratio the ASR changes its character from a destructive for the constructive one. These findings coincide well with the results of the studies reported by Tang and Han [14]. Basing on them, Malek and Roy [15] draw a conclusion that elimination of hazardous effect of alkalis when a glassy slag was added to portland cement could be attributed not only to the lower permeability of slag pastes, but also to an ability of the slag to bind

the alkali metal ions with the formation of insoluble alkaline aluminosilicate hydrates of the  $\text{Na}_2\text{O}\cdot\text{Al}_2\text{O}_3\cdot 5\text{SiO}_2\cdot 5.4\text{H}_2\text{O}$  type. Further, the results of works reported in [16–19] showed that possible formation of the alkaline or mixed alkaline–alkaline earth aluminosilicate hydrates in the ITZ depended not only upon an aggregate type, but upon a composition of the aluminosilicate constituent of the alkali activated cement. A conclusion was made that by changing the  $\text{Al}_2\text{O}_3$ -content of the cement aluminosilicate constituent by introduction of the active  $\text{Al}_2\text{O}_3$ -containing additives the risk of destructive consequences of the ASR in the concretes made from the cements with high content of alkali could be brought to minimum.

The paper covers the results of study on behavior of aggregates from the alkali-susceptible rocks in the environment of various cement pastes, including the alkali activated cement pastes, and discusses ways of how to eliminate destructive processes in the concretes made from the cements with the increased alkali contents.

Moreover, a necessity of such studies was initiated by the increasing use of the alkali activated cements and concretes both in Ukraine and over the world.

## 2. Materials and examination techniques

Finely crushed dense rocks of polyfractional composition with the particle size between 0.5 and 10 mm were used in the study as potentially alkali-susceptible aggregates. Chemical composition of the selected aggregates is given in Table 1. A river quartz sand with a grading factor equal to 2 was used as fine aggregate in preparation of the reference specimens.

The following cements were used to study a behavior of the alkali-susceptible aggregates in the alkali activated cement concretes:

- portland cement Type IIII II/A-III with compressive strength = 400 kgf/cm<sup>2</sup> and 0.22%  $\text{Na}_2\text{O}_{\text{eq}}$  by mass (according to national standard of Ukraine);
- portland cement Type IIII II/A-III with compressive strength = 400 kgf/cm<sup>2</sup> and 1.3%  $\text{Na}_2\text{O}_{\text{eq}}$  by mass;
- alkali activated slag cement Type IIII IEM I (2.5%  $\text{Na}_2\text{O}_{\text{eq}}$  by mass);
- alkali activated slag portland cement Type IIII IEM IV (60% slag by mass) (2.5%  $\text{Na}_2\text{O}_{\text{eq}}$  by mass)
- alkali activated pozzolanic cement Type IIII IEM III-3 (60% fly ash by mass) (2.5%  $\text{Na}_2\text{O}_{\text{eq}}$  by mass) [20].

Fineness of the cements under study expressed as a Blaine specific surface was 4400...4600 cm<sup>2</sup>/g.

Chemical composition of the major cement constituents and metakaolin additive are given in Table 2.

Sodium carbonate and sodium metasilicate pentahydrate were used in the study as  $\text{Na}_2\text{O}_{\text{eq}}$ . They were introduced into the cement composition in dry form in the process of intergrinding and blending the cement constituents or in liquid form (aqueous solutions). Metakaolin was used as active mineral additive.

Water was used in preparation of the compositions in which the alkaline activator was introduced in dry form ( $W/C = 0.35\text{--}0.4$ ).

Beam specimens (4 × 4 × 16 cm) were prepared for testing strength in compression and in flexure and prism specimens (2.5 × 2.5 × 28.5 cm) with copper bench marks – for testing deformations. The concrete mixture used in the experiments contained cement and aggregate taken as 1:2.25.

After 2 days the specimens were taken from the moulds and placed for further hardening in the corresponding conditions. A basic measurement was taken preliminary on the specimens intended for control of deformations (reference specimens). A device with indicating gage (division value = 0.01 mm) was used for taking measurements.

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