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The use of lithium compounds for inhibiting alkali-aggregate reaction effects in pavement structures

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Abstract. Internal corrosion of concrete caused by the reaction of reactive aggregate with sodium and potassium hydroxides from cement is a threat to the durability of concrete pavements. Traditional methods for reducing the negative effects of the reaction include the use of unreactive aggregates, low alkali cements, mineral additives or chemical admixtures, incorporated during mixing. Lowering the relative humidity of the concrete below 80% is another measure for limiting the destructive reaction.
The incorporation of lithium compounds, in particular lithium nitrate and lithium hydroxide, to the concrete mix is a method of limiting alkali-silica reaction effects. The challenge is to reduce the negative effects of aggregate reactivity in members in which the reaction has occurred because the aggregate happened to be reactive. The paper presents ways of limiting the deterioration of ASR-affected concrete in road pavements and other forms of transportation infrastructure, mainly through the use of lithium compounds, i.e. lithium nitrate. Impregnation methods that allow the penetration of lithium ions into the concrete structure were characterized, as was the effectiveness of the solutions applied.

1. Introduction

Alkali-aggregate reaction (AAR) in concrete is an example of internal corrosion that occurs in the entire volume of the material. Two currently recognized types of AAR are reactions between alkalis and aggregates containing reactive silica (ASR) and reactions between alkalis and carbonate aggregates (ACR). Due to more frequent presence of reactive forms of silica such as opal, tridymite, chalcedony, and strained quartz, in aggregates used in concrete, ASR is usually the main process responsible for the deterioration of concrete structures [1]. The reaction between aggregates and sodium and potassium hydroxides, which originate mainly from the cement, produces a sodium potassium-calcium silicate gels, which absorbs water and swells. Alkali-aggregate causes expansion and cracking in the concrete.

Experience in dealing with the devastating ASR has been reported in many countries, including United States, Canada, England, Denmark, Germany, India, China, Japan, or Korea [2-4]. In some cases the destruction of concrete was incorrectly attributed to other deterioration processes, excluding the reaction with alkalis [4]. In Poland, no cases of destructive alkaline reaction have been reported so far, however, the presence of reactive aggregates has been found in the regions of north-eastern and south-west Poland [5-7]. It is further necessary to take into account that concretes in road structures are highly exposed to AAR, which results from the limited possibility of protecting them from moisture. Also, the use of de-icing agents (sodium chloride) as an external source of alkalis increases

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their concentration in the pore solution of the concrete. In the ASR risk classification system introduced by the British Cement Association UK, concrete pavements are classified in the high-risk category [8].

The ASR preventive measures focus on eliminating the factors contributing to the destructive processes by using unreactive aggregates, low alkali cements, or protection against moisture. Cement with mineral additives may also be used, where it is possible to increase the permissible content of alkali in cement [9]. The methods of preventing the effects of ASR include the use of lithium compounds, mainly lithium nitrate and lithium hydroxide [10-12]. The risk of alkali attack is only possible here at the mixing stage of concrete production.

In ASR-affected elements, further protection of the material against damage can be provided by adding lithium compounds and reducing the relative humidity of the concrete. The present study aims at outlining the ASR preventive methods for use on concrete, especially on concrete pavements.

2. Alkali-silica reaction in concrete pavements

Cracking of road surfaces due to AAR starts in sites exposed to high moisture. Cracks are observed near free edges and joints as parallel to the free longitudinal edges of the road and perpendicular to the transverse joints. A model of crack development due to AAR is shown in Fig. 1:

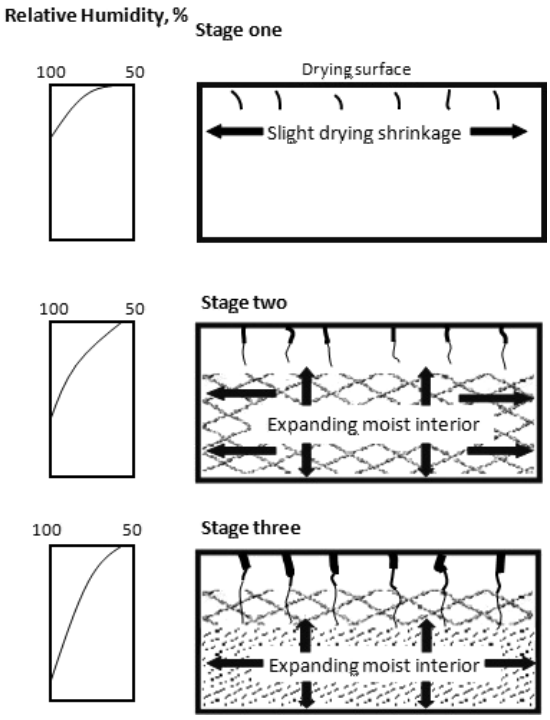


Figure 1. Model of AAR crack extension in concrete pavement [13].

The development of cracks in concrete pavements can be represented in the form of a three-stage model, depending on the relative humidity of the concrete. In the first stage, very small cracks are observed; these are caused by drying shrinkage. The effects of the alkaline reaction are not visible at this stage. In the second stage, at a relative humidity above 80%, a swellable silicate gel forms and migrates towards the surface of the concrete. Surface cracks expand as a result of concrete expansion. In the third stage, when the top layer dries, the reaction in the surface layer is reduced. A high humidity level inside the concrete ensures the progress of deterioration. The gel that exerts tensile stress on the surface of the concrete causes the growth and widening of the surface cracks. In zones with high moisture, the reaction continues until depletion or reduction in the amount of one of the substrates. The presence of reactive silica and the right amount of alkalis in the pore solution is

insufficient when the concrete dries. The process is then suppressed until the moisture content in the concrete increases.

Concrete road surfaces are exposed primarily to the deterioration caused by cyclic freezing and thawing. ASR cracks affect the durability of concrete exposed to negative temperatures. The absorption of water by alkali silicate gels increases its quantity in the element, thus lowering the freeze-thaw resistance of the concrete [14]. De-icing agents used in winter, such as NaCl, increase alkali content in concrete, which results in the acceleration of their reaction with aggregate. Sodium chloride increases the pH of the pore solution, and the solubility of silica and calcium hydroxide also increases, contributing to a faster progress of the AAR and the formation of Na+K-S-H and C-N + K-S-H gels [13]. Rangaraju et al. stated that deicers such as sodium acetate or potassium acetate can trigger ASR [15].

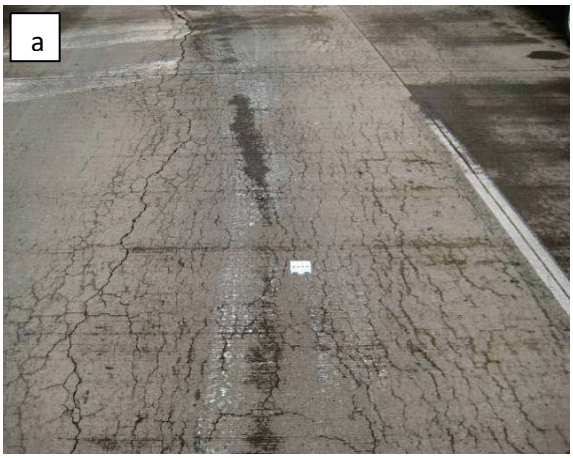
3. ASR symptoms

The types of distress in non-reinforced and reinforced concrete pavements are summarized in Table 1.

Table 1. Distress in concrete pavements [16].

Jointed concrete pavements	Continuoustly reinforced concrete pevements
Corner break	Longitudinal cracking
D-cracking	Transverse cracking
Longitudinal cracking	D-cracking
Transverse cracking	Map cracking
Joint deficiencies	Scaling
Map cracking	Popouts
Scaling	Blowups
Popouts	Joint deterioration
Joint Faulting	
Blowups	

ASR in road pavements is manifested mainly in the form of a network of cracks, mainly on the whole concrete surface, joint deficiencies/deterioration, silicate gel exudation, local concrete chipping and ravelling (Fig. 2).





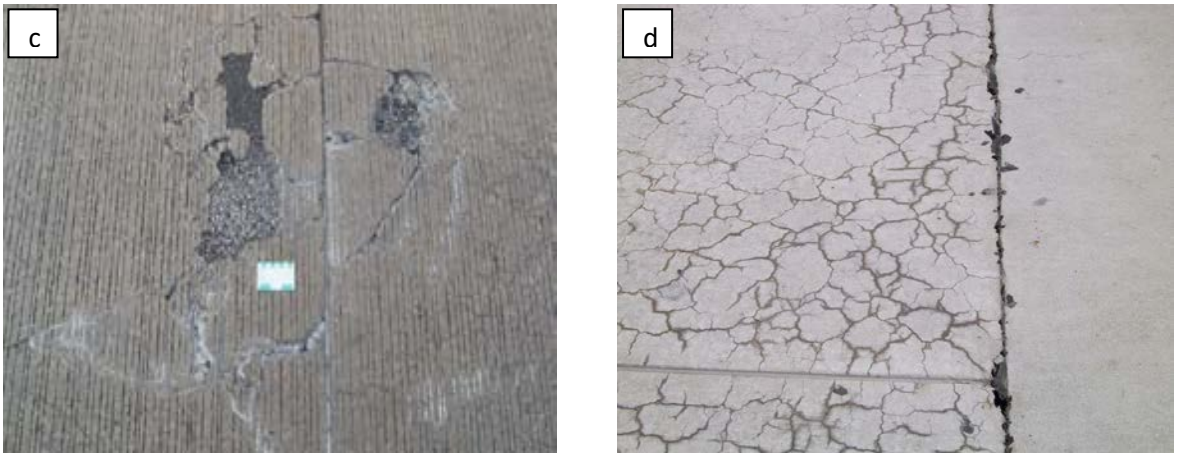


Figure 2. Examples of ASR induced distress to pavements (a-b), gel exudation and delamination (c), cracking and joint spall (d) [14,17-19].

Damage occurs mainly in the vicinity of joints and is similar in appearance to D cracking, also characteristic of frost attack. The crack pattern is closely related to the geometry of concrete, reinforcement layout, stress fields and environmental impacts. Mesh (map) cracks typically have a random orientation. Restraining the shape in one or more directions makes the cracks be oriented in the direction in which the stresses are lower. In concrete surfaces, cracks are usually parallel to the side edges restrained by curbs [20].

4. Methods of limiting ASR inducing factors

The reaction of alkali with aggregate takes place when the following three factors occur simultaneously:

- sufficient concentration of sodium and potassium hydroxides in the pore solution. Alkalis derive mainly from Portland cement, although the source may also be aggregate, mineral additives and chemical admixtures used in concrete or de-icing salts,
- the presence of reactive silica in the aggregate,
- sufficient moisture of around 80%. Water, introduced into the concrete mix as mixing water and that coming from external sources, transports the ions and when absorbed by silicate gels leads to concrete cracking.

If any of these factors is eliminated, ASR will not take place. It should be noted that the elimination of the first two factors is only possible at the mixing stage. Aggregates that do not have alkali reactivity and low alkali cements should be used, along with pozzolans or hydraulic mineral additives as a partial cement replacement. Reducing the amount of water by using a lower w/c ratio (w/s) can increase the alkali concentration in the pores in the concrete, thereby increasing the rate of silica dissolution. Another way is to incorporate a chemical admixture in the form of lithium compounds, mainly lithium nitrate and lithium hydroxide, into the concrete mix. It is believed that lithium ions protect the reactive silica grain against reaction with sodium and potassium hydroxides, forming a protective layer on its surface. This layer consists of nonexpansive lithium silicates or less expansive silicates with a low sodium and potassium contents enriched with lithium ions [21, 22].

In a concrete member, in which the aggregate happened to be reactive, limiting further destruction is possible in several ways classified into two categories [23]:

Treat the cause			Treat the symptoms		
CO_2	Lithium compounds	Drying	Crack filling	Restrain	Relieve stress

In the case of concrete pavements, recommendations include the use lithium compounds, reduction of humidity (treat the cause) and slow cutting/saw cutting (treat the symptoms). These methods will be discussed in more detail in the following sections.

4.1 Treat the cause

Lithium compounds applied to ASR-affected concrete is the promising method. Although the mechanism of lithium ions behaviour is not fully understood, it is believed that lithium modifies the properties of ASR gels changing their expansive character into nonexpansive [22]. The first studies conducted in 1993 -2004 indicated high efficiency of lithium compounds in limiting the expansion of AAR affected concrete samples analyzed in laboratory conditions. Promising research results became the driving force of further experiments, mainly in the United States as part of the Federal Highway Administration (FHWA) projects. The main application method for lithium compounds is placing them as a surface layer on pavement slabs, airport plates, road barriers, bridges. In order to increase the depth of lithium penetration, other techniques such as vacuum and electrochemical methods are used [23].

4.1.1. Topical application

The application of lithium compounds, primarily lithium nitrate, that has become quite common in the recent years is the surface overlay used on road and airfield pavements and on highway dividers. Lithium nitrate in the form of 30% $LiNO_3$ solution is applied to vertical surfaces using a handheld pressure spray applicator and a truck-mounted spray bar for depositing the solution on horizontal surfaces (Fig. 3) [24]. The $LiNO_3$ solution is most often used at a rate of 0.06 L/m^2 to 0.40 L/m^2 in one or several applications. It has been observed that better penetration of lithium ions is achieved by several applications of smaller amounts [25]. Considering the depth of penetration, the surface treatment with lithium nitrate is not as effective as vacuum or electrochemical impregnation, but in the case of large areas it is more economical.

The efficacy of lithium treatment on ASR affected surfaces of concrete, pointed out by the Federal Highway Administration U.S. Department of Transportation is affected by:

- the type of lithium compound. Lithium nitrate is the only recommended compound because it does not increase the pH of pore solution or, unlike lithium hydroxide, induce the pessimum effect;
- the quantity of lithium nitrate used and the number of repetitions. The surface needs to be covered evenly. Too large amounts of $LiNO_3$ solution may lead to runoff from the treated surface or through evaporation, to $LiNO_3$ precipitation in the form of a salt visible as a white residue on the concrete. Minimum two applications at least 30 minutes apart;
- adequate surface preparation – clean, debris-free, wet surface;
- temperature and wetness of concrete during treatment. The treatment cannot proceed during rainfall or if the rain is forecast within 6h;
- properties of concrete, i.e., the degree of damage due to ASR, permeability,
- additional environmental factors that reduce the durability of concrete, e.g., freeze-thaw cycles. If in addition to ASR, frost attack occurs, other protective measured should be applied such as crack sealing or overlays.

If the precipitated lithium nitrate covers more than 5% of the surface area, water should be applied and until the surface is deemed safe for vehicles [25].

Research on the effectiveness of lithium compounds introduced into hardened concrete, in which the reaction proceeds, was conducted in the 90s of the 20th century. Sakaguchi et al. studied mortars and concrete with reactive aggregates (Pyrex glass) impregnated with the $LiNO_2$ solution after reaching the sufficient level of expansion [26]. The impregnation was found to limit or inhibit further expansion in the specimens. Stokes et al. added a surfactant to the $LiNO_3$ solution to enhance the penetration of lithium ions deep into the concrete structure. They also demonstrated that the higher the extent of expansion, the extent of cracking and the interconnection of microcracks, the easier the path of the lithium solution penetration to a depth of 50 mm (Fig. 4). The effectiveness of concrete surface impregnation with a 30% lithium nitrate solution was investigated on State Route 1 in Delaware. Lithium nitrate was applied two times a year at a rate of 0.24 L m^2 for three years. Four years after treatment, untreated area revealed serious distress in the form of numerous cracks, chipping of concrete at transverse and longitudinal joints. The area of concrete subjected to six cycles of surface treatment with lithium nitrate showed less damage, observed in the form of smaller spalling of concrete on joints (Fig. 5) [27].

Hence the degree of damage to concrete has an impact on lithium ions penetration. Johnston pointed out that ASR cracks facilitate the penetration of the lithium nitrate solution but if the amount of deterioration is too large, it is no longer possible to protect the member against further corrosion by applying lithium nitrate [28].



Figure 3. Surface treatment with lithium compounds [24].

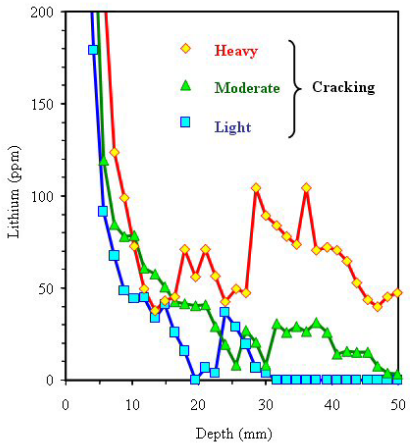


Figure 4. Lithium concentration profiles for concrete pavement after six treatments [27].

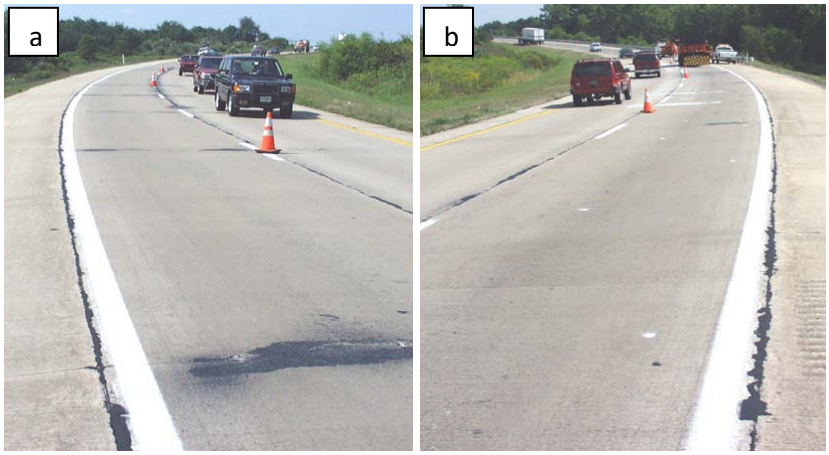


Figure 5. ASR affected pavement in Delaware a) control (untreated) concrete, b) after topical treatment with 30% lithium nitrate solution [27].

Numerous studies carried out as part of FHWA projects have shown limited penetration of lithium applied by spraying and limited effectiveness of the treatment at the long term protection against expansion and cracking. Folliard and co-workers demonstrated that after three lithium applications on the surface of concrete in Idaho, the $LiNO_3$ concentration exceeded 100 ppm, the dosage considered sufficient to limit further deterioration of concrete only in the surface layer, at a depth of 3-4 mm [29]. Similar results were reported when $LiNO_3$ was applied to a 26-kilometre concrete road in Geogetown, DE, at a dosage of $0.061 L/m^2$. The lithium levels were determined form the cores extracted immediately after treatment and several months later. The lithium concentration above 100 ppm was found at a depth of 3-10mm. The probability of ASR inhibition was in that case very low [30].

4.1.2. Vacuum impregnation

The lithium nitrate solution can be introduced into the concrete using the vacuum impregnation method as an alternative to pressure injection. The method consists in creating a suitable underpressure to remove air and moisture from the cracks in concrete, thus creating more space for impregnation. After a vacuum of about 0.5 atmosphere was achieved, lithium is drawn (sucked) into the evacuated area in the concrete. The solution fills the air voids, microcracks and cracks [24].

Vacuum impregnation was used to repair ASR-affected structures in South California and Pennsylvania. The Pennsylvania Department of Transportation applied this method to bridge elements – abutments, sidewalks and wing walls. However, due to the difficulty in performing vacuum impregnation on larger members, this method is not used in large road sections. Vacuum impregnation, used mainly in the repair of bridge pillars and road barriers, provided a deeper penetration of lithium ions compared to the topical application. The concentration of lithium above 100 ppm was recorded at a depth of 2-4mm in road barriers and 8-10mm in bridge pillars [29]. FHWA Project's research on the effectiveness of vacuum impregnation indicated that the penetration of lithium ions is limited only to the surface layer. Drimalas and colleagues used, among others vacuum impregnation method for the application of $LiNO_3$ in columns of road bridges in Houston, which have been damaged to a different degree as a result of alkaline corrosion. A 30% solution of lithium nitrate was used at $1.63 L/m^2$. The effectiveness of the vacuum impregnation was also investigated in the ASR-affected road barriers located to the west of Boston in Massachusetts. Monitoring of the columns of the bridge in terms of expansion and surface cracks indicated low effectiveness of the impregnation method. Similar results were obtained in the case of road barriers, where no reduction in expansion levels was observed. Improvement was achieved after applying hydrophobic agents [31]. High costs and complexity of the process are the main drawbacks and limitations of this method, with a small difference in the depth of lithium ion penetration compared to the topical method.

4.1.3. Electrochemical methods

Electrochemical methods have been developed for the electrochemical chloride extraction (ECE) to protect reinforcement against corrosion. By applying a low-voltage direct current, the chlorine ions located close to the reinforcement (cathode) migrate towards the surface anode. After several modifications, this method has been implemented for the application of lithium ions in the concrete structure [32, 33]. Reinforcement is usually the cathode and the anode is attached to the surface of the member. Following the application of the current of adequate voltage, usually up to 40V (bridge deck), positively charged lithium ions travel towards the cathode. The duration of the entire process is from several days up to 8 weeks [24].

Due to the greatest depth of lithium penetration achieved, this method is the most favourable of all the methods. The electrochemical impregnation with lithium ions is not used on road pavements, but offers promising results in the case of other forms of transportation infrastructure. Studies on the depth of lithium penetration in ASR-affected bridge elements showed that the concentration of lithium sufficient to limit the expansion of concrete (above 100ppm), occurred at a depth of up to approximately 50 mm [34]. One side effect of the electrochemical lithium treatment is its ability to accelerate corrosion processes. In the electrochemical process hydroxyl ions are formed on the

reinforcement (a cathode). To offset the negative charge, the sodium and potassium ions from the concrete migrate towards the cathode, increasing the susceptibility of concrete in the reinforcement area to AAR. Researchers recommend proper monitoring of the structure over time for cracks and changes in the microstructure around the reinforcement to see whether the benefits from the increased depth of lithium penetration outweigh the potential effects of the pH increase in the concrete pore solution in the vicinity of the reinforcement [23, 35].

4.1.4. Reducing humidity

As mentioned before, high relative humidity is one of the factors necessary for ASR to initiate. A drainage system can be improved, concrete can be sealed using suitable materials or external cladding can be applied to reduce water penetration into concrete. However, the mixing water used for concrete mix production is often sufficient for the ASR processes to start and proceed. As indicated by Diamond, concrete degradation is possible without an external source of water, which can just be the capillary pore solution [36]. A more adequate way to protect a member against deterioration is the use of protective coatings (sealers). The coatings retard external water penetration while allowing the water vapour exchange - vapour-permeable coatings. The most commonly used agents include silanes and siloxanes. In concrete pavements, where water is available through capillary action from the base and subbase layers, the use of silanes may be ineffective and the reduction of internal humidity is only possible in the surface layer. One advantage of using agents based on silanes and siloxanes is the reduction of external chloride penetration.

Silane was used to treat a 19 km long section of pavement in Pine Bluff, Arkansans where the signs of ASR were observed [30]. Petrographic analysis revealed that the cracks and exudations were caused by alkali reactive chert. A 100% and 40% siloxane-based agent was mechanically sprayed at a dosage of 3.1L/m^2 on the sections where the distress was a) mild, b) moderate. Seven months after the treatment, the temperature, the extent of expansion and the relative humidity of the concrete at a depth of 25 mm and 75 mm were analyzed. No significant differences were found compared to the reference samples, but a need to extend the monitoring period was pointed out to be able to determine if the siloxane agent was effective in reducing the humidity and ASR-induced expansion. Pavements in Arkansas, USA were subjected to topical treatment with silane to observe the effects of both the reaction between alkalis and moderately- reactive fine aggregate and the frost attack [37]. The silane sealer was sprayed at a rate of 150 L/m^2 (40% water-based silane) and 250 L/m^2 (100% water-based silane). During the three-year long monitoring period, strains (pavement panels were instrumented with a grid for measuring strain) and the internal RH were measured. Results showed a reduction in the relative humidity and a significant reduction in strain levels following the 100% silane application. A need for further monitoring was reported.

4.2 Treat the symptoms

4.2.1. Crack filling

Filling cracks is particularly important in concrete pavements as the cracks allow water and aggressive ions, including chlorides to easily penetrate the concrete. Flexible grouts or sealers can be used as the filling material. Rigid materials, including cement-polymer mortarsepoxy resins may cause cracks around the bonded area [23].

4.2.2. Saw cutting/slow cutting, concrete replacement

Cutting slots or expansion joints can be an effective, but mostly temporary, method of relieving ASR induced stresses in the concrete. This does not, however, eliminate the causes of ASR. These methods are temporary solutions for use in the members in which the concrete expansion proceeds and the treatment needs to be applied repeatedly.

A frequent method of extending the service life of concrete pavements is the removal of some of the concrete around the joints damaged by the expansion, where ASR is most frequent. The

replacement of concrete in the vicinity of joints by saw cutting is recommended when ASR is accompanied by freeze-thaw action. Soundproof concrete can be used as a complementary material. Joint details (opening, dowel bars) have to be restored [17, 19].

5. Summary

Alkali-aggregate reaction reduces the durability of concrete in road infrastructure facilities. Numerous cracks become the migration paths for corrosive agents, such as chlorides and sulphates. The increased humidity of concrete members accelerates their deterioration due to freezing and thawing cycles. AAR control or mitigation is thus important.

Minimizing the ASR effects when reactive aggregates were used in the mix is difficult. Multiple examples of internal corrosion of concrete in road infrastructure assets indicate that the problem of aggregate reactivity needs to be addressed. One of the methods for minimizing the progress of ASR is the use of lithium compounds, which have the ability to change the properties of reaction products - sodium-potassium-calcium silicate gels - from swelling to non-swelling. Road infrastructure at many sites was treated with a solution of lithium nitrate introduced into the concrete structure by surface treatment or by vacuum or electrochemical impregnation, leading to the reduction in the number of cracks and spalls. The key problem is obtaining the appropriate depth of lithium ion penetration, which depends on the specificity of the reaction occurring in the entire volume of concrete. In road pavements, the application of $LiNO_3$ is confined mainly to surface and vacuum treatments (small area) providing insufficient lithium penetration depth to ensure the efficacy of the methods. Further research and monitoring of the ASR-affected members treated with lithium compounds is thus necessary.

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