Committee Report : JCI- TC211A

Technical Committee on Assessment and Prediction of Expansion due to Internal Swelling Reactions in Concrete Structures

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Abstract

This committee focused on the internal swelling reactions (ISRs) of concrete, such as alkali-silica reactions (ASRs), delayed ettringite formation (DEF), and oxidation of iron sulfide-containing aggregates. The goal of this committee is to propose appropriate assessment and prediction methods for risk assessment of ISRs-affected structures. We discussed the basic theory of the swelling reaction and presented the state-of-the-art report and future prospects of a structural scale analysis and modeling based on microscopic mechanisms, mainly with respect to ASRs. In addition, we investigated and summarized the accelerated test methods and analysis methods necessary for understanding the mechanism of DEF and for risk assessment. Furthermore, the risk of swelling due to the oxidation of iron sulfide-containing aggregates, which is not widely recognized in Japan, was investigated.

Keywords: Alkali-silica reaction, delayed ettringite formation, iron-sulfide aggregate, internal swelling

1. Introduction

The various risks of deterioration due to the internal swelling reactions (ISRs) of concrete, such as an alkali-silica reaction (ASR), delayed ettringite formation (DEF), and ettringite formation due to oxidation of iron sulfide minerals in aggregates, is attracting attention both in Japan and other countries. However, appropriate assessment and prediction methods for these swelling phenomena is still under investigation. This committee has reconsidered the basic theory of swelling mechanism that is common to ISR, examined the ideal direction of the test methods for ISR and the model of materials and structures, and conducted research with the aim of presenting a more effective and feasible ISR risk assessment method. In particular, we investigated the microscopic mechanisms of the ASR and DEF and the interaction between microscopic-swelling pressure generation processes and macroscopic deterioration processes of structures; organized the numerical analysis model, accelerated test method, deterioration process, and crack analysis method for the assessment and prediction of internal swelling in concrete structures based on microscopic mechanisms; organized the deterioration mechanism by iron-sulfide-containing aggregates; and used a geological approach to examine the risk of occurrence in Japan.

Table 1. Committee members

Chairman of Committee	Yuichiro KAWABATA	Port and Airport Research Institute
Secretary of Committee	Yuya TAKAHASHI	University of Tokyo
Secretary of Committee	Yasutaka SAGAWA	Kyushu University
Secretary of Committee	Taito MIURA	Nagoya University
Secretary of Committee	Shintaro MIYAMOTO	Tohoku University
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	Daisuke YAMAMOTO	National Institute of Technology, Oita College
	Go IGARASHI	Nagoya University
	Hiroki SAKURABA	Public Works Research Institute
	Ippei MARUYAMA	University of Tokyo
	Kazuo YAMADA	National Institute for Environmental Studies
	Kennosuke SATO	University of Yamanashi
	Naoshi UEDA	Kansai University
	Ryo KURIHARA	University of Tokyo
	Shingo ASAMOTO	Saitama University
	Takashi KAWAKAMI	Kyushu University
	Tomonori SEKI	Sumitomo Osaka Cement
	Yoko ANDO	Taiheiyo Consultant Co. Ltd.
	Yoshifumi OHGI	Taiheiyo Cement
	Yoshihiko TODA	JIP Techno Science
	Yukiko NISHIOKA	Takenaka Corporation

Table 1 lists the members of the committee. The committee consists of four Working Groups. In the Basic Theory WG (WG1), the basic theory of the ISR was discussed with reference to basic theories in other fields. In the Modeling WG (WG2), we discussed the ideal numerical analysis model based on the microscopic mechanism of the ASR. With a focus on the interaction between microscopic mechanisms and macroscopic behavior, the microscopic mechanisms that should be focused on and the importance of considering them are summarized

in assessing the structural responses of ASR-affected structures. In the Test and Analysis WG (WG3), especially in the case of DEF, we discussed the ideal direction of expansion tests, implemented interlaboratory tests, and summarized the significance and purpose of testing and equipment analysis. In the iron-sulfide-containing aggregate WG (WG4), we referred to overseas cases of swelling due to the oxidation of aggregates containing iron sulfide, in addition to summarizing the possible mechanisms, and examined methods for assessing potential risks.

2. Direction of modeling based on microscopic mechanism

In the case of WG1 and WG2, mutual relationships between the swelling, cracking, and changes in the mechanical properties common to concrete swelling deterioration were discussed based on the characteristics caused by its anisotropy. Based on the results of the literature surveys conducted globally, we reviewed the structural analysis model that introduced the swelling anisotropy model and discussed the necessity of modeling the swelling anisotropy from a microscopic perspective with respect to the performance assessment of structures that undergo internal swelling deterioration. As an example of a literature research, Fig. 1 presents the statistical results of the analytical research aimed at the structural performance assessment of concrete that has undergone ASR expansion. Only a limited number of countries have developed analytical methods capable of assessing the mechanical behavior of ASR-deteriorated structural members, and it was found that Japan has a high proportion of these methods. For detailed information regarding these methods, the readers are recommended to refer to the review paper[1)].

2.1 Discussion on modeling of swelling deterioration

With respect to ASR and DEF, within the committee, we sorted out topics and discussed numerical analyses that introduced deterioration models and discussed the modeling necessary for realizing a swelling deterioration analysis. The target numerical analyses are as follows: (1) a structural analysis with the models such as the ASR expansion anisotropy proposed by IFSTTAR (currently Université Gustave Eiffel)[2)] into the general-purpose finite element method (FEM); (2) a loading analysis of prestressed concrete (PC) beam members deteriorated by ASR and DEF, wherein a simple swelling model was applied to the general-purpose FEM; and (3) a swelling analysis of the beams and deck slabs modeled from a microscopic perspective while considering ASR, which is being developed by the University of Tokyo[3)]. From the discussion with the use of these numerical analyses, it was confirmed that some phenomena are difficult to be reproduced by the currently proposed analytical methods. In particular, we shared the importance of modeling while taking into consideration the microscopic viewpoint of the formation process of ASR gels in precisely assessing and predicting changes in the swelling over time. Moreover, we confirmed that reproducing changes in the stiffness and strength of members with the general constitutive laws required modeling of not only the swelling anisotropy under the constraint conditions, but also the anisotropy of the mechanical property change due to the orientation of the cracks caused by the constraint.

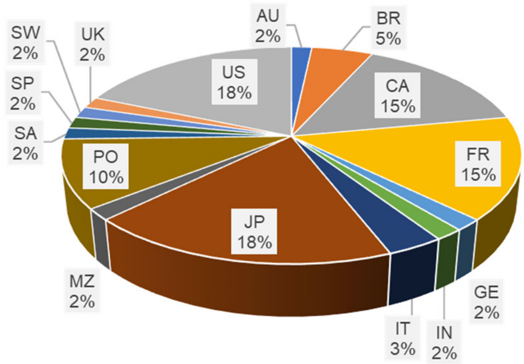

Fig. 1: Country count results of research institutions to which authors are affiliated[1)]

2.2 Necessity of microscopic modeling

To predict the future change in performance of ASR-deteriorated structures, cores are generally obtained from the structures and accelerated tests are performed to determine the residual swelling potential of the concretes.

When conducting analytical studies, especially in macroscale models, it is common to perform a structural analysis using the free expansion obtained in the laboratory concrete expansion test as an input data. However, the validity of the performance assessment/prediction using free expansion is unclear. For example, the core has smaller dimensions than the structure and is more sensitive to boundary conditions during acceleration test. In laboratory tests, when the core comes into contact with water, the alkali contained in the core is leached out[4)]. As a result, the expansion converges early, and the residual expansion may be determined to be small. In contrast, in structures, especially in the central part of the structure, a sufficient amount of alkali may exist, and the swelling may continue. When calculating the amount of swelling using the amount of reaction products from the chemical reaction process between reactive minerals and alkali ions, if the effects of alkali leaching are not taken into consideration, the expansion that can occur in real structures may be underestimated.

In this manner, it is considered possible to appropriately assess and predict the potential amount of concrete expansion in structures by considering microscopic viewpoints such as the effects of alkali leaching that may occur at the structure core and specimen scale.

2.3 Necessity of modeling anisotropic expansion

When assessing the mechanical properties (strength, elastic modulus, etc.) of concrete in ASR-deteriorated structures, there are cases wherein cores are obtained from the structure, and performance assessment is performed using the physical properties as input values. However, constrained ASR expansion exhibits anisotropy, and the distribution of cracks is strongly affected by the constraint conditions[5), 6)]. Therefore, even if the core is obtained from one direction of the structure for assessing the physical properties, the meaning of the physical properties varies greatly depending on the constraint conditions[7), 8)]. At present, there is few data on the effect of the constraint conditions on the physical properties of concrete with ASR deterioration, and further research is necessary.

2.4 Mutual relationship between expansion, cracking, and mechanical properties

Two major problems are encountered in understanding and modeling the expansion anisotropy of ASR and DEF. One is that the mechanism of expansion transfer (expansion redistribution) is unclear, and the other is that there is very little experimental data on the anisotropy of mechanical properties under constraint.

The former is frequently discussed in the modeling of ASR expansion, and researchers are divided on whether expansion transfer occurs, wherein the expansion in the direction orthogonal to the constraint is greater than the free expansion, and the cause of its occurrence remains unclear[5), 6)]. This is related to whether the model of expansion anisotropy is considered in the structural analysis. Therefore, it is necessary to understand whether expansion transfer occurs in the concrete used to predict the future expansion of ASR-deteriorated structures. A recent study conducted by Miura et al.[9)] clarified that the formation mechanism of sharp cracks and onion-skin cracks observed in an aggregate depends on the distribution of the expansive sites in the aggregate and also mentioned the possibility that the distribution of this expansive site differs depending on the rock type of the aggregate (Fig. 2). Furthermore, the results suggest that the susceptibility to expansion transfer depends on the crack pattern inside the aggregate[10)].

Regarding the latter, the anisotropy of the mechanical properties under the constraint can be confirmed when the crack propagates in the direction parallel to the constraint. For example, if an ASR occurs in a PC structure, the cracks propagate in the longitudinal direction owing to the prestress force applied to the longitudinal direction. In this case, the deterioration of the mechanical properties in the bridge axis direction (parallel to the cracks) is small, but the mechanical properties in the direction perpendicular to the cracks are considered to decrease significantly. At present, experimental data on ASR-deteriorated concrete have been published by Hansen et al.[7] and Anca et al.[8], but the data are still limited. Furthermore, at present, sufficient experimental data to discuss the anisotropic mechanism of the deterioration of mechanical properties have not been available.

Internal crack information is important for expansion transfer and mechanical property anisotropy. Therefore, it may be possible to solve these issues related to expansion anisotropy by identifying quantitative indices based on the crack information (width, density, angle, etc.) inside aggregates and in mortar. Therefore, the method of extracting crack information from inside concrete is important. In the future, it is expected that experimental data will be expanded to elucidate the mutual relationship between the anisotropy of expansion under restraint; crack area (aggregate, cement paste), distribution, density, and direction; and the anisotropy of mechanical properties.

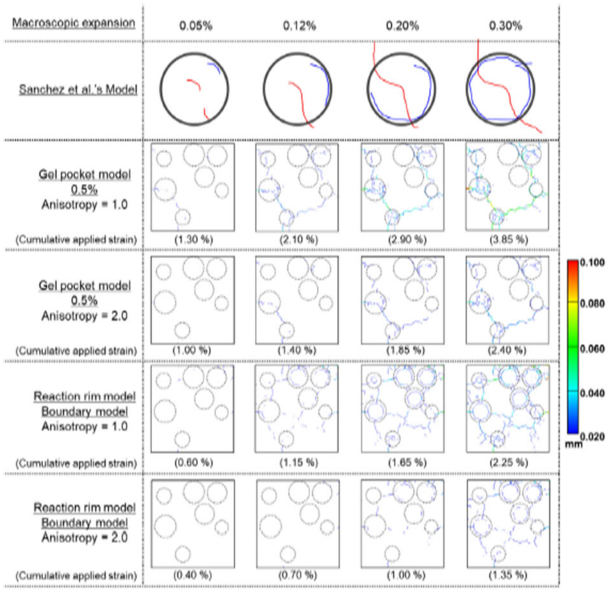

Fig. 2: Crack propagation in each expansive site model[9]

3. Examination of accelerated test method for internal swelling reaction

While ISR risk is attracting attention globally, the required tests and assessment methods are yet to be standardized. Especially, in Japan, no case of DEF-affected structure has been reported whist laboratory tests show that the mortar/concrete using Japanese cement show large expansion. Therefore, in WG3, we focused on DEF in the ISR and used test methods for understanding the mechanism of DEF and risk assessment, in addition to assessment and diagnosis methods based on various analytical methods. We conducted a literature survey on accelerated tests in the global research on DEF, proposed an accelerated DEF test method, and started a common test.

3.1 DEF acceleration test method

The French Public Works Research Laboratory, LCPC (currently Université Gustave Eiffel) has proposed the LPC Test method No.66 as a method for conducting the swelling test of concrete specimens[11)]. A test specimen fabricated from concrete with a job mix is subjected to high-temperature curing while simulating the actual environment, followed by two cycles of water at 20 °C for 7 days and 38 °C at RH30% for 7 days, and then immersion in water at 20 °C for at least 12 months. In the high-temperature curing, the expected thermal history of concrete should be simulated and thus the specific temperature and duration are undefined.

The proposal for the LPC Test method No. 66 is based on the studies of Duggan et al.[12)] and Pavoine et al.[13), 14)]. In the "Duggan test", a 3 × 3 × 14 inch prism specimen is prepared, and it is subjected to precuring for 2 h, followed by accelerated curing (85 °C, 4 h). A core of 22 mm diameter and 50±5 mm height is then obtained from the prism specimen, and the change in the length of the specimen is measured. After curing in water for 3 days and repeating the heating and cooling at 82 °C and 21 °C twice, the change in the length of the specimen in distilled water is measured. The cycle of heating and cooling is used to generate microcracks in the specimen, which facilitates the supply of water into the concrete, which is believed to promote the DEF. In a previous study[12)], tests were conducted under the condition that the water–cement ratio (W/C) was 0.40, while changing the type of cement, type of aggregate, and the SO_3/Al_2O_3 ratio; it has been reported that the swelling reaches a maximum when the SO_3/Al_2O_3 ratio is approximately 1.0.

In addition, Pavoine et al.[13), 14)], with references to Duggan test and similar tests, proposed the method used in the LPC Test method No. 66. Dry–wet cycles have the effect of locally increasing the saturation of ettringite and of increasing the occurrence of microcracks or increasing the width of cracks and facilitating moisture migration. In reference[13)], the Taguchi method was used to optimize the curing method after performing heat curing at 90 °C for 10 h, with parameters such as the time until the start of repeated drying and wetting, period of immersion in water, cycle, and number of repeated drying and wetting cycles, drying temperature, and type of immersion solution. Based on the results, Method No.66 was proposed. It has also been reported that the nature of immersing solution (tap water, saturated $Ca(OH)_2$) plays a secondary role. In addition, in reference[14)], verification experiments were conducted on concrete used for bridges and concrete for precast members using the above proposed method. As a result, when the bridge concrete was subjected to the same temperature history as the actual member (maximum of approximately 80 °C), for the mix of W/C = 0.47, DEF index = 0.88, and SO_3/Al_2O_3 = 0.92) showed approximately 0.10% of the swelling at 170 days whilst another mix of W/C = 0.54, DEF index = 1.07, and SO_3/Al_2O_3 = 1.03 exhibited severe swelling (1.5%) at 130 days. In contrast, PC concrete did not show any swelling (maximum swelling of approximately 0.01–0.02%) when it was subjected to a temperature history of 80 °C for 1 h. This is because the concrete was subjected to less heating as it was not manufactured in the summer and the member had a shape which promoted heat loss.

Paris et al.[15)] compared three methods: (a) preheating (23 °C) for 4 h, heating at 95 °C and RH 95% for 12 h, and then soaking in saturated $Ca(OH)_2$ solution, (b) heating for 36 h, and (c) preheating for a short duration of 1 h with an added step of heating to 85 °C. As a result, it was experimentally demonstrated that method (b) accelerates the initiation and extent of damage caused by DEF in mortar.

In countries other than Japan, the DEF index or SO_3/Al_2O_3 of cement is originally high and is promoted by repeating hot and cold cycles after high temperature curing or by repeating dry and wet cycles; however, in Japan, the combination of K_2SO_4 addition and high-temperature curing is the primarily used method. In both cases, DEF occurs, but the physical and chemical phenomena occurring inside the mortar or concrete may be different. Further research is required to determine whether the assessment can be based solely on the swelling rate and whether accelerated testing can provide the data necessary for understanding and modeling the reaction mechanism.

3.2 Laboratory test

The accelerated test of DEF is considered to have the following two significances. One is to determine whether DEF is likely to occur under conditions such as the materials used, mixture proportions, and curing method and to consider measures to suppress the DEF if there is a possibility of DEF. The other is to obtain information that contributes to the identification of model parameters that are necessary for improving the accuracy DEF expansion predictions and assessments. There are almost no direct comparisons between the acceleration method of repeated heating and cooling after high-temperature curing, or repeated drying and wetting, which is mainly used overseas, as shown in the previous section, and the method of adding K_2SO_4, which is mainly used in Japan. Therefore, we established test procedures for method A (underwater storage), which is commonly used in Japan to promote DEF with the addition of K_2SO_4, and method B (underwater storage after repeated heating and cooling), which was created based on LPC Test method No. 66. The mixing conditions were W/C = 0.50 (using high-early-strength Portland cement) and W = 175 kg/m^3. There were several discussions on the method for setting the amount (or rate) of K_2SO_4 to be added, but three additive levels of SO_3 amounts (0%, 1%, and 2%) were set (ratio to cement mass) herein. The conditions for heat curing (or steam curing) were as follows: 4 h for pretreatment, 20 °C/h for heating and cooling speed, and 90 °C as the maximum temperature during 12 h. As long as there is a heating device (such as a programmable thermostat), the above can be carried out using general concrete-related laboratory equipments and without the use of any other special equipment. The experiments are ongoing.

4. Precautions in DEF Diagnosis and Quantitative Assessment of Cracks

4.1 Diagnosis of DEF considering other deterioration phenomena

In WG3, we also summarized the precautions in diagnosing ISRs. When diagnosing DEF, it is important to distinguish it from ASR, which has similar appearance deterioration phenomena, and other deterioration phenomena with similar microscopic characteristics (external sulfate attack, aggregate drying shrinkage, and frost damage)[16)].

When concrete with DEF is observed using a polarizing microscope or an electron microscope, it is characterized by a "gap" around the aggregate (Photo 1)[17)]. This is hypothesized to be owing to the growth of ettringite in the paste and the swelling of the paste[18), 19)], which is often accompanied by the formation of ettringite in the gap. However, when observing the gap, it should be noted that the gap width under the constraint condition reduces owing to the DEF, thus making observation difficult[20)].

In ASR, cracks that extend from the aggregate particles to the surrounding paste and ASR gel are observed (Photo 2)[21)]. It has been reported that ASR and DEF not only occur independently, but also influence each other chemically, which causes combined deterioration[22)]. Currently, it is difficult to clearly distinguish between ASR and DEF. However, as mentioned above, it is important to focus on the gaps and ettringite around the aggregate as a symptom of DEF whilst the cracks extending from the inside of the aggregate to the paste and ASR gel is a typical feature of ASR. From the microscopic view, it is also important to observe the continuity of the cracks and the type of the crack-filling product in detail. A core swelling test has also been proposed to distinguish between the DEF and ASR potential[23)] and should be used in combination with an observation.

Similarities with microscopic feature of DEF damage have also been reported, which include external sulfate attack, drying shrinkage of aggregates, and frost damage. External sulfate attack can be distinguished from DEF by characterizing the permeation of sulfate ions and coexisting substances other than ettringite (such as gypsum)[16]. As aggregates containing clay minerals may develop cracks around aggregate particles, it is necessary to distinguish between aggregate types[24]. In the case of frost damage involving the action of water, ettringite generated through a solution is universally observed in air bubbles, etc., but it can be distinguished from DEF by carefully observing the presence or absence of gaps around the aggregate[21].

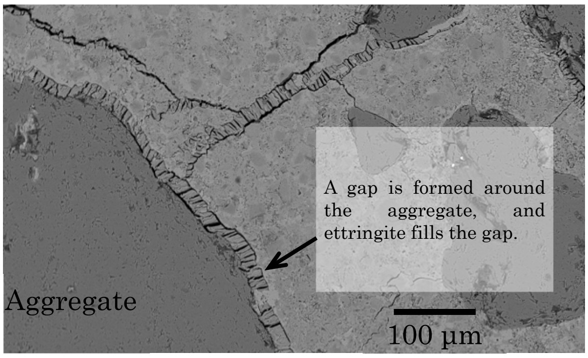

Photo 1: Gap around aggregate due to DEF expansion[17]

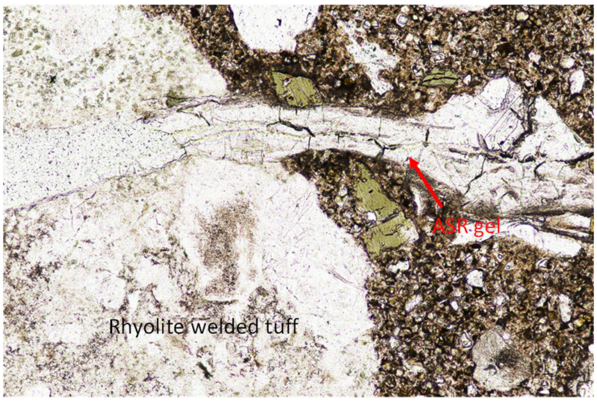

Photo 2: Cracks caused by ASR[21]

4.2 Quantification of cracks

The DRI (damage rating index) method has been proposed as a method for semi-quantitatively assessing the state of deterioration of concrete damaged by an ASR from the observation results of cracks[e.g., 25]. In the DRI method, grid lines are drawn on the surface of a plate-shaped sample cut from concrete, and the features listed in **Table 2** are counted within each grid, multiplied by a weighting factor, and summed up[26]. The calculated DRI value is used to assess the degree of concrete deterioration. The petrological features and weighting factors observed using the DRI method differ depending on the researcher, and the process of calculating the DRI value is currently non-standardized.

Data that quantitatively captures cracks via "observation," such as that in the case of the DRI method may be useful for the analysis of deterioration mechanism. Various methods have been reported for the quantitative assessment of cracks, including advanced digital technology and artificial intelligence. Specific examples include a method for analyzing photographed images of concrete structures and specimens without sample preconditioning; a method of obtaining photographs after polishing; a method of drawing and observing grid lines as in the case of DRI; a method in which the specimen is impregnated with a fluorescent epoxy resin; and a method of drawing a random dot pattern on the photographed surface and assessing it using the digital image correlation (DIC) method.

In the unprocessed-image technology, a technology has been proposed that extracts cracks from digital images via image analysis using the wavelet transform, semi-automatically draws crack diagrams of concrete structures, and automatically calculates a histogram of the crack length, total crack extension, average crack width, and crack density[27]. It is expected that such technology will be applied to concrete deterioration analysis in the future.

Table 2: Examples of features and weighting factors of the DRI method[26]

Petrographic features	Weighting factors
Crack in coarse aggregate	0.25
Opened cracks in aggregate	2
Cracks with reaction product in coarse aggregate	2
Coarse aggregate debonded	3
Corroded aggregate particle	2
Crack in cement paste	3

5. Investigation of internal swelling deterioration of concrete using aggregate-containing iron sulfide

5.1 Overview of deterioration

Although this problem has received little attention in Japan thus far, it has been highlighted that the use of aggregates containing iron sulfide in concrete may cause deterioration. Expansion of concrete and/or pop-outs, owing to the reaction of iron sulfide, have been reported, and the latter is commonly reported in Japan. In WG4, we summarized the cases of deterioration that have been reported worldwide, as listed in **Table 3**[28]. From **Table 3**, there are various types of iron-sulfide minerals that have caused the deterioration of structures, but among these, pyrrhotite and pyrite are the most common minerals of deterioration. Furthermore, it was found that pyrite rarely existed as a single phase and coexisted with pyrrhotite in many cases. Furthermore, it was found that pyrrhotite was detected in many of the cases wherein severe deterioration such as large crack opening occurred. Based on a comprehensive evaluation of these results, it was suggested that among iron sulfides, pyrrhotite could have a strong influence on the severe expansion that causes cracks in concrete structures.

Table 3: Real case of deterioration of concrete with iron sulfide-containing aggregate

Reference	Origin	Year	Structure	Deformation	Aggregates used			Estimated cause of deterioration
					Rock type	Mineral	Coexisting mineral	
Lugg et al.5)	U.K.	1900–1950	Block	A	Iron sulfide slag	Py, Po	—	・Aggregate swelling
Moum et al.6)	Norway	1959	Housing foundation	A	Alum shale	Po	—	・Sulfate deterioration (thaumasite) ・Sulfate deterioration
Martna et al.7)	Sweden	1960–	Water power plant	A, B	Shale	Po	—	・Sulfate deterioration
Vasquez et al.8)	Spain	1962–1995	Building, housing foundation, bridge, dam	A, B, F	Shale	Py, Po	Limestone, phyllite	・Sulfate deterioration (ettringite)
Bérard et al.9)	Canada	1975	Housing foundation, bridge	C, D	Shale	Po	Limestone, diatomite	・Swelling due to gypsum
Geiss et al.10)	U.S.A.	1980–2016	Housing foundation	A, C	—	Po	—	・Identification of Po and comparison with past cases
Oberholster et al.11)	South Africa	1984	Housing foundation, floor slab	A, B	Slate	Py,Po,Ccp	Magnetite	・Sulfate deterioration (thaumasite)
Shayan12)	Australia	1988	Floor slab	D	Shale	Py	—	・Swelling due to jarosite, gypsum, halotrichite
Macleod et al.13)	Scotland	1990	Bridge	D, F	Diabase	Py, Po	—	・Swelling due to rust (iron hydroxide) ・Sulfate deterioration (ettringite)
Ayora et al.14)	Spain	1998	Dam	A, B, C	Schist	Po	—	・Sulfate deterioration (ettringite) ・Swelling due to gypsum, melanterite
Rodrigues et al.1)	Canada	2004–2008	Housing foundation, floor slab	A, B	Gabbro	Py, Po, Ccp, Pn	Anorthite, siderite	・Swelling due to rust (iron oxyhydroxide) ・Sulfate deterioration (ettringite, thaumasite)
Dubey et al.15)	India	2004	Dam	B, E	Schist	Po	—	・Sulfate deterioration (ettringite) ・Swelling due to gypsum
(Tagnit-Hamou et al.16)	Canada	2005	Housing foundation, floor slab	B, C	Anorthite	Po	Mica	・Swelling due to rust (goethite) ・Sulfate deterioration (ettringite, thaumasite)
Schmidt et al.17)	Switzerland	2011	Dam	A, E	Schist	Py, Po, Ma	Feldspar, quartz, biotite, muscovite	・Swelling due to rust (iron oxide, iron hydroxide)
Hawkins et al.18)	Ireland	2012	Floor slab, pavement	A, B	Calcilutite	Py	—	・Sulfate deterioration (ettringite)

Deformation: A: swelling and deformation; B: macroscopic cracks; C: tortoise-shell cracks; D: pop-out and peeling; E: rust and rust juice; F: spots and discoloration

Containing iron sulfides: Py (Pyrite), Po (Pyrrhotite), Ccp (Chalcopyrite), Pn (Pentlandite), Ma (Marcasite)

5.2 Overview of deterioration mechanism

Pyrrhotite is a stoichiometrically amorphous mineral represented by the chemical formula $Fe_{1-x}S$, where x ranges from 0 to 0.125[29]. According to some studies[30), 31)], internal swelling of concrete caused by iron-sulfide-containing aggregates can be attributed to the oxidation of pyrrhotite—which is more reactive than pyrite—to the formation of Fe-containing rust such as goethite, ferrihydrite, and limonite, and this reaction causes primary swelling. Sulfate ions are subsequently released into the system from the oxidized pyrrhotite, which results in precipitation of ettringite and thaumasite in the cement matrix, thus resulting in secondary swelling.

Schmidt et al. prepared concrete samples using concrete cores obtained from a dam and aggregates similar to those used in dams and conducted a chemical analysis; their results showed that pyrrhotite accounted for approximately 80 mass% of the sulfide minerals contained in the aggregate, and pyrite and pyrrhotite were oxidized and altered from the surface, first changing to iron oxide (Fe_2O_3) and secondarily to iron hydroxide (goethite and $Fe(OH)_3$)[32]. In addition, when assuming that goethite and $Fe(OH)_3$ produced by the oxidation of pyrrhotite and pyrite are produced at a ratio of 1:1, then the main cause of the internal swelling of concrete using iron sulfide-containing aggregates is the volumetric swelling of sulfide minerals because the maximum swelling rate of iron-sulfide-containing aggregates during oxidation is 0.07%, which is sufficient to cause cracks in aggregates. In addition, the effect of ettringite on internal swelling remains unclear.

5.3 Potential risk assessment method for iron-sulfide-containing aggregates

Rodrigues et al. proposed tests to confirm the safety of using iron-sulfide-containing aggregates in concrete[33), 34)]. In this test, as Phase 1, the amount of sulfur contained in the aggregate and the content of sulfide minerals are assessed. If the total sulfur content in the aggregate exceeds 1 mass%, it should be excluded, and if it is less than 0.1 mass%, it is determined to be safe. Herein, if the total sulfur content is 0.1 mass% or more and less than 1 mass%, the test proceeds to phase 2.

In Phase 2, the aggregate is enclosed in a column, and the oxygen consumption is measured. If the oxygen consumption is less than 5.0%, it is determined to be safe, and if it is 5.0% or greater, the test proceeds to Phase 3.

The Phase 3 is divided into first stage and second stage. The first-stage test is focused on the ASR and the oxidation reaction of iron sulfide aggregates and exposes the mortar bar to an oxidizing environment using an oxidizing agent for 90 days. If swelling of more than 0.1% is observed in the first-stage test, it is assumed that internal swelling caused by the oxidation of ASR or iron sulfide aggregates has occurred, and the aggregate is determined to be harmful. If the swelling is 0.1% or less in the first stage, the test proceeds to the second stage.

In the second stage, focusing on the precipitation of ettringite and thaumasite, the mortar bars are transferred to a low-temperature, high-humidity environment, wherein these hydrates are likely to precipitate, and expose it for 90 days. Here, if it moves to the second stage and swelling does not progress, it can be used as aggregate for concrete. However, if the internal swelling has progressed further, it is determined that the internal swelling has occurred owing to the precipitation of thaumasite and ettringite, and it cannot be used as an aggregate.

5.4 Verification of potential risk assessment method

In WG4, we conducted the mortar bar test introduced in Section 5.3 for the purpose of elucidating the detailed mechanism of the internal deterioration of hardened cement-based materials using iron-sulfide-containing aggregates and verifying the validity of the assessment method. This test yielded the following results.

(1) From the observation of the appearance of the mortar bar, pop-outs were observed on the mortar surface. Furthermore, the pyrrhotite that existed from the surface to a depth of approximately 5 mm was oxidized.

(2) On using scanning electron microscopy–energy dispersive X-ray spectrometry (SEM-EDS) to observe the elemental composition inside the mortar, the formation of ettringite and thaumasite was not observed in the deteriorated part comprising cracks (**Table 4**).

(3) As a result of the elemental mapping using SEM-EDS of the mortar after the test, it was found that the sulfate ions inside the mortar were eluted outside the system. This is hypothesized to be because sodium hypochlorite, which was used to promote oxidation, stabilized the Friedel's salt in the system, thus causing sulfate ions to dissolve in the liquid phase and eventually to be eluted out of the system. Therefore, it was suggested that it may be difficult to assess the secondary swelling due to the formation of ettringite and thaumasite in this study.

Table 4: Chemical composition of iron sulfide its vicinity (EDS, wt %)

Spot	SiO_2	TiO_2	Al_2O_3	Fe_2O_3	MnO	MgO	CaO	Na_2O	K_2O	SO_3	P_2O_5	Cl	Total
E1*	0.12	0.00	0.01	59.06	0.00	0.00	0.69	0.13	0.10	36.39	0.01	0.08	96.58
E2	0.34	0.00	0.03	63.68	0.01	0.03	12.20	0.71	0.02	2.91	0.00	2.01	79.93
E3	7.53	0.15	14.04	4.94	0.00	0.18	39.65	0.48	1.36	1.36	0.00	8.00	69.69
E4	1.42	0.28	13.90	2.21	0.04	0.15	40.15	0.18	0.65	0.65	0.15	10.43	59.78

*E1 represents the mass concentration of the element as a non-oxidized metal.

<Composition formula normalized to the theoretical value for each mineral>

E1 : pyrrhotite $Fe_{0.93}S_{1.00}$

E3 : Friedel's salt $(Ca_{3.35}, Na_{0.05})_{3.40}(Al_{1.30},Si_{0.59},Fe_{0.29})_{2.19}[(2Cl)_{0.53},(SO_4)_{0.08},(2OH)_{0.39}]_{1.00}O_6.nH_2O$

E4 : Friedel's salt $(Ca_{4.08}, Na_{0.02})_{4.10}(Al_{1.56},Si_{0.13},Fe_{0.16})_{1.85}[(2Cl)_{0.84},(SO_4)_{0.05},(2OH)_{0.11}]_{1.00}O_6.nH_2O$

6. Conclusion

In this Technical Committee, we reviewed the underlying microscopic mechanism of the swelling deterioration common to ISR, investigated the necessary basic technologies from the viewpoint of the performance assessment of ISRs-damaged structures, and discussed the ideal ISR risk assessment method. Although there still exist many unclear aspects of the microscopic mechanism of ISR, it was confirmed that the method of dealing with the (micro) cracks that occur as a result is crucial in terms of both observation and analysis. In this technical committee report, we did not present a specific risk assessment method, but we were able to present part of the elemental technology and framework necessary for that purpose. At present, there is a large gap between observation (particularly materials) and analysis (structural), and bridging this gap is the most important in realizing the entire process from diagnosis to performance assessment/prediction.

The results of the activities of this technical committee were compiled in a report, submitted as review papers mainly to the Concrete Journal and to the Journal of Advanced Concrete Technology. This is intended to increase the motivation of the members of the technical committee, which started its activities during the COVID-19 pandemic. It is hoped that this technical committee will serve as an impetus to vigorously advance the research activities of each committee member and the research in this field.

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