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EFFECTS OF ENVIRONMENTAL CONDITIONS ON ALKALI-AGGREGATE REACTION AND PREVENTIVE MEASURES

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1. INTRODUCTION

At first the **alkali aggregate reaction** (AAR) is discussed. The effects of environmental conditions and the preventive measures are derived from the factors influencing the AAR. They are summarized at the end of the paper.

There are four factors that must arise simultaneously to exhibit alkali aggregate reactions in mortar and concrete:

- soluble alkalies,
- reactive lime,
- alkali reactive aggregates and
- unfavourable environmental conditions.

Preferably the **alkalies** are constituents of cements and should be reduced especially in the case of ordinary Portland cement (OPC) to a level commonly assumed to be <0.6 wt.-% Na_2O-equiv.. Finally, the total alkali content should be related to the entire concrete mix.

In the case of **blended cements** containing admixtures which are highly effective with respect to their chemical and physical reactivity and their amount of replacement, the alkali content is of minor influence. Especially, this is true for the use of Portland blast furnace slag cement (PBFSC) with a sufficiently high rate of qualitatively good slag.

Effective admixtures as replacement of OPC or as addition to the concrete mix diminish the available **lime content** in the cement-stone of the hardened concrete and are of predominant influence. In this context the available lime is

- the free lime as portlandite and partly
- the lime softly bonded within high-lime calcium silicate hydrates and calcium aluminate hydrates.

This active lime is able to precipitate gel-like alkali silicate hydrates to solid alkali calcium silicate hydrates and to form semipermeable membranes permeable to alkali ions and water molecules. The membranes are impermeable to silicate anion complexes.

Alkali reactive aggregates (ARA) of the silica type and the silicate type are predominantly fairly dense silicious materials forming alkali silicate hydrates containing more or less lime.

In the case of opaline sandstone, for example, with bulk densities < 1.6 g/cm^3 corresponding with total porosities >43 vol.-% no difference to the control with respect to an alkali silicate reaction (ASR) causing a measurable damage was gauged (1). Partly there are reactive dolomitic limestone and/or clay mineral containing limestones. Their reactivity depends on a destructive dedolomitization or on the expansion of special clay minerals in limestones. This paper mainly deals with the alkali silica reaction or the alkali silicate reaction. Possibly there are some small differences in the chemical and physical behaviour of the resulting gels and solids but here the overall reaction mechanism is supposed to be equal and therefore summarized as ASR.

Besides soluble alkalies, reactive lime and ARA the **environmental conditions** of the concrete structure that could be affected by the AAR are of fundamental influence on the occurence of damaging ASR or AAR. In laboratory tests as well as in concrete structures and members in practice moisture content and temperature as well as their time dependent changes are the most important conditions influencing the degree of attack. Besides there is a considerable effect given by the size of mortar and concrete probes and structures.

Frost-thawing cycles and wet-dry cycles will increase the extent of the deterioration and its visible signs.

This short report attempts to combine all these complex features based on personal experience and on literature, to evaluate them and to compare them to existing findings on the basis of the **ASR mechanism** called **dynamical osmotic equilibrium** (2).

This model is in progress. It considers the ideas of many well known scientists who partly did their work about 80 years ago up to those still working today (2-11). The main fundamentals are

- the Donnan membrane equilibria,
- the semipermeable membrane formation and
- the behaviour of the alkalies within the structure of cement mortars and concretes.

The semipermeable membrane is formed by the cement stone and by the hydrated calcium alkali silicate gel blocking the permeable voids of the cement stone.

2. INFLUENCES ON AAR

In the literature sometimes it is referred to

- visible damages on structures
  (map cracking, pop outs, gel exudations),
- measurement of expansion and, only occasionally,
- measurement of mechanical resistance.

In the following the influences on AAR will be discussed in detail.

As mentioned before the discussion is based on literature and on the work at the GHI. At the latter, work was done with mortar bars according to DIN 1164 (cement : sand : w/c = 1 : 3 : 0.5; size 4 x 4 x 16 cm^3) and needed deviations to achieve a given aim. In deviation to ASTM C 227-87 the cement content is about 500 kg/m^3. On the mortar bars besides the visible signs of AAR the changes in mass, length and resonance frequencies were gauged at given times. Additionally, mineralogical examinations were performed (1,2,12,13). The same examinations were done on specially taken and handled concrete cores (14). According to the work done at the GHI it is very important to point out that it is insufficient to measure the expansion only, since it does not give a complete picture of the time dependent slope of the resistance of attacked mortar or concrete. One possibility to calculate this slope is to measure the resonance frequency.

2.1 Soluble Alkalies in Cements

The need for the limitation of the **alkali content of OPC** to 0.6 wt.-% Na_2O-equiv. in concrete structures with ARA is known since the work of Thomas E. Stanton published in 1940 (15) and is confirmed since that time in numerous publications. But frequently it has been stated that in special cases <0.6 wt.-% Na_2O-equiv. is an insufficiently high value. He already calculated the K_2O+Na_2O as wt.-% Na_2O-equiv.. These wt.-% Na_2O-equiv. of OPC are predominantly varying in the range of 0.2 to 1.2 (16).

The ASR of equivalent amounts of alkalies decreases with the following order: K_2O - Na_2O - Li_2O. There are several causes for this behaviour:

- decreased diffusion rate (9),
- decreased limiting concentration value for silicid acid coagulation (2) and
- decreased alkali silicic acid reactivity (12).

Some researchers measured the water soluble alkalies according to ASTM C 114-85 as there is only a relatively low coefficient of correlation between mortar bar or concrete test results and the alkali content of OPC or the alkali content of mortar or concrete mix. But finally after 28 to 90 days the alkalies of OPC are soluble to an extent of 90 wt.-%. (17). The solubility of alkalies might be increased by higher fineness of the cement and by additives or elevated temperatures increasing the overall hydration reaction (11,18). On the other hand, with less reactive aggregates, coarse sized RA, or low alkali OPC in practice or in laboratory tests the damaging reaction will start occasionally after 1 to 4 years (13) or later. That means the evaluation of OPC should be done by the amount of the alkalies soluble in hydrochloric acid.

This method can be applied to **blended cements containing latent hydraulic blast furnace slag** (BFS) like BFSC as the wanted good quality BFS also is completely soluble in hydrochloric acid. The benefit of BFSC with respect to AAR is commonly accepted (10, 13,19,20,21). To achieve a sufficiently high diminuition of ASR the needed amount of OPC replacement by BFS is proved to be between 35 and 65 wt.-% with a mean value at 50 wt.-% (10,13,19, 20,22). There is only less influence of the alkali of the BFS itself. With respect to the adopted value of 0.6 wt.-% Na_2O-equiv. for OPC, Bryant Mather pointed out that "a similar limit for portland blastfurnace slag cement might well be established at a considerably higher point, perhaps somewhere in the vicinity of 1.20 % Na_2O-equivalent". With the help of test series and statistical methods the effective alkalies (A_{eff}) of BFSC were calculated anew (20),

$$A_{eff} = A \left(1 - \left(\frac{H}{H_o}\right)^2\right) \qquad (1)$$

A - total wt.-% Na_2O-equiv. in BFSC
H - BFS content
H_o - needed BFS content to prevent expansion

This work is part of the German regulations (21) on **low alkali cement** (LAC).

The higher values of permissible Na_2O-equiv. in PBFSC compared to OPC are resulting from

- the intergrowth of the hydration products of clinker and slag particles (10) and
- the lower lime content (24,25)

in the hardened cementstone of PBFSC.

This leads to a considerably lower permeability (10,25) and a higher alkali fixation in the mortar and concrete designed with PBFSC. The higher alkali fixation of hydrated PBFSC can be concluded from experimental data showing an increasing alkali content of solid CSH with a decreasing c/s-ratio (27) and other data showing that the wt.-% Na_2O-equiv. concentration in the pore solution of hydrated PBFSC is about 5-fold lower than that of comparable hydrated OPC (27).

In the European countries the replacement of cement clinker by BFS and other metallurgical slags is based on many years of experience. The BFSC with 35 wt.-% replacement of OPC is standardized since 1909 and the BFSC with up to 80 wt.-% slag since 1917. Thus the quality of the slag has been a problem between the blast furnace operator and cement manufacturer for a long time. The quality of the slag is defined by the chemistry, the quenching or the glassiness and the fineness.

The effect of pozzolans on damaging ASR is comparible to that of BFS discussed above. But, in general, an evaluation of pozzolans in **Portland pozzolan cement** (PPC) is more difficult whilst the rating of the alkali content of OPC and PBFSC has arrived at a satisfactory solution. Generally, the evaluation of hydraulic reactivity of cementing materials is in the following sequence

$$OPC > BFS > P \qquad (2)$$

That means, the hydration of OPC immediately starts with the water addition, that of BFS in PBFSC within minutes to hours and that of pozzolans in PPC within days to weeks. The chemical and mineralogical composition of OPC and BFS varies only within a small range. On the other hand, the chemical, mineralogical and surface physical measures of pozzolans vary within a wide range. Predominantly, the silica content varies from about 45 to nearly 100 wt.-%, the effective alkali content, calculated as wt.-% Na_2O-equiv., varies from about 0 to 7 wt.-% and the BET-surface area from about 0.5 to >50 m^2/g.

Because of this wide range of variations in the chemical and physical properties a simple formula for the calculation of effective alkalies in PPC does not exist. Moreover the cement replacement must be related to the activity and the alkali content and release of the pozzolan along with the alkali content of the applied OPC. That means, the activity of the different types of pozzolans is of importance in the case of PPC. The test method according to ASTM C 311-87 to determine the pozzolanic activity index with OPC after curing for 28 days at 38°C under moist conditions is insufficient for this purpose.

The effective alkalies of pozzolans are water soluble or silicate bonded. The water soluble ones are easy to determine. The silicate bonded alkalies are released at the reaction with free lime (28) and there exists a strong correlation between alkali release and lime fixation in the case of some puzzolanic fly ashes (29). These experimental data were maintained at the hydration with excess water. But up to now there is no evidence that the amount of hydrochlorid acid soluble alkalies is the same as that released at the hydration with lime. The ASTM C 311-87 test method to determine the available alkali after curing for 28 days reaction with lime at 38°C is sufficient.

During paste hydration of OPC blended with opal, shale or fly ash, decreasing c/s-ratios of CSH in the OPC and the corresponding blends caused an increased alkali absorbtion (30).

The effect of high active pozzolan on the diminuition of damaging ASR is caused by lime and alkali fixation and by improved tightness of the structure of hardened PPC. But frequently the chemical and physical behaviour of the pozzolans are insufficient. Therefore, in practice, for a safe construction a larger amount of OPC replacement or an improvement of pozzolan is necessary.

2.2 Reactive Lime in Cements

A measure for the presumable amount of reactive lime in hydrated OPC is the lime standard

$$\mathrm{KStIII} = \frac{100\ (\mathrm{CaO} + 0.75\ \mathrm{MgO}^{*})}{2.8\ \mathrm{SiO_2} + 1.18\ \mathrm{Al_2O_3} + 0.65\ \mathrm{Fe_2O_3}} \quad (\%) \qquad (3)$$

The active lime content increases with raised lime standard. The available lime in hydrated cements is bond in portlandite, in high lime calcium silicate hydrates and in calcium aluminate hydrates. It can be determined by the improved Franke method. With this method not only free lime is extracted but also the lime which is liberated when the c/s-ratio in hydated calcium silicates is reduced to 1.5 and the c/a-ratio in tetra calcium aluminate hydrate to 3.

The free lime in the hydrated OPC measured by this method amounts to 20 to 30 wt.-% CaO. At room temperature the lime liberation is finished after about 3 to 7 days hydration (28).

The lime fixation of 7 different slags amounts to about 50 to 73 mg CaO/g BFS after 1 day hydration and to 106 to 125,0 mg CaO/g BFS after 14 days hydration. The water/cement (w/c)-ratio was 5 and the proportioning was 2.5 g clinker + 2.5 g BFS + 0.35 g anhydrit. With slag contents up to 75 wt.-% the lime fixation decreased and at lower w/c-ratio especially the early lime fixation increased (24).

The pozzolan lime reaction was examined at 2, 10 and 20°C and a water/binder (w/b)-ratio of 1 with high reactive silica fume and lower reactive Rhenish trass (31). The results clearly indicate the reduced lime fixation at 10 and 2°C, especially with Rhenish trass. On the other hand the lime liberation of POC is not as much retarded at low temperature as the lime fixation of highly reactive pozzolan.

2.3 Alkali Reactive Aggregates

There are only some facts on ARA that should be mentioned here without going into detail. Main influences are

- the pessimum amount of ARA,
- the reactivity of silica and silicate and
- the grain size distribution.

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*) with MgO contents $\leq$ 2.0 wt.-%

The reactivity of the ARA is given by the cristallinity of the silica, the lattice distortion and/or the grain size of the quartz. The most reactive ARA in Germany has an amorphous opaline character.

Increased grain size provokes a delayed reaction (13). In practice a broad range of grain size distribution of ARA has to be taken into account (14). Partly the ARA have a low frost-thaw resistivity with or without di-icing agents.

But there are different kinds of opaline sandstone from the same gravel deposit.

- A less deliterious acting calcitic grey-white material (~30 wt-% calcite),
- a high destructive acting less calcitic grey-green material (~4 wt.-% calcite),
- a less destructive acting calcitic grey-green material (~50 wt.-% calcite).

On the other hand opaline sandstone with bulk densities <1.6 g/cm^3 and those with values $\geq$ 2.3 g/cm^3 will exhibit no damaging ASR (1,13). The high reactivity of opaline sandstone and the lower reactivity of some porous flints were taken into account in the German regulations on ARA from Schleswig-Holstein. ARA with grain size < 1 mm are neglected. This question has to be examined since especially the small sized ARA can provoke an early rapid expansion accompanied with a dangerous decrease in modulus of elasticity. At the valuation of concrete of bridge side walls after about 6 to 7 years service life time the visible size of ARA was about 2 to 8 mm in diameter and the amount was calculated to 1 to 2 wt.-% (14). This result, however, cannot be a final valuation of the occurence and the behaviour of damaging ARA sized < 1 mm in practical concrete structures!

2.4 Concrete Mix Design

An important influence on AAR is given by the design of the concrete mix:

- proportion of cement,
- w/c-ratio,
- admixtures,
- additives and
- mixing process.

The proportion of cement gives a value of the amount of A_{eff} in concrete. But what might be the maximum value for safe concrete at pessimum conditions of all factors influencing ASR? It is supposed to be < 2 kg/m^3 effective Na_2O-equiv. (13,14,32). That means that the limitation of wt.-% Na_2O-equiv. of OPC should amount in maximum to about 0.4 instead of 0.6 as being commonly adopted so far.

From 0.4 to 0.7 increased w/c-ratios will exert raised losses in relative E-modul related to the maximum value. The final expansion is within a narrow range from 8 to 9 mm/m. Only the mortar with a w/c-ratio of 0.7 has an expansion of 7 mm/m (13). With concretes increased w/c also exhibits worse damage (33).

High active pozzolans or BFS as admixtures in concrete are reducing ASR, as already discussed for PBFSC and PPC, especially if they are serving as OPC replacement (1,13). There is no experience on the influence of inert fillers in conrete.

Alkali containing agents like some superplasticizers will increase predominantly the sodium content of the pore solution of mortar or concrete from about 0.5 to 1.8 mg Na_2O/ml and thus increase the early expansion rate and the drop in resonance frequency (13). In mortar bars air entraining agents exerted a beneficial effect. At about 8 vol.-% void space the expansion was reduced from initially 14 to 0 mm/m (34). This favourable effect could not be repeated with concretes. Within the range of 3 to 4.8 vol.-% air voids no measurable positive effect was proved (33).

At the mixing process addition of dry opaline sandstone along with the standard sand to the premixed cement slurry exerts a 2-fold initial expansion and an accompanying stronger drop in resonance frequency than the addition of wet opaline sandstone. The total expansion is the same in both cases. But the delayed ASR with wet addition results in a more severe damage of the structure established by the resonance frequency measurement (1).

2.5 Environmental Conditions

The main influencing environmental conditions are

- the temperature and
- the moisture

and their time dependent changes including frost-thawing cycles and wet-dry cycles and their influence on dimensions of the mortar or concrete pavement or member.

The accelerating influence on damage formation of increased temperatures is well known for 45 years (35) and has been repeatedly confirmed since that time. These tests were performed

- with 22 different aggregates of non to high reactivity and a grain size from 0.3 to 4.8 mm,
- with a high alkali cement (without giving a value) and partly plus 0.5 wt.-% Na_2O added to the mixing water and
- moist cured at 21.5, 43,5 and 65,5 °C for 2 years.

Predominantly raised temperatures caused increased and accelerated expansion. In some cases the 43.5°C cured prisms achieved the highest values. Partly with inert quartz expansions were measured only at higher temperatures, especially with extra Na_2O

addition. Extended research at room temperature and at 43.5°C with OPC containing 0.98, 0.59 and 0.14 wt.-% Na_2O-equiv. (corresponding to high, medium and low alkali content) and three different ARA leads to the conclusion that "elevated temperatures increase the initial rates of mortar expansion but they decrease the total expansion" (36).

Other researchers confirmed a maximum expansion at about 40°C (37,38) examining the temperature range from 20 or from 4 to 60°C respectivly. They used mixes with about 6 or 9 kg Na_2O-equiv./m^3 respectively. After 1 year at 4 and 10°C the mortar bar expansion amounts only to about 30% of that one achieved at 20°C. But the reaction was still incomplete (38). Thus, a final conclusion with respect to damaging effect of low temperature can not be drawn by now.

The moisture content of concrete needed to exhibit damaging ASR is present in concrete buildings, members and pavements exposed to open air weathering. Destruction of mortar or concrete by ASR was achieved at about 20°C and 80 to 85% r.h.. The most severe damages were observed at delayed ASR and at ASR performed at low humidities like 95, 90 and 85% (1,13). A delayed restorage from 20 to 40°C curing temperature provokes less severe damage (13). The existing results of concrete structures are in good agreement with the data given above (39). ASTM mortar bars with about 9 kg Na_2O-equiv./m^3 showed already expansion at 73% r.h. and 38°C curing temperature (40).

Direct measures of deflection and expansion on concrete structures show the importance of changes of temperature and humidity during the seasons (41,42). These measures show an increased expansion at depths of 12 to 120 inch inside of the Parker Damm concrete (41).

The comparison of humid cured concrete beams and cubes at 40°C with those stored in open air after 10 years showed a more severe damage at normal weathering. Bigger sized specimen are more affected than smaller ones (33). In the case of bridges that are affected by ASR in Schleswig-Holstein the piers facing the south are signed by map cracking. At the north side no cracking is visible though the concrete cores are showing the same visible and measurable signs of ASR. That means additional temperature and humidity cycles by sun shine, wind and rain fall will increase the visible signs of ASR (14). Capillary transport mechanisms of alkali solution in the pore system of concrete may also increase pessimum conditions. Furthermore, reinforced concrete will show minor or increased cracking with respect to the positioning of reinforcement.

2.6 Surface Treatment of Concrete

The damaging influences on ASR by the environmental conditions can be significantly reduced by a suitable surface treatment. This treatment might consist in a sealing or a water repellent impregnation of the concrete surface.

The water repellent impregnation was done with an alkyl-alkoxy silane. The research definitely revealed that dense and porous mortars were better protected against ingress of water by this silane than by the hardened silicone resins commonly used at that time. This behaviour is attributed to the better bonding and the better penetration into the surface (43). The first results on the protection of mortar bars with ASR were very promising (1). Partly they found consideration with or without further improvement of the silanes (44-47).

3. CONCLUSIONS

On the basis of the ASR mechanism model of the existing experimental work the following conclusions can be drawn:

The temperature necessary to cause damaging ASR in concrete structures does normally exist. Raised temperatures exceeding 20°C accelerate ASR and decrease the damaging effects. Temperatures >55°C may be of lower interest. They can provoke decreased expansion with ARA or increased expansion with inert aggregates. At curing temperatures < 20°C only insufficient results are available unless the slope of measures on structures clearly indicates the poor reaction at lower temperatures. The changes of temperature including frost-thaw cycles will increase the damage.

The moisture content of concrete necessary to exhibit damaging ASR is present in concrete buildings, members and pavements exposed to open air weathering or being in contact with water. Decreased equilibrium humidities will delay the beginning of expansion and damage but will induce more severe destruction and will prevent the damaged structure from healing up. At about 20°C the minimum humidity causing ASR was determined to be 80 to 85%. This value might be lower at higher environmental temperatures. The changes in humidity and the migration of pore solution and alkalies will increase the damage and its visible signs in structures.

ASR can be prevented by dropping the total effective alkali content of a concrete mix to a value below 2 kg Na_2O-equiv. per m^3 concrete. This can be achieved by

- lowering the alkali content of the OPC,
- lowering the cement content of the concrete mix,
- OPC replacement by a sufficient amount and quality of BFS and
- OPC replacement by a sufficient amount and quality of pozzolan.

There is no knowledge on the effect of inert fillers admixtures on concrete designed with RA.

The effective alkali content of agents like superplasticizers must be considered. Some accelerating agents or higher w/c-ratios will also increase the ASR. Air voids are only partly found to be effective.

Water repellent impregnation of concrete surfaces can reduce damaging influences of ASR.

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