LATE-EXPANSIVE ASR DUE TO IMPORTED SAND AND LOCAL AGGREGATES IN OKINAWA ISLAND, SOUTHWESTERN JAPAN

Tetsuya Katayama ^{1,*}, Takeshi Oshiro ², Yoshinori Sarai ¹, Kiyoshi Zaha ³, Takahide Yamato ³

¹ Kawasaki Geological Engineering Co. Ltd., <u>TOKYO</u>, 108-8338 Japan

² Okinawa Forum of Concrete Durability, KAIHOCHO, Okinawa, 904-2162 Japan

³ Okinawa Operation Office, West Nippon Expressway Co. Ltd., <u>URASOE</u>, 901-2101 Japan

Abstract

Highway bridges and viaducts constructed 19 years ago are undergoing late-expansive ASR with crack density up to $12m/m^2$. The reacted mineral is chiefly cryptocrystalline to microcrystalline quartz in metamorphosed sedimentary rocks, contained in river sands imported from Taiwan and in sea-dredged sands from Okinawa, as well as in crushed limestone coarse aggregate from Okinawa. Locally, ASR is superimposed by highly reactive cristobalite in andesite, occurring in a dike exposed in the limestone quarry. Associated dolomitic limestone presented dedolomitization, producing brucite without cracking. Despite general late-expansive nature of ASR, cracking appeared in concrete with estimated total alkali even at Na₂Oeq < 2.5 kg/m³. Concrete core expansion test identified deleterious expansion, made only in alkali immersion. The complexity of reactions explains why previous studies failed to diagnose AAR.

Keywords: cryptocrystalline quartz, dedolomitization, EPMA, imported aggregate, late-expansive ASR

1 INTRODUCTION

Map-cracking of concrete bridges and viaducts has been noticeable since 1996 in the southern half of the Okinawa Expressway, which came into service in Okinawa Island in 1987. Because the mechanism of alkali-aggregate reaction (AAR) in the Ryukyu Islands had not been well understood, a technical committee was organized to clarify the nature and the extent of AAR to provide the basis for planning remedial measures [1]. Detailed petrographic diagnosis was performed of concrete core specimens taken from affected substructures in 2006, based on RILEM AAR-6 part 1 [2]. Besides crack densities, concrete core expansion under accelerated conditions, and water-soluble alkalis were measured. This paper summarizes results of investigation by this committee showing that late-expansive ASR occurred in combination with harmless dedolomitization of imported sands and local aggregates, along with early-expansive ASR of local aggregate. A procedure to diagnose AAR in Okinawa will be presented.

2 MATERIALS AND METHODS

2.1 Sample origin and extraction

Construction records indicate that a local crushed limestone coarse aggregate from Motobu, Okinawa was used throughout the expressway. Of the southern half of the expressway, the southern section (Arakaki to Futenmagawa) contained river sands imported from Hualien in Taiwan, while the northern section (Kitanakagusuku to Matsumoto) used well-desalinated sea-dredged sands from off the Arakawa coast of Okinawa, along with a crushed limestone fine aggregate also produced in Motobu. Concrete core samples (dia.100 \times L200mm, dia.50 \times L130mm) were taken from representative portions of 8 structures, including four bridges, three viaducts and one retaining wall (Table 1). They were wrapped with a polyethylene film on site, maintaining moisture before the laboratory work, to enhance exudation of ASR sol onto the core surface, which looked dark before solidifying into ASR gel.

2.2 Methods of investigation

Crack density and grade of deterioration

Structures were visually inspected to assess the progress of AAR. The grade of deterioration (field severity) was categorized in four stages, adjusting the Japanese guidelines on concrete practice to fit (JSCE 2001): I) the cryptic stage (no cracks); II) the developing stage (all cracks <0.2 mm); III) the accelerating stage (some cracks >0.4mm); and IV) the deteriorating stage (detachment of concrete cover). Representative concrete members were studied as to crack density, and cored. Cracks in the concrete

^{*} Correspondence to: <u>katayamat@kge.co.jp</u>

surface were traced with a marker onto an attached transparent vinyl sheet ($1 \times 1m$; at 1m height from ground). Crack lengths were gauged with a measuring wheel in the laboratory, and the crack density was calculated as a total length per unit area (m/m²) in four ranges (<0.2mm, 0.2-0.4mm, >0.4mm, full scale).

Core scanning, binocular and polarizing microscopy

A full sequence of concrete petrography was performed, from core scanning to EPMA analysis based on RILEM AAR-6 part 1 [2]. After removal of the cling film wrapper from the core specimens, a continuous image of their lateral surface (dia.100 \times L200mm, 360°) was obtained by the digital core scanner for modal analysis of the coarse aggregate (size >5mm, vol%) aided by PC software. Stereomicroscopy was done on every aggregate particle to check the presence of ASR sol/gel exuded onto the core surface. Polished thin sections of concrete (25 \times 35mm, thickness 15µm, not fluorescent) were subjected to polarizing microscopy to identify reacted rocks and minerals, cracks and gel products around them, and the extent of reaction (Tables 1, 2). Modal analysis of sand-fraction aggregates (<5mm) was made with an automatic point counter. Unhydrated cement particles were identified under reflected light.

The progress of ASR in each reacted rock type in thin section was categorized into four stages (Table 1): 1) the formation of reaction rims and exudation of ASR sol/gel around the reacted aggregate; 2) the formation of gel-filled cracks within reacted aggregate; 3) the propagation of gel-filled cracks from the reacted aggregate into surrounding cement paste; and 4) the migration of ASR gel into air voids [3]. Then the petrographic severity of ASR was rated, combining the above reaction stages with the extent of damage (crack widths and frequency) in concrete, i.e. (1) traceable, (2) minor, (3) moderate, and (4) severe.

SEM observation, EPMA analysis, minimum cement alkali and water-soluble alkali

After completing the polarizing microscopy, SEM observation and quantitative EPMA (EDS) analysis were done on ASR gel, CSH gel and unhydrated cement particles, using the same polished thin sections (JEOL JSM 5310LV/JED 2140: at 15KV, 0.12 nA, x 2000, data acquisition time 100s, dead time 30%, ZAF correction). Compositions were plotted on the [Ca/Si]-[Ca]/[Na+K] diagram (Tables 3, 4).

The minimum amount of cement alkali (Na₂Oeq), exclusive of water-soluble alkali sulfates, was estimated based on EPMA (EDS) analysis of unhydrated cement phases in concrete, assuming their content in the original cement clinker [3]. This was converted to the total cement alkali with a correction factor for the missing alkali of alkali sulfates. The alkali-budgets in concrete were also estimated, combined with wet-chemical analysis of water-soluble alkali by atomic absorption (40°C, <0.3mm, solid: water =1:10, shaking 30 minutes) of both bulk concrete and separated coarse aggregate (Table 5). Finally, the total available alkali in concrete was calculated as the sum of cement alkali and water-soluble alkali of coarse and fine aggregates. In this assumption, mix proportions in the construction records were used.

Concrete core expansion tests

To assess the expansion due to AAR, large core specimens (dia. $100 \times L200$ mm) were taken, attached with measuring studs in the field, and then stored in humid containers (20°C, 1 month; 40°C, 6 months) according to JCI-DD2. For comparison, small core specimens (dia. $50 \times L130$ mm) were immersed in alkaline solution (80°C in 1M NaOH, 4 weeks) to see the expansion potential of aggregates. These core tests have no RILEM equivalent, and are used when the same aggregate is no longer available.

3 RESULTS

3.1 Crack density and grade of deterioration

The expressway is located inland, without an external supply of alkali (Na) from sea-water. Mapcracking was most evident on the abutments where rain water was leaking from overlying girders. Such structure members in the southern section developed moderate deterioration (crack density: $6-12m/m^2$; >0.4mm: up to $3m/m^2$) of the accelerating stage of ASR, while northern structures presented minor deterioration with fine cracks (mostly <0.2mm: $3-10m/m^2$) of the developing stage (Fig.2, Table 2).

The severity of ASR roughly correlated with the petrography and field diagnosis within different boundaries. In Arakaki bridge, cracking has continued, with an average growth rate of $0.5m/m^2$ per year. One abutment was severely cracked in part (8-12m/m²; >0.4mm: 1-3 m/m²), with local detachment of concrete cover on the corner (crack 2mm) suggestive of a transition to the deteriorating stage, along with advanced reactions of various rock types cracking the cement paste. In Kitanakagusuku, where concrete columns were built in three stages, fine cracks appeared only in the block of the middle level (all <0.2mm, 10m/m²) indicative of the developing stage, with minor reactions detected microscopically. The sound lower block of this column was at the cryptic stage, with only traceable reactions (gel formation).

3.2 Alkali-aggregate reaction of coarse aggregates

Alkali-silica reaction

Limestone aggregates in the expressway came from a huge limestone body in the Mesozoic accretionary complex in Motobu, Okinawa, quarried and supplied by several plants for each construction section. In the southern structures, AAR was identified in the crushed limestone coarse aggregate where impure limestone (dark gray, argillaceous, mostly 20-30 vol%) and less than 2 vol% of andesite, derived from a thin dike exposed in the same limestone quarry, were found in concretes. Impure limestone with a few vol% percent of crypto- to microcrystalline quartz, produced a moderate degree of ASR with gelfilled cracks (width <40µm). These cracks originated from cryptocrystalline quartz that had converted to ASR gel, extending into surrounding cement paste (Figs.1 A, B). (Table 4). Andesite particles presented abundant ASR gel with wide expansion cracks (<300µm). Polarizing microscopy and SEM observation showed that rounded patches of primary cristobalite (<7 vol%) in andesite had converted to ASR gel, leaving a pseudomorphic texture with expansion cracks extending into the cement paste (Figs.1 G, H).

In the northern section, impure limestone coarse aggregate was only in the range of 5 vol% and andesite was absent, but crushed limestone fine aggregate was used. This caused no problem in concrete.

Dedolomitization

This paper first reports the late-expansive ASR and dedolomitization in the crushed limestone coarse aggregate in Japan. Dolomitic limestone and dolostone, irrespective of the type of aggregates or the size of dolomite crystals, produced dedolomitization with a marked reaction rim on their periphery, along with a carbonate halo in the surrounding cement paste. No expansion cracks were detected unless ASR was involved. SEM observation indicated that dedolomitization had produced a "mottled texture [4]", composed of fine grained brucite and secondary calcite (size 1-3µm), without cracking the cement paste. By contrast, gel-filled cracks derived from cryptocrystalline quartz extended into cement paste (Figs.1 C, D). On the dolomite rhombs contacting the cement paste, an alumino-bearing hydrate with Mg/Al atomic ratio around 3, suggestive of hydrotalcite 6MgOAl₂O₃CO₂:12H₂O, was identified by EPMA (EDS) analysis and mapping of elements (Figs.1 E, F, Table 4). In the dolomitic aggregate, cryptocrystalline quartz was not preserved but has been altered to Mg-silicate gel, instead of real ASR gel.

3.3 Alkali-aggregate reaction of sand aggregates

The Taiwanese river sands in the southern structures had similar lithologies rich in regionally metamorphosed sedimentary rocks (Table 1). ASR was identified as gel-filled fine cracks (up to 10μ m) extending from slate, phyllite (Fig.1 I), meta-chert (granular) and quartzite (Figs.1 L, M), as well as from non-metamorphosed mudstone, shale, siliceous shale, chert and some sandstone, constituting about 40 vol% of the sand fraction (Fig.3). Crypto- to microcrystalline quartz, a typical mineral of late-expansive ASR, reacted (Table 2). Nearly 30 vol% of the sand was metamorphosed carbonate rocks, of which dolomitic rocks exhibited dedolomitization, forming a carbonate halo in the surrounding cement paste without causing cracks. They came from the metamorphic terrain on the eastern coast of Taiwan.

The Okinawan sea-dredged sands in the northern structures also contained regional metamorphic rocks, i.e. slate, phyllite (Figs.1 J, K), meta-shale (siliceous phyllite), meta-chert (layered) and quartzite (Figs.1 N, O). These were lithologically similar to the Taiwanese sands, but their metamorphic grade may be lower, since the grain size of quartz was generally smaller. These contained sea shells (foraminifera, barnacles, calcareous algae, etc.) suggestive of varying dredge-sites, and non-metamorphosed sedimentary rocks with cryptocrystalline quartz. ASR occurred in the same rock types as in the Taiwanese sands, forming gel-filled fine cracks (<10µm) and reaction rims. In Kitanakagusuku, the cracked concrete block was rich in cracked particles of slate, whereas the sound block contained only rimmed particles, along with abundant crushed limestone fine aggregate occupying nearly 50 vol% of the sand fraction.

3.4 Compositions of ASR gel detected by EPMA

On the [Ca/Si]-[Ca]/[Na+K] diagram, alite and belite in concrete were hydrated to CSH gel along each compositional line, from the least hydrated state with [Ca/Si] atomic ratio of 3.1 and 2.1, respectively, down to final hydration products with [Ca/Si] =1.5 or less (Figs.4 A-D). Alite retained optical and compositional zoning, composed of a low-birefringent core and a high-birefringent rim with a corresponding scatter of [Ca]/[Na+K] ratios. This suggests that the original cement clinker was rich in SO₃ and that some alkali sulfates were present. Belite was richer in alkali than alite even during hydration.

ASR gel in four structures had essentially the same compositional lines, passing a point at [Ca/Si] =1/4-1/6, [Ca]/[Na+K] =1.0 (Figs.4 A-D), despite the variety of reacted rock types analyzed, for Arakaki (andesite coarse, quartzite and siliceous shale fine), Arakaki retaining wall (andesite and limestone coarse), Noborimata (limestone coarse, quartzite and slate fine) and Kitanakagusuku (quartzite and shale

fine). Crack-filling ASR gel had higher alkali (lower [Ca]/[Na+K]) ratios in the reacted aggregate, often rosette-like, than that in the mostly amorphous cement paste. In Noborimata, crack-filling ASR gel occupied a narrow, higher alkali range, while in Kitanakagusuku, ASR gel had a wide compositional range.

3.5 Alkali-budgets in concrete

Among the cement clinker phases expected, there was less alkali-aluminate, but interstitial glass occurred in some structures. The estimated minimum amount of cement alkali, assuming the content of each phase (e.g. alite 60 wt%, belite 20 wt%, aluminate 5 wt%, ferrite 5 wt%, glass 10 wt%), ranged Na₂Oeq 0.50-0.68%, i.e. total cement alkali 0.60-0.79% (empirically \times 1.2). Thus the total cement alkali in concrete corresponded to Na₂Oeq 2.0-2.7 kg/m³, based on the construction records of cement mix (mostly 340 kg/m³) and its water-soluble alkali (empirically \times 0.6) (Table 5). The measured water-soluble alkali of concrete was Na₂Oeq 1.3-2.5 kg/m³, of which 0.1-0.5 kg/m³ was in the separated coarse aggregate. The latter was higher in southern structures, where reacted impure limestone and andesite were present. Because the aggregates used in concretes could not be obtained, water-soluble alkali of total aggregates (plus chemical admixture), was calculated as the difference between that of concrete and of cement (0.1-1.1 kg/m³). Water-soluble alkali of fine aggregate, estimated including chemical admixture, was higher in northern structures. So the total available alkali in concrete, i.e. total cement alkali + water soluble alkali of coarse and fine aggregates + other derived alkali, amounted to Na₂Oeq 2.2-3.5 kg/m³.

3.6 Concrete core expansion tests

JCI expansion test of the concrete core specimens all gave less expansions than the criteria for deleterious expansion adopted by several organizations in Japan (0.05% at 3 months: Ministry of Construction; 0.1% at 6 months: Hanshin Expressway). (Fig.5). By contrast, the accelerated concrete core expansion test using NaOH solution presented expansions that all exceeded the deleterious limit in this test (0.1% at 3 weeks [3]). At 3 weeks, concrete cores from less deteriorated northern structures gave larger expansions (0.20-0.33%) than those from more deteriorated southern structures (0.12-0.20%).

4 DISCUSSION

4.1 AAR in Okinawa in previous investigations

In Okinawa Island, map-cracking was first noticed in a coastal bridge around 1985. Since then, investigations of AAR have been done by the Okinawa General Bureau, by the Okinawa Prefectural Government, and by Japan Highway Public Corporation (presently West Nippon Expressway Co. Ltd.), to find remedial measures for affected bridges and viaducts. Recently, a comprehensive review of AAR with additional tests was made to provide the background data to select concrete aggregates to be used for Irabu bridge [5]. In summary, the affected structures commonly contained crushed limestone coarse aggregate from Motobu, Okinawa, plus river sand and/or gravel aggregates imported from Hualien, Taiwan, the latter two being responsible for ASR. However, despite studies in the past decades, there has been little diagnosis of AAR and confident screening of reactive aggregates. This was due to inadequate methodologies being applied in Okinawa to identify late-expansive aggregates, as discussed below.

Previous petrographic examinations

Early examinations in Okinawa were done by non-petrographers, confined to the coarse aggregates separated from cement paste by acid treatment for XRD analysis. It was naturally not possible to identify what minerals had *actually* reacted to form ASR gel with expansion cracks in concrete. XRD analysis was also useless, because reactive crypto- to microcrystalline quartz was indistinguishable from inert quartz. Since concrete thin sections were used, no observation had been made of reaction sites on the *very* boundary between the aggregate and cement paste, but only potentially reactive forms of quartz had been identified in the aggregate. Some reports claimed an "abnormal expansion" of concrete, failing to detect ASR gel in cracks within concrete. Recently, ASR gel has been identified by SEM observation with qualitative EDS analysis, but it was not in the polished thin section, but on the fracture surface. Thus, all the petrographic data did *not* link with each other, far from authentic concrete petrography.

Non-petrographers used a fluorescence method (ASTM C856, annex) with uranyl acetate $UO_2(C_2H_3O_2)_22H_2O$, an internationally regulated radioactive substance, to identify ASR sol/gel in the concrete core specimens, skipping other confirmation methods. However, close-up photography of this method yielded too small an image to identify each reacted sand particle. Since Japanese regulations require the user to get permission to handle and store this hazardous material, and to keep waste solution permanently at the user's site, barring the shipment to waste processing plants, this method is not usable in Japan. Instead, this study showed that core-scanning with stereomicroscopy made upon unwrapping is an adequate substitute for identifying ASR sol/gel on the aggregate particles on the core surface.

Conventional standard tests

At the time of construction of the expressway, there was no standard AAR test in Japan, but it was revealed later that aggregates from the same sources passed Japanese standard mortar bar and chemical tests (JIS A1145 & 1146)[5]. This indicates that the current JIS standard tests will most likely miss potentially deleterious late-expansive aggregates containing crypto- to microcrystalline quartz, irrespective of whether it is a domestic or an imported aggregate. Hence, some accelerated tests may be useful.

Conventional JCI concrete core tests failed to detect deleterious expansion in the ASR-affected Okinawan concretes. The reason for this is that the expansion limits of this test were based on early-expansive andesite aggregates in other areas of Japan, where structures showed cracking within several years of construction. However, our data in the present study indicate that, at age of 19 years, late-expansive ASR in Arakaki bridge produced comparable deterioration but with a small expansion rate (field severity IV; crack density: >0.2mm: 3-5m/m²). This suggests that core specimens will also produce slow and small expansion in this test, thus escaping being judged as deleterious. Therefore, the owner of the structures should not falsely believe that deleterious ASR is absent or has deceased in the structures.

4.2 The type of AAR in this study

Late-expansive ASR and early expansive ASR

Sea-dredged sands from Okinawa produced fine cracks in concrete (width <0.2mm) due to lateexpansive ASR in the northern structures, where other reactive aggregates were absent. Since they produced larger expansion in the accelerated concrete core test than in the core specimens from southern structures with the Taiwanese sands, they have a large potential for developing further ASR.

Cristobalite is one of the highly reactive silica minerals causing early-expansive ASR with a small pessimum proportion (<10% [6]). Although the concrete contained less than the pessimum proportion of andesite with this mineral, this probably contributed to the cracking of southern structures, because cracks associated with this andesite in concrete were the widest. The andesite was crystalline and volcanic interstitial glass was absent as a result of geological alteration to smectite through diagenesis. Selective quarrying of the limestone has been removing this andesite dike at the limestone quarry, since the discovery of its ASR by the second author in a bridge in another area of Okinawa in around 2003 [5].

Dedolomitization and sweetening effect

Dedolomitization has been known in the so-called "alkali-carbonate reaction" in Ontario, Canada, in which brucite forms in the dolomitic aggregate without producing expansion cracks in the surrounding cement paste [4]. This is also the case in Okinawa, and cracking of concrete was due to ASR of cryptocrystalline quartz in the limestone, indicating that dedolomitization is a harmless reaction. Mg-silicate gel formed after crypto- to microcrystalline quartz in the dolomitic aggregate suggests that brucite had reacted with ASR gel around quartz. Hydrotalcite found on the periphery of dolomitic aggregate may be gel-like, since it had a low analytical total of 45wt%, while the crystal phase exclusive of CO_2 and H_2O , should be 56.9wt%. This material is readily synthesized at ambient temperature in the chemical industry.

The crushed limestone fine aggregate did not produce ASR in the northern structures where cracking was minor. It is interesting to note that, in Kansas and Nebraska, USA, the addition of crushed limestone coarse aggregate, called "limestone sweetening", has been successfully applied to suppress ASR of the local sand-gravel aggregates, and limestone fine aggregate was more effective in Iowa [7]. It should be worthwhile to examine if the Okinawan limestone fine aggregate has more sweetening effect on ASR of the sea-dredged sands than simple dilution, based on petrographic comparisons of northern structures.

4.3 Implications of composition of ASR gel

ASR gel migrates from the reacted aggregate into cement paste, releasing alkalies and taking up calcium from CSH gel, and transforms compositions into CSH gel toward a "convergent point" at around [Ca/Si] = 1.5, [Ca]/[Na+K] = 100, where a chemical equilibrium is attained between ASR gel and CSH gel. The narrow, higher alkali range of the crack-filling ASR gel in Noborimata (Fig.4 C) is indicative of the most expansive nature of ASR gel among the structures examined, explaining the largest expansion obtained in the JCI-DD2 test. The wide compositional range of ASR gel in Kitanakagusuku (Fig.4 D) suggests that ASR of the Okinawan sea-dredged sand has not deceased yet in this concrete.

At the point of [Ca/Si] = 1/6, [Ca]/[Na+K] = 1.0, ASR gel corresponds to 0.5(Na, K)₂OCaO 6SiO₂nH₂O, and if Na₂O (or K₂O) is replaceable with CaO, keeping the cation valent (2Na=Ca), this is then equivalent to makatite Na₂O4SiO₂5H₂O or kanemite Na₂O4SiO₂7H₂O. If cation numbers are kept constant (Na=Ca), then they correspond to okenite CaO2SiO₂2H₂O. It will be worth examining further to clarify the nature of the ASR gel and relationships between these silicates, by means of micro-XRD.

4.4 Alkali-threshold of concrete

The contribution of water-soluble alkali from coarse and fine aggregates in concrete (Na₂Oeq 0.1-1.1 kg/m³) should not be neglected. Of these, the water-soluble alkali of the extracted coarse aggregate could contain absorbed alkali from cement paste, and that of the fine aggregate is subject to error by a higher alkali sulfate ratio of the cement clinker than expected (i.e. total alkali/min alkali > 1.3) plus introduced alkali from the chemical admixture (Table 5). In Kitanakagusuku, the highest value of water-soluble alkali of fine aggregate may be due to reacted slate particles abundantly contained in concrete.

The cracking of concrete occurred with total available alkali down to Na₂Oeq 2.2 kg/m³, i.e. cement alkali Na₂Oeq 2.0 kg/m³ (Table 5), far below the common alkali threshold of 3.0 kg/m³. Highly reactive andesite with a pessimum phenomenon may have contributed to this, but this was not always found in concrete. It is noteworthy that, in such a low alkali condition, both the sand and the crushed limestone coarse aggregate produced late-expansive ASR of cryptocrystalline quartz.

Similar crushed limestone coarse aggregate, capable of causing late-expansive ASR due to cryptocrystalline quartz, has been known in Canada (Spratt aggregate), which developed cracks in concrete with low alkali cement (Na₂Oeq 0.46%) and total concrete alkali Na₂Oeq 1.9 kg/m³, after 12 years of exposure in Kingston [8]. In Okinawa, where freeze-thaw cycle is absent, sub-tropical climate with higher average annual temperature and precipitation (Naha 23°C, 2000mm) most probably accelerated the lateexpansive ASR than in other areas of the world where this reaction has been known (e.g. Halifax 7°C, 990mm; Kingston 7°C, 800mm). Thus, the currently accepted alkali threshold for causing ASR in Japan (Na₂Oeq 3.0 kg/m³; JIS A5308) needs to be reviewed for local situations in Okinawa.

5 CONCLUSIONS

It follows from the above discussion that the methodology of concrete petrography, described by the first author in RILEM AAR-6 part 1 [2], is suitable for the diagnosis of late-expansive ASR and dedolomitization in concrete, particularly in Okinawa.

- They are: 1) core scanning, 2) modal analysis of coarse and fine aggregates, 3) stereomicroscopy to identify ASR sol/gel, and, using the same polished thin section, 4) polarizing microscopy, 5) SEM observation and 6) EPMA analysis of reaction products to evaluate the evolutionary trend of ASR, and 7) EPMA analysis of unhydrated cement phases to estimate the minimum alkali content in the cement used. Measurements of 8) crack density, and of 9) water-soluble alkali to examine the alkali-budgets in concrete, and 10) accelerated concrete core expansion test to assess potential reactivity of aggregate, are also useful.
- The southern section of the southern Okinawa Expressway is undergoing moderate degrees of cracking, due to late-expansive ASR of cryptocrystalline quartz contained in the river sands imported from Taiwan and in the crushed limestone coarse aggregate from Okinawa, and by early-expansive ASR of cristobalite in andesite that accompanied the limestone aggregate. Dedolomitization was harmless.
- The northern section of the southern Okinawa Expressway presented no problem with the limestone coarse aggregate, but the sea-dredged sands from Okinawa, with broadly similar lithologies to those of the Taiwanese sands, produced fine cracks due to late-expansive ASR of cryptocrystalline quartz.
- Late-expansive ASR is taking place with total available concrete alkali, even less than Na₂Oeq 2.5kg/m³. This underlines the need for reviewing the commonly accepted alkali threshold of 3kg/m³ in Japan, to accommodate with the sub-tropical conditions in Okinawa which are likely to accelerate ASR.

6 ACKNOWLEDGEMENTS

This research has been performed in parallel with a technical committee chaired by the second author Dr. Takeshi Oshiro, Prof. Emeritus of University of Ryukyus. The authors are grateful to Mr. Hideto Imuta, deputy director of Okinawa Operation Office, West Nippon Expressway Co. Ltd. for his managing support in this project. Special thanks go to the following governmental organizations for providing the authors with access to unpublished reports on AAR in Okinawa: the Civil Engineering & Construction Division of Miyako Branch, and the Department of Civil Engineering & Construction, both of Okinawa Prefecture; the Naha Ports & Airport Office, and the Road Management Division, both of Development Construction Department, the Okinawa General Bureau, Cabinet Office of Japan.

7 REFERENCES

[1] Investigation of the current state of structures in Okinawa Expressway between Naha and Ishikawa. Okinawa Operation Office, West Nippon Expressway Co., Ltd. March, 2007: pp180, +

appendix report of Technical Committee on AAR in Okinawa Expressway (in Japanese).

- [2] RILEM AAR-6. Guide to diagnosis and appraisal of AAR damage to concrete in structures, part 1: diagnosis, 5th draft AAR-6, July, 2006: pp57 + appendices.
- [3] Katayama T, Tagami M, Sarai Y, Izumi S, Hira T (2004): Alkali-aggregate reaction under the influence of deicing salts in the Hokuriku district, Japan. Materials Characterization, Vol. 53, 2-4, Special Issue 29: 105-122.
- [4] Katayama T (2004): How to identify carbonate rock reactions in concrete. Materials Characterization, Vol.53, 2-4, Special Issue 29: 85-104.
- [5] Report of committee on the concrete durability for the construction of Irabu bridge (2006): Civil Engineering & Construction Division, Miyako Branch, Okinawa Prefecture, Japan (in Japanese).
- [6] Katayama T, Helgason TS, Olafsson, H (1996): Petrography and alkali-reactivity of some volcanic aggregates from Iceland. Proceedings, 10th International Conference on Alkali-Aggregate Reaction in Concrete. Melbourne, Australia: 377-384.
- [7] Hadley DW (1968): Field and laboratory studies on the reactivity of sand-gravel aggregates. Journal of the PCA Research and Development Laboratories, January: 17-33.
- [8] Hooton D, Rogers C, Ramlochan T (2006): The Kingston outdoor exposure site for ASR after 14 years what have we learned? Marc-André Bérubé Symposium on Alkali-Aggregate Reactivity in Concrete, Montreal, Canada: 171-193.

				So	uthern sec	tion			Norther	n section	
Reactio		Structures	Arakaki	Arakaki	Nobo r i- mata	Adaniya	Futenm a-gawa	Kitanaka -gusuku	Kitanak a- gusuku	Uechi	Matsu- moto
n			Bridge	Ret.wall	Viaduct	Viaduct	Bridge	Viaduct	Viaduct	Bridge	Bridge
	Ro	ck types	Abut- ment	Retaining wall	Abut- ment	Abut- ment	Abut- ment	Column cracked	Column no crack	Abut- ment	Pier
Dedolo-	С	Dolomitic ls.	XX	х	XX	х	х	х	х		
itization	F	Dolostone, dol.ls	х	XX	XX	х	XX		х	х	x
ASR		Dolomitic ls.		x 2-3							
	С	Limestone	x 2-3	x 2-3	x 2-3	x 2-3					
		Andesite	xx 1-4	xx 1-4		xx 1-4					
	F	Andesite		x 1					x 1-3		
		Sandstone			x 1-3		x 1	x 1	x 1	x 1	x 1-3
		Mudstone, shale		x 1	x 1-3	x 1	x 1-3	x 1-3	x 1	x 1	
11010		Siliceous shale	x 1-2	x 1-3		x 1-2	x 1-2	x 1-3	x 1	x 1	x 1
		Chert	x 1-3	x 1	x 1	x 1	x 1-3	x 1	x 1	x 1-3	x 1-2
		Slate, phyllite	x 1-3	x 1	x 1-3	x 1	x 1-3	x 1-3	x 1-3	x 1-3	x 2-3
		Meta-sil.shale								x 1	x 1-3
		Meta-chert	x 1-3	x 1-3	x 1	x 1-3	x 1			x 1	
		Quartzite	x 1-3	x 1-3	x 1-3	x 1-2	x 1	x 1-3	x 1-2	x 1	x 1-4
Severity	<i>v</i> of	Petrographic	4	3	2	3	2	2	1	2	2
ASR		Field	IV	III	III	III	II	II	Ι	II	II
Intensit Progress Petrogra	y of 1 s of 1 aphic	AAR: xx conspicuou ASR: 1 reaction rim 4 gel-fill voids severity of ASR: 1 t	s, x preser & gel-exuc in cement raceable, 2	nt C: coa lation, 2 ge paste 2 minor, 3 r	rse aggrega l-fill cracks noderate, 4	ate, F: fine a s in aggrega 4 severe	aggregate te, 3 gel-fil	l cracks in	cement pas	ste,	

 TABLE 1: Intensity and stage of AAR in the southern half of the Okinawa Expressway with reference to the reacted rock types of the aggregate in concrete as determined by thin section petrography.

TABLE 2: The type of AAR and reacted minerals in the aggregate of concrete in the Okinawa Expressway as determined by thin section petrography.

Structure	Aggi	regate	Reacted rock type	Туре	of reaction	acted mineral	Crack
	Faiwan	River	Mudstone, shale, sandstone, siliceous shale, chert, slate, phyllite, meta-chert, quartzite	ASR	Late-expansive	Crypto- crystalline quartz	Fine
Southern section		sanu	Dolomitic limestone, dolostone	Dedolomiti- zation	Harmless	Dolomite	None
			Andesite	ASR	Early-expansive	Cristobalite	Wide
		Crushed limestone	Limestone (argillaceous)	ASR	Late-expansive	Crypto- crystalline quartz	Medium
Northern section	Dkinawa	coarse	Dolomitic limestone	Dedolomiti- zation	Harmless	Dolomite	None
		Sea- dredged sand	Mudstone, shale, sandstone, siliceous shale, chert, slate, phyllite, meta-chert, quartzite	ASR	Late-expansive	Crypto- crystalline quartz	Fine

TABLE 3: Selected compositions of ASR gel in the fine aggregates in concrete as determined by EPMA (EDS) analysis.

\setminus	Arakaki #			N	oborimata	#	Kit	anakagusul	Matsumoto+		
	Qua	rtzite	Sil.sh.	Qua	rtzite	Slate	Qzt.	Sla	ate	Chert	Slate
	ASR	ASR	ASR	ASR	ASR	ASR	ASR	Crypt.	ASR	ASR	ASR
	gel*	gel**	gel**	gel*	gel**	gel**	gel*	quartz	gel*	gel**	gel**
SiO ₂	46.28	46.02	39.19	42.92	44.82	47.95	39.68	97.30	51.04	46.03	37.95
Al ₂ O ₃	0.15	0.18	3.07	1.06	0.25	2.56	1.50	1.31	1.22	0.48	2.57
Fe ₂ O ₃	0.00	0.00	1.36	0.48	0.00	0.00	1.63	0.55	3.12	0.42	1.47
MgO	0.00	0.00	0.00	0.00	0.00	0.00	0.39	0.00	0.26	0.00	0.30
CaO	18.73	7.23	7.70	19.74	9.41	9.29	17.50	0.04	5.72	10.77	13.41
Na ₂ O	1.13	2.40	1.61	2.90	1.74	2.40	0.65	0.07	0.22	1.05	0.81
K ₂ O	3.62	6.18	7.84	3.73	6.96	6.15	3.15	0.06	3.50	7.00	5.04
SO3	0.00	0.00	0.00	0.10	0.00	0.06	0.00	0.00	0.05	0.19	0.08
Total	69.91	61.99	60.76	70.93	63.18	68.41	64.51	99.35	65.14	65.94	61.62
1	0.43	0.17	0.21	0.49	0.23	0.21	0.47		0.12	0.25	0.38
2	2.95	0.62	0.63	2.04	0.82	0.80	3.55		1.25	1.05	1.80
#: River	sand from	Taiwan, +:	Sea-dredge	ed sand fro	m Okinawa	a, *: Am	orphous, *	*: Rosette-l	ike		
1. Atomi	c ratio[Ca]	/ISil 2: Atc	mic ratio	Cal/(Nal+	-IKI)						

Ν	Ara	kaki	Arakaki retaining wall										
\backslash	Lime	stone		Limestone				Dolomitic	limestone#	ŧ			
\backslash	Crypt.	ASR	Crypt.	ASR	ASR	ASR	ASR	Mg-	Secd.	Brucite	Hydro		
	quartz	gel*	quartz	gel*	gel*	gel*	gel*	sil.gel	calcite		-		
	1	0		Ö	0	0	Ŭ	Ŭ			talcite		
											#		
SiO ₂	97.81	57.19	98.40	50.57	45.58	29.95	27.00	28.95	0.23	5.22	0.11		
Al ₂ O ₃	0.21	0.21	0.38	0.50	0.47	1.34	0.39	0.00	0.00	0.00	13.13		
Fe ₂ O ₃	0.36	0.07	0.00	0.47	0.56	0.91	0.31	0.07	0.00	0.00	0.45		
MgO	0.00	0.00	0.00	0.00	0.00	0.01	0.00	24.59	2.02	59.31	<u>28.82</u>		
CaO	0.18	10.01	0.27	9.81	18.34	34.52	29.93	13.78	52.20	2.66	2.22		
Na ₂ O	0.00	0.97	0.19	0.75	2.00	0.17	0.24	0.53	0.25	0.71	0.32		
K ₂ O	0.16	4.39	0.00	5.49	4.77	0.11	0.82	0.22	0.13	0.00	0.05		
SO3	0.00	0.00	0.40	0.03	0.13	0.00	0.00	0.00	0.05	0.02	0.13		
Total	98.73	72.84	99.63	67.63	71.85	67.00	58.85	68.14	54.88	67.94	45.23		
1		0.19		0.21	0.43	1.23	1.19						
2		1.43		1.24	1.97	77.43	17.61						
\backslash	l	Noborimat	a	Adaniya Limestone				Arakaki	Ada	iniya			
		Limestone						Andesite+	And		esite+		
	Crypt.	ASR	ASR	Crypt.	ASR	ASR	Cristo-	ASR	ASR	Cristo-	ASR		
	quartz	gel*	gel**	quartz	gel*	gel**	balite	gel*	gel**	balite	gel**		
SiO ₂	96.78	43.56	41.06	96.94	44.37	42.70	95.90	50.96	52.58	98.39	51.21		
Al ₂ O ₃	0.77	0.62	0.34	0.62	0.00	0.16	1.82	0.53	0.28	0.63	0.23		
Fe ₂ O ₃	0.00	0.00	0.36	0.00	0.42	0.33	0.00	0.28	0.07	0.33	0.58		
MgO	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
CaO	0.45	11.69	13.49	0.98	23.19	10.35	0.35	14.65	11.73	0.09	10.49		
Na ₂ O	0.03	2.16	0.92	0.25	1.45	0.97	0.47	3.52	1.49	0.72	1.94		
K ₂ O	0.32	4.13	6.36	0.41	2.97	7.16	0.63	6.15	9.24	0.09	7.77		
SO3	0.09	0.17	0.00	0.00	0.00	0.00	0.05	0.30	0.00	0.00	0.04		
Total	98.44	62.33	62.53	99.19	72.40	61.78	99.23	76.40	75.38	100.26	72.27		
1		0.29	0.35		0.56	0.26		0.31	0.22		0.22		
2		1.33	1.46		3.76	1.01		1.07	0.86		0.82		
#: Accor	npanying tl	he limestor	ne, +: Deriv	ved from a	dike in the	limestone	quarry, *:	Amorpho	us, **: Ros	ette-like			
1. Atomic ratio [24]/[S1] 2. Atomic ratio [Ca]/[Na]+[K]													

TABLE 4: Selected compositions of ASR gel and dedolomitization products in the crushed coarse aggregates in concrete as determined by EPMA (EDS) analysis.

TABLE 5: Alkali-budgets of concrete as estimated from EPMA (EDS) analysis of unhydrated cement and wet chemical analysis of water-soluble alkali of concrete and separated coarse aggregate.

\setminus	Clinker Cement						Con	crete		Coarse aggregate (extr.)			Others
\setminus	Mi	Minimum alkali Total Water- by EPMA alkali sol.alk			Wate	er-soluble	alkali	Total alkali	Wate	Total alkali			
$ \rangle$		measured	l	estin	nated	measured est.				est.			
$ \rangle$	%			Na ₂ Oeq	kg/m ³	0	6	Na ₂ Oeq	kg/m ³	0	/0	Na ₂ Oeq	kg/m ³
	Na ₂ O	K ₂ O	(1)	(2)	(3)	Na ₂ O	K ₂ O	(4)	(5)	Na ₂ O	K ₂ O	(6)	(7)
1	0.54	0.18	0.66	2.69	1.61	0.063	0.049	2.19	3.27	0.028	0.025	0.36	0.22
2	0.52	0.25	0.68	2.04#	1.22	0.039	0.034	1.40	2.22	0.019	0.017	0.21	-0.03
3	0.43	0.16	0.53*	2.16	1.30	0.042	0.035	1.50	2.36	0.015	0.013	0.20	0.00
4	0.46	0.26	0.63	2.57	1.54	0.065	0.042	2.14	3.17	0.038	0.036	0.47	0.13
5	0.44	0.19	0.57	2.33	1.40	0.074	0.054	2.53	3.46	0.030	0.023	0.40	0.73
6	0.50	0.22	0.64*	2.62	1.57	0.066	0.058	2.39	3.44	0.011	0.011	0.14	0.68
7	0.41	0.14	0.50	2.04	1.22	0.038	0.030	1.33	2.15	0.007	0.006	0.07	0.04
8	0.41	0.17	0.53	2.16	1.30	0.058	0.049	2.07	2.90	0.011	0.012	0.16	0.61

1 Arakaki, 2 Arakaki retaining wall, 3 Noborimata, 4 Adaniya, 5 Futenmagawa, 6 Kitanakagusuku, 7 Uechi, 8 Matsumoto (1): Na₂Oeq=Na₂O+0.658K₂O * including interstitial glass (2): $1.2 \times ((1)/100) \times 340 \text{ kg/m}^3$, 1.2 = 1.3 (total alkali/min alkali) $\times 0.97$ (dilution by gypsum) $\times 0.96$ (dilution by additives)

(3): 0.6 x (2), 0.6= water-soluble alkali ratio, (4): as 2300 kg/m^3 , # as 250 kg/m^3 (5): Total available alkali of concrete = (2) + (6) + (7) = (2) + (4) - (3)

(6): Water-soluble alkali of separated coarse aggregate, including absorbed alkali from cement paste

(7): Water-soluble alkali of fine aggregate plus chemical admixture = (4) - (3) - (6), subject to error by absorbed alkali in the

coarse aggregate (6), or a higher alkali sulfate ratio of cement clinker (total alkali/min alkali), e.g.. > 1.3 Total available alkali from coarse and fine aggregates plus chemical admixture = (6) + (7) = (4) - (3)



Figure 1: Reacted crushed limestone (A) with ASR gel from cryptocrystalline quartz (B). Dolomitic limestone with gel-filled crack (C, D), forming spots of brucite, calcite (D, E) and hydrotalcite (F) on the periphery through dedolomitization. Andesite with gel-filled cracks from reacted cristobalite inside (G H). A-H: Arakaki retaining wall. Reacted slate (phyllite) in Taiwan sand (I Futenmagawa) and in Okinawa sand with crystallized ASR gel (J, K: Kitanakagusuku). Reacted quartzite: rosette-type ASR gel in Taiwan sand (L, M: Noborimata) and amorphous ASR gel in Okinawa sand (N, O: Kitanakagusuku).



Figure 2: Crack density of concrete structures Figure 3: Compositions of fine aggregates in concrete. * ASR-reacted rock types



Figure 4: Compositional trends of ASR gel and CSH gel in concrete as determined by EPMA (EDS): A) Arakaki, B) Arakaki retaining wall, C) Noborimata, and D) Kitanakagusuku



Figure 5: Concrete core expansion tests: A) JCI-DD2 in humid air, B) acceleration in NaOH solution