



Variations in the composition and relations between alkali-silica gels and calcium silicate hydrates in highway concrete

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ABSTRACT

Alkali-aggregate reactions (AAR) are a form of severe concrete deterioration that can be found anywhere in the world. The rock type of aggregate is the crucial factor for AAR occurrence as well as for the chemical and morphological nature of their deleterious products – alkali-silica gels (ASG). The paper focuses on highway cement concrete pavement with aggregate composed of metamorphosed volcano-sedimentary complex rocks, such as proterozoic chert, black shale, spilite, and green schist, and alluvial sandy gravels. These petrographic types of aggregate were found to be potentially hazardous. The microchemical composition of ASG was analysed and compared to calcium silicate hydrates present in the vicinity of one another. The qualitative elemental composition of both is the same. Gradual variations of ASG within a single position and among different positions in cement concrete pavement slabs were monitored and compared.

1. Introduction

Alkali–aggregate reactions (AAR) can be defined as chemical reactions between alkali hydroxides (sodium and potassium) present in the pore solution of concrete and certain minerals in the aggregate. The product is typically a hygroscopic gel (alkali silica gel, ASG) that expands on hydration and may introduce cracking to the surrounding hardened concrete, thereby causing its mechanical properties to deteriorate.

Geological approaches which combine methods such as polarizing light microscopy [1, 2], electron microscopy and microanalysis, powder X-ray diffraction, differential thermal analysis or calorimetry and thermogravimetry, and infrared spectroscopy [3] are applied in the investigation of expansion reactions in concrete. Though originally auxiliary methods, they have become crucial for the prediction of the expansion processes.

A combination of geological methods makes it possible to analyse of the complex processes taking place in the “stones”. In addition, it can reveal, clarify and gradually specify the dependences and sequences of degradation processes taking place between the binder and aggregate in cement concrete pavement (CCP) slabs. However, their deciphering is made the more difficult by the fact they can be caused by a number of non-linear factors, whether internal or external.

Available literature on AAR is vast. Since 1943, when AAR were first described, the number of publications on this topic has been increasing each year.

1.1. Characteristic Manifestations of AAR

Manifestations on macroscopic scale:

- 1) A net of fine micro-cracks can be observed on the surface of concrete. They become more easily visible after rain.
- 2) The micro-cracks gradually open and form a web that spreads across the surface.
- 3) Deep cracks often produce a thick net of polygonal or radial failures. Joint action of additional negative factors increases the depth of the cracks and the size of the affected area.
- 4) Subsequently, aggregate clasts become loose and fall off and the binder suffers spalling. The surface becomes crater-like and, later, small depressions, of up to several centimetres large, appear.

Manifestations on microscopic scale:

- 5) Gel covers the rims of air voids.
- 6) Filling of air voids and the first formation of expansion micro-cracks.
- 7) Gradual filling of the cracks with gels.
- 8) Permeation of gels through weakened zones of aggregate; formation of a breccia-like texture.
- 9) Gel forms “mound- and crater-like” formations.
- 10) Due to the decrease in moisture, the plasticity of gels is reduced.

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Contraction cracks often filled with second-generation gels begin to appear.

Development of ASG chemical composition:

- 1) The C/S (CaO/SiO_2) ratio increases as the gel ages.
- 2) H_2O content is variable.
- 3) Increase of Alk/C ($(\text{Na}_2\text{O} + \text{K}_2\text{O})/\text{CaO}$) ratio during crystallization processes.
- 4) Low to trace contents of Al_2O_3 and its absence in crystalline phases developed from gels.
- 5) Sulphates such as ettringite, thaumasite or a solid solution of these end members occur.

Unless the basic conditions are met, i.e. the presence or reactive SiO_2 forms, highly alkaline environment, favourable temperature and moisture, the reactions leading to the formation of AAG do not proceed.

The occurrence and course of AAR is mostly affected by the composition of aggregate, binder, pore solutions and the climate. A successful prediction of whether AAR will occur requires knowledge of the conditions and course of AAG formation.

Calcium silicate hydrates (CSHs) form in highly alkaline environments ($\text{pH} > 12$). These phases can be stable only in an equilibrium concentration with the $\text{Ca}(\text{OH})_2$ solution. Hydroxyl ions in the pore solutions react with reactive aggregate. If CSHs contain higher amounts of sodium and/or potassium (usually expressed as $\text{Na}_2\text{O}_{\text{ekv}}$) the content of $(\text{OH})^-$ ions in the pore solution increases and the pH value rises. The higher the pH value is, the faster and more intense the reactions with silica are. These reactions result in the formation of alkali-silica gels (ASG). The limit content of alkalis in cement is considered to be 0.6% of $\text{Na}_2\text{O}_{\text{ekv}}$ [4]. This value is often used for the assessment of alkali-aggregate non-reactivity. Tuthill [5] recommended decreasing the limit $\text{Na}_2\text{O}_{\text{ekv}}$ value to 0.4%.

Alkali-silica gels hydrate and increase their volume (they swell). If ASG completely fills intergranulars and/or air voids, the resulting expansion pressure begins to affect the concrete. Within a relatively short period, the expansion pressure deforms and damages the microstructure of both binder and aggregate in sensitive locations. The first microcracks appear in the vicinity of aggregate grains and air voids in places where reaction rims appear. A publication by Binal [6] presents the results of expansion pressure measurements in samples with various reactive aggregate rocks. The highest ASG pressure was identified in a mixture containing opal aggregate (2.81 MPa), while the lowest was measured in samples with orthoquartzite rocks (0.26 MPa).

AAR do not need to start immediately after concrete has hardened; in fact there may be a delay of several years. Because the hardened cement paste is at risk of being infiltrated by alkalis from the aggregate or the outside environment, the rate of AAR may later gain in intensity. However, in some cases, AAR do not occur in concretes with aggregate considered as highly reactive. The most probable explanation as to why some of the potentially reactive rocks do not cause AAR is due to a lack of alkalis.

The focus of the paper is to examine mutual relations of alkali-silica gels, as the main manifestation of AAR, and calcium silicate hydrates. Another aim is to study the reactivity of volcano-sedimentary rocks such as green schists and spilites. There were several publications that focused on the same issue as this paper. A handbook for the identification of alkali silica reactivity in CCB was published by Stark [7]. Modrý et al., Lukschová and Šachlová et al. [8–11] and others focused on the procedures of ASR and AAG identification including associated macroscopic and microscopic features. Gregerová and Všíanský [12] summarized the applicability of microscopic examination methods for the identification of deterioration products in concrete. Pertold et al. and Pertold and Lukschová [13–15] focused on the state of CCB in Czech Republic.



Fig. 1. Macroscopic appearance of the examined concrete.

2. Material and Methods

The subject of the study are concrete cover blocks (CCB) with the dimensions of ca 6×3.7 m, which were placed below the surface of a highway and exposed to the traffic for 18 years. Drill cores (15×30 cm) and edge fragments of the CCB were examined. Samples were systematically extracted from the drill cores with steps of 10 cm. Marked polygonal cracks were found in the surface. Macroscopically, the parts that showed the most damage were rim areas around majority of the aggregates (Fig. 1), however the gels can penetrate rock fragment as well.

Since the extraction of the samples, temperature and humidity were kept constant until the time of the analysis. This was because temperature and humidity fluctuations cause drying up and swelling of gels and with this connected leaching of salts.

A set of polished thin sections was prepared from each drill core and fragment sample. The samples were cut using water cooling. Subsequently, the cut plates were immediately placed into a vacuum chamber. Thin sections were prepared in a dry way and cleaned with isopropyl alcohol. The thin sections were examined by means of optical and electron microscopy and X-ray microanalysis. The petrographic analysis of fine aggregate was done by polarizing microscopy using an Olympus BX 51 microscope. An estimation of the contents of coarse aggregate rock types was made by image analysis of pictures of longitudinal sections of the drill cores. Scanning electron microscopy and energy dispersive spectroscopy (SEM/EDS) using a Tescan Vega LSU was the principal method of examination of ASG and CSH phases. EDS was used for point and line analyses. The area distribution of selected elements was determined as well.

Given the fact that the EDS analysis does not allow the detection of water, the water content was calculated as the difference between the measured sum of oxides and 100%. The same method was used e.g. by Poole and Bérubé and Fournier [16, 17].

3. Results

3.1. Petrographic Composition of Aggregate

The petrographic composition of aggregate is very diverse (Tables 1 and 2).

3.1.1. Coarse Aggregate (Fraction of 4–32 mm)

The aggregate consists of green schists, spilites and black shales. The macroscopic appearance of the coarse aggregate is shown in Fig. 2. The reactive potential of this fraction is lower than reactivity of the fine one due to its granulometry and the absence of reactive SiO_2 forms.

Greenschists are grey-green, very fine-grained rocks. A plane-parallel texture is visible on a macroscopic scale while microstructure is nemato-, lepido-, up to fibroblastic. Locally, the fragments are massive.

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