

ALKALI-AGGREGATE REACTION IN NORTHERN FRANCE : A REVIEW

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A statistical survey was conducted on 140 bridges located in the northern part of France (Lille, Reims and Paris). This survey indicates that 5% of the studied bridges present important degradation signs likely related to AAR; 24% of the bridges show evident signs of AAR having caused up to now only minor damages; 57% of the bridges show little degradation signs; and 14% of the bridges show no degradations. This study enlighten the close relationship existing between the used aggregates (siliceous aggregates or metamorphic limestones aggregates containing pyrite) and the observed deteriorations. The chemical characteristics of the AAR gels are presented. The negative correlation existing between the concrete compressive strength and the gel alkali contents argue for a more expansive nature of the alkali enriched gels.

INTRODUCTION

Until the early eighties only few cases of alkali-aggregate reactions (AAR), mainly affecting dams, were described in France. In the recent years, however, more and more cases have been recognized. An intriguing feature is that most of the observed cases are located in the northern part of France. Two reasons may explain this distribution : the geological characteristics of this region affecting the aggregate choices and the cold and humid weather requiring important additions of de-icing salts in winter. A typical characteristic of the AAR affecting northern France concrete is the association, in many cases, of ettringite (Deloye (1), Larive (2), Brouxel and Valiere (3)). This association of ettringite and AAR products is classically described in AAR affected concretes (Regourd et al (4), Regourd-Moranville (5), Jones and Poole (6), Jones (7), Durand and Berard (8)). A possible raison of such association is the presence, together with the reactive siliceous aggregates, of impure metamorphic limestones aggregates (3). The presence of pyrite, providing sulfur, and clay minerals, providing alumina, may explain the formation of ettringite in these samples ((1), LeRoux and Toubeau (9, 10)). This paper presents the results of a statistical survey conducted on 140 bridges constructed in the seventies in an area between Paris, Reims and Lille. This survey was conducted to detect the bridges showing deterioration due to AAR. Indeed, AAR were known to affect several bridges in this area and at least two bridges had to be destroyed. To eventually prevent any further development of the reaction, for example by stopping the water circulation, and therefore any ionic exchange, an early diagnosis of the reaction is necessary.

RESULTS

Bridge inspection

The survey consisted of a on site visit to observe all the degradations and to take two samples per bridge. The samples were taken in the most deteriorated areas. All the samples were examined with a polarizing microscope. Systematic X-Ray diffraction analyses were also conducted to determine the nature of the used aggregates as well as the occurrence of abnormal amounts of

ettringite. A detailed study (scanning electron microscope observations and energy dispersive X-Ray analysis) was conducted on the four most deteriorated bridges. The detailed study of four other bridges, also located in this area, is additionally presented in this paper to address a global situation of the AAR phenomenon in the northern part of France.

The bridges were divided into four groups (deteriorated, damaged, fair, good) taking into account the observed degradations on the structures and the presence of cracks at the microscopic scale. Seven bridges (5%) present numerous signs of alkali-aggregate reaction (deteriorated group): extensive surface cracking on all the structural members with locally gel exudation; occurrence of abundant cracks observed in thin section together with the presence of reaction rims around certain minerals. Moreover, they often present important amounts of ettringite (> 4.5 %). Map cracking was observed at different scales. Usually, on the entire surface of the affected structural member, a map cracking was observed every 0.4 m. and 2 m. in both longitudinal and transversal directions. These fractures are likely structural fractures related to the bonding of the AAR affected layer on the non affected layer (Diab and Prin (27)). A map cracking, at the cm scale, is observed in the most deteriorated areas. These small cracks are likely the first to be formed. For two bridges, the number of cracks as well as their apertures were compared to a 1983 detailed inspection: the number of cracks was multiplied by a factor ranging between 2.5 and 7 depending on the structural element while their apertures increased in the same time by a factor ranging between 1 and 4. These observations led to the conclusion that a continuing expansive phenomenon is taking place in these bridges. Moreover, 33 bridges (24 %) show evident signs of AAR having caused up to day only minor damages (damaged). They are many chances that most of these bridges will show in the next 10 years the degradation signs observed today in the seven bridges described previously. All the other bridges show few (57%; fair) or no (14%; good) degradations.

Petrographic examination

Siliceous and calcareous aggregates were found in all the studied concretes; the calcareous aggregates being the biggest ones. The calcareous aggregates are of two types: biotectric limestones likely of Cretaceous or Early Tertiary age and metamorphic limestones showing a strong foliation marked by oxides and sulfur minerals (pyrite). These limestones are similar to the Paleozoic limestones described in the Ardennes massif and near Boulogne (Debelmas (11)). It should be pointed out that some of these calcareous aggregates (less than 5 %) are in fact dolomitic limestones as revealed by their X-Ray diffraction analyses.

Many different types of siliceous aggregates have been observed. The main types of siliceous minerals are : small rounded quartz, undulatory extinction quartz, pegmatitic quartz, microcrystalline quartz, opal, trydimite, chalcedony. Dark reaction rims were observed in many cases and particularly around the microcrystalline quartz crystals. Moreover, many quartz crystals are fractured, especially along their rims. The main types of siliceous rocks are : sandstones, granites, gneiss, porphyritic basalts and andesites, diabases, and gabbros. The basic magmatic aggregates (porphyritic basalts and andesites, diabases, and gabbros) were observed only in the bridges located near the Belgium border. They show often intense alteration signs : the plagioclase are saussuritized and the clinopyroxene or amphibole crystals are often transformed into epidote and/or chlorite. These aggregates are very similar to those described in Belgium as coming from the quarries of the Queynast region where large quantities of aggregates have been produced (Soers and Meyskens (12)). The only difference between our description and the description given by these authors is the presence of gabbros which have never been described in this Queynast region.

Many types of aggregates used in northern France concretes are considered as reactive for AAR; for example opal, micro-cristalline quartz and quartz with undulatory extinction (e.g. Coull (13)). The aggregates of the Queynast region are also likely very reactive to AAR because of the presence of abundant deteriorated siliceous glass. Some aggregates contain moreover alkali enriched minerals like feldspar (basalts, andesites, diabases and gabbros of the Queynast region). Glauconite aggregates are also commonly found in the concretes of this region. This mineral, also

enriched in alkalis $[(K,Na)_2(Fe^{3+},Fe^{2+},Al,Mg)_4[Si_6(Si,Al)_2O_{20}](OH)_4]$, is present in all the Late Cretaceous formations North of Paris (11). Up to day, however, there is no relationship between the presence of glauconite and the occurrence of AAR.

SEM and EDXA results

The samples were observed and analyzed using an Hitachi scanning electron microscope coupled with a Link energy dispersive spectrometer. The analytical procedures followed in this study are similar to those reported in detail by Brouxel (14) and are briefly described in the paper by Remy and Brouxel ((15), this issue). The analytical errors used in this study are : $SiO_2 \pm 4\%$; $CaO \pm 1.1\%$; $Na_2O \pm 1.2\%$; $K_2O \pm 0.9\%$.

AAR gel aspects. Most of the gel types described in the literature (e.g. (5)) were observed in these samples; the most common type being the massive gel (Fig. 1, 2 and 3). Lamellar and rosette-like gels are quite rare while the association with ettringite and/or thaumasite is common (Fig. 3 and 4; see also (3)).

AAR gel analyses. The AAR gels are chemically characterized by an important variation of their silicium, calcium, sodium and potassium contents (Table 1). These elements represent around 100% of the analyzed elements : $SiO_2 = 21$ to 95% , $CaO = 3$ to 77% , $Na_2O = 0$ to 4% , and $K_2O = 0.3$ to 28% . In a silicium versus calcium diagram these two elements, as already noted by Knudsen and Thaulow (16), are negatively correlated (Fig. 5). The deviation of the point away from this 100% line is related to the alkali enrichment of the AAR gel. Many points are close to the 100% line indicating that their alkali content is quite low. In figure 6, silicium is plotted against $Na_2O + K_2O$. The analyzed gels show a maximum alkali enrichment for SiO_2 ranging between 40 and 50% (see also Brouxel (17)).

DISCUSSION

The fact that most of the AAR problems are located in the northern part of France is likely the consequence of two causes : the nature of the used aggregates and the weather conditions. However, AAR have been reported for many years in two nearby countries (Belgium and Great Britain : (6), (12), Fookes et al (18), Fookes et al (19), Wood (20), Soers (21), Besem and Demars (22), Hobbs (23), Jones (24), VanGemert (25), DeCeukelaire (26)). The geological characteristics and weather conditions of France, Belgium and Great Britain are similar : they are no differences between the geological features of the Paris basin and the London basin, and the weather characteristics require in these countries the addition in winter of important amounts of de-icing salts. Moreover, Belgium aggregates are commonly used in France. Nonetheless, well documented AAR phenomenon were described much earlier and more often in Great Britain (since 1975) and Belgium (since 1984). This is likely related to the fact that detailed studies of concrete structures are more recent in France. It is possible that in the future, with the increasing number of studies like this one or those now conducted by the Laboratoire Central des Ponts et Chaussées (Larive and Louarn (28)), a similar number of cases will be described.

The compilation of around 300 X-Ray diffraction analyses conducted on 140 bridges shows some variations of the nature of the used aggregates related to the geographic position of the bridges, and therefore related to the geological features of the different areas. The quartz/calcite ratio, reflecting the siliceous/calcareous aggregate ratio, range between 1 in the northern part of the studied area to 6 in the southern part of this area. These values represent of course averages and some of the calcite analyzed by X-Ray diffraction is probably due to the carbonation of the cement paste. In the south, where little limestones aggregates are used, biodetritic Cretaceous or Tertiary limestones are usually observed. In the north, where higher amounts of limestones aggregates are used, these limestones are mainly metamorphic Paleozoic limestones. The presence of abundant pyrite grains in these limestones may explain the common association of AAR gel and ettringite (3).

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TABLE 1 - EDXA microanalyses of the AAR gels observed in seven northern France bridges

Sample	Na2O	MgO	Al2O3	SiO2	S	Cl	K2O	CaO	FeO	Total	Na2O+K2O
1-23	0,0	0,5	1,3	42,4	0,0	0,1	0,8	54,7	0,2	100,0	0,8
1-23	0,0	0,1	0,9	37,8	0,0	0,0	3,6	57,7	0,0	100,0	3,6
1-23	0,8	0,1	0,0	88,2	0,0	0,2	0,2	10,6	0,0	100,0	1,0
1-23	0,0	0,0	0,0	71,8	0,0	0,0	3,4	24,8	0,0	100,0	3,4
1-23	0,3	0,0	0,2	84,0	0,0	0,0	1,4	14,0	0,1	100,0	1,7
1-23	0,8	0,0	1,8	40,5	0,0	0,2	3,7	52,8	0,3	100,0	4,5
1-23	0,2	0,0	0,0	76,6	0,0	0,1	0,1	23,0	0,1	100,0	0,3
1-23	0,0	0,3	0,6	76,6	0,0	0,0	0,9	21,7	0,0	100,0	0,9
1-23	0,0	0,0	0,0	71,8	0,0	0,0	3,4	24,8	0,0	100,0	3,4
1-23	0,8	0,1	0,0	88,2	0,0	0,2	0,2	10,6	0,0	100,0	1,0
1-23	0,9	0,1	0,4	68,9	0,0	0,2	0,9	28,7	0,0	100,0	1,9
1-23	0,0	0,0	0,0	54,5	0,0	0,0	5,1	40,4	0,0	100,0	5,1
1-23	0,0	0,1	0,1	65,3	0,0	0,0	0,9	33,4	0,2	100,0	0,9
1-23	0,0	0,0	2,1	48,7	0,0	0,1	5,3	43,7	0,1	100,0	5,3
1-23	0,0	0,0	0,2	53,6	0,0	0,0	3,8	42,3	0,0	100,0	3,8
1-23	0,0	0,0	0,0	54,6	0,0	0,0	4,8	40,6	0,0	100,0	4,8
1-23	0,0	0,3	0,0	61,1	0,0	0,1	5,5	33,1	0,0	100,0	5,5
1-23	0,2	0,3	0,1	46,9	0,0	0,0	0,8	51,8	0,0	100,0	1,0
1-23	0,0	0,1	0,8	44,7	0,0	0,0	1,0	53,4	0,0	100,0	1,0
1-23	0,9	0,1	2,1	38,2	0,0	0,0	1,7	56,9	0,2	100,0	2,6
1-23	0,0	0,7	1,3	33,3	0,0	0,0	0,9	64,0	0,0	100,0	0,9
1-23	0,5	0,0	0,9	48,5	0,0	0,0	2,0	48,1	0,0	100,0	2,5
1-23	0,0	0,0	1,5	32,5	0,0	0,0	1,9	63,9	0,1	100,0	1,9
1-23	0,0	0,0	0,0	30,2	0,0	0,0	1,9	67,8	0,0	100,0	1,9
1-24	1,2	0,7	1,0	48,7	0,0	0,0	15,7	32,4	0,2	100,0	16,9
1-24	1,4	0,1	2,2	51,3	0,0	0,2	9,7	35,1	0,1	100,0	11,1
1-24	2,7	0,3	0,0	64,9	0,0	0,0	10,9	20,9	0,3	100,0	13,5
1-24	2,4	0,5	0,0	62,3	0,0	0,1	10,7	24,1	0,0	100,0	13,1
1-24	0,2	0,0	0,0	91,5	0,0	0,0	1,3	7,0	0,0	100,0	1,5
1-24	0,3	0,0	0,0	42,4	0,0	0,0	11,9	45,2	0,3	100,0	12,2
1-24	0,3	0,0	0,3	40,2	0,0	0,0	11,3	47,5	0,2	100,0	11,7
1-24	0,9	0,0	2,7	36,0	0,0	0,0	4,9	55,2	0,3	100,0	5,7
1-24	0,5	0,7	1,4	34,5	0,0	0,0	1,8	60,7	0,5	100,0	2,2
1-24	0,8	0,0	2,0	43,5	2,0	0,0	4,3	47,3	0,2	100,0	5,1
1-24	1,1	0,3	2,1	39,4	0,0	0,0	10,7	45,7	0,8	100,0	11,8
1-24	0,7	0,0	8,0	33,3	6,4	0,0	3,1	48,4	0,2	100,0	3,8
2-2	0,6	0,0	0,2	94,4	0,0	0,4	1,6	2,7	0,2	100,0	2,2
2-2	3,0	0,0	5,5	60,1	0,0	1,3	7,3	22,7	0,2	100,0	10,3
2-2	0,9	0,0	0,0	95,1	0,0	0,6	0,4	2,9	0,0	100,0	1,3
2-2	1,5	0,0	9,2	51,8	0,0	0,4	9,1	27,3	0,8	100,0	10,6
2-2	4,1	0,0	14,8	47,6	0,0	1,1	5,7	24,3	2,4	100,0	9,8
2-2	0,0	0,0	7,1	35,1	0,0	0,1	12,6	42,1	3,1	100,0	12,6
2-2	0,0	0,0	9,2	90,3	0,0	0,0	0,9	8,7	0,0	100,0	0,9
2-2	0,2	0,0	0,5	82,2	0,0	0,0	5,4	11,7	0,0	100,0	5,6
2-2	0,8	0,0	0,7	50,4	0,0	0,0	20,4	27,7	0,0	100,0	21,2
2-2	3,1	0,3	25,6	37,4	0,0	0,5	9,1	21,2	2,9	100,0	12,1
2-2	0,0	0,4	0,8	42,9	0,0	0,1	20,5	32,6	2,8	100,0	20,5
2-2	0,2	0,2	3,6	39,4	0,0	0,1	10,9	45,2	0,5	100,0	11,1
2-4	0,4	0,2	0,8	44,0	0,0	0,0	28,2	26,1	0,3	100,0	28,6
2-4	0,5	0,1	1,3	48,6	0,0	0,0	19,5	29,5	0,4	100,0	20,0
2-4	0,0	0,1	0,0	59,2	0,0	0,0	3,1	37,6	0,0	100,0	3,1
2-4	0,1	0,1	0,3	42,4	0,0	0,1	3,1	53,9	0,0	100,0	3,2
2-4	0,0	0,1	5,2	29,5	0,0	0,1	0,9	62,4	1,9	100,0	0,9
2-4	0,0	0,6	5,1	26,6	0,0	0,0	0,6	66,4	0,7	100,0	0,6
2-4	0,0	0,4	0,9	31,7	0,0	0,0	1,3	65,0	0,7	100,0	1,3
3	0,3	0,3	0,8	29,8	0,0	0,3	4,4	64,1	0,1	100,0	4,7
4	0,7	0,9	0,0	45,2	0,0	0,1	3,1	50,1	0,0	100,0	3,8
4	3,1	0,0	0,0	33,2	0,0	0,0	3,2	56,4	4,1	100,0	6,3
4	0,0	0,0	0,0	41,1	0,0	0,1	13,3	45,4	0,2	100,0	13,3
5	0,6	0,0	1,0	30,9	0,0	0,0	3,1	64,4	0,0	100,0	3,7
6-1	0,2	0,1	2,8	31,3	0,0	0,0	1,2	64,3	0,1	100,0	1,4
6-1	0,0	0,2	2,8	42,4	0,0	0,0	3,2	51,4	0,1	100,0	3,2
6-2	0,0	0,0	0,9	21,0	0,0	0,6	0,8	76,6	0,1	100,0	0,8
6-2	0,0	0,0	0,3	23,1	0,0	2,1	0,9	73,0	0,6	100,0	0,9
7	0,0	0,0	1,8	29,1	0,0	0,0	0,3	68,4	0,4	100,0	0,3

A close relationship between the quartz/calcite ratio and the degradations was observed. Indeed, on an average basis, the concretes presenting a quartz/calcite ratio close to 1 are much more often deteriorated than the concretes made mainly with quartz aggregates (Fig. 7). Two explanations can be given : first of all, the pessimum content of the reactive siliceous aggregates used in these concretes is obtained for a 50% content; second, the presence of metamorphic siliceous limestones containing pyrite grains is necessary for the reaction to develop. This second hypothesis seems more logical taking into account the fact that ettringite is commonly observed in the most deteriorated concrete structures (Fig. 7).

We compared our EDAX results on the observed AAR gels with the data published in the literature since 1942 : 160 analyses of AAR gels were compiled from 20 papers (this list is of course not exhaustive and the data are available on request: (4), (8), (26), Stanton (29), McConnell et al. (30), Hester and Smith (31), Idorn (32), Buck and Mather (33), Arsberetning (34), Thaulow and Knudsen (35), Poole (36), Poole (37), French (38), Bérard and Lapierre (39), Buck and Mather (40), Cole et al (41), Bariono (42), Oberholster and Krüger (43), Bérubé and Fournier (44), Davies and Oberholster (45)). Only four elements (SiO_2 , Na_2O , K_2O and CaO) were taken into account and normalized to 100%. It can be seen in figure 5 and 6 that the distribution of the published points is clearly bimodal. Most gels present high SiO_2 contents ($\text{SiO}_2 > 70\%$) and a maximum alkali enrichment for SiO_2 contents ranging between 65 and 85 %. Some data show however an important alkali enrichment for SiO_2 contents ranging between 40 and 50 %. This second peak fits perfectly with the maximum obtained in this study. It should be pointed out that many published data come from gel analyzed in concrete enriched in alkalis or stored in alkali enriched water (17). Moreover, gel produced under natural environment conditions are shown to be richer in calcium than gels produced under accelerated conditions (4). These observations question the reliability of accelerated AAR.

These variations may be explained by the amount of available alkalis and/or by differences in the kinetics of the reaction. When the kinetics of the reaction are normal, calcium, if present in quantities large enough, is able to reach the reaction sites : lime-alkali-silica gels are formed. When the reaction is accelerated, calcium, which has a higher hydraulic radius and therefore a lower diffusion rate, does not have time to reach the reaction sites, and alkali-silica gels instead of lime-alkali-silica gels are formed (Wang and Gillott (46)). These authors (46) stated that alkali-silica gel has a high affinity for water and will be able to sorb water and therefore show expansion. However, none of the studied gels can be considered as alkali-silica gels (the calcium content is always higher than the alkali content). It is possible nonetheless to suggest that the gels showing the highest alkali contents are the most able to produce expansion and consequently degradations (17). We have plotted the alkali contents of the AAR gel against the compressive strengths of the structural elements they belong to (Fig. 8). A rather well defined negative correlation was obtained. If we consider that the average standard concrete compressive strength is 35 MPa, it should be pointed out that three structures, affected by AAR, present higher compressive strength (45 to 50 MPa). They present gels with low alkali contents (less than 2 %). These gels are likely rather immature and present in small quantities filling all the voids and giving to the concrete a higher compressive strength. The concrete structures presenting the lowest compressive strengths (around 20 MPa) present on the contrary gels with high alkali contents ($\text{Na}_2\text{O} + \text{K}_2\text{O} = 6$ to 9 %). These observations agree with the hypothesis that gels with high alkali contents are the most able to produce expansion and therefore microstructural degradations.

CONCLUSION

This survey conducted on 140 bridges in the northern part of France shows that 5% of the studied bridges present important degradation signs that may be related either to AAR or to AAR + ettringite development : e.g. extensive surface cracking and gel exudation. Twenty four percent of the bridges show evident signs of AAR having caused up to now only minor damages. Extensive studies are necessary on these structures to evaluate the possible evolution of the phenomenon in terms of degradations and structural consequences. The development of the AAR is mainly related

to the presence in the concrete of two types of reactive aggregates : microcrystalline quartz crystals showing often reaction rims and metamorphic siliceous limestones containing pyrite grains. The presence of these sulfur enriched minerals explain why ettringite is often associated with the AAR gels. This survey enlighten also the close relationship between the nature of the used aggregates and the AAR development.

The EDAX studies of the AAR gels confirm the important variation of their chemical composition. A negative correlation was defined between the alkali content of the gel and the concrete compressive strength: gel with low alkali contents (< 3%) were found in structures presenting high compressive strength (40 to 50 MPa) while gel with high alkali contents (> 6%) were found in structures presenting lower compressive strength (\approx 20 MPa). Immature AAR gels, characterized by low alkali contents, and filling all the existing voids improve the concrete compressive strength. On the contrary, well developed AAR gels, characterized by higher alkali contents, are most able to show expansion and therefore to produce degradations of the concrete diminishing its compressive strength.

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Figure 1 SEM micrograph of a massive AAR gel

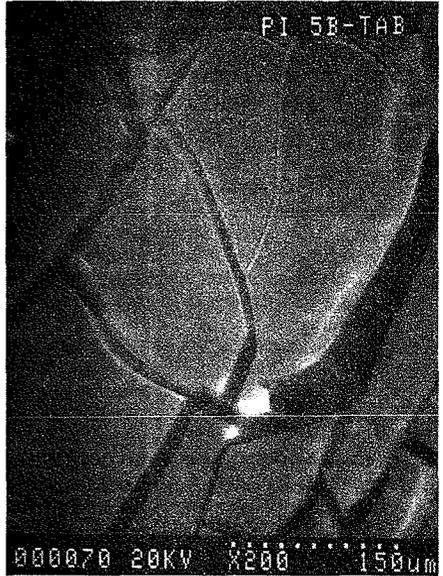


Figure 2 SEM micrograph of a massive AAR gel

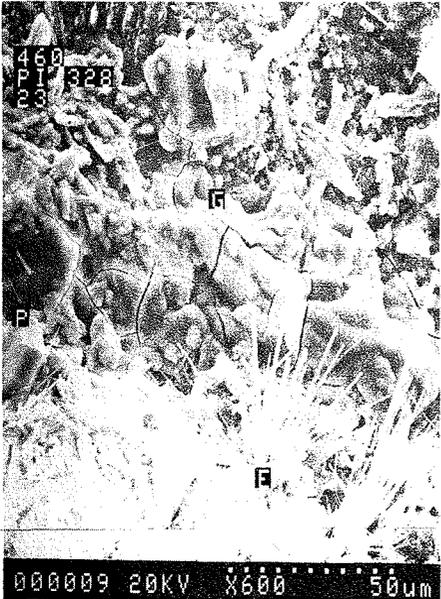


Figure 3 SEM micrograph of AAR gel associated with ettringite



Figure 4 SEM micrograph of massive ettringite + thaumasite + calcite patch on a reactive aggregate

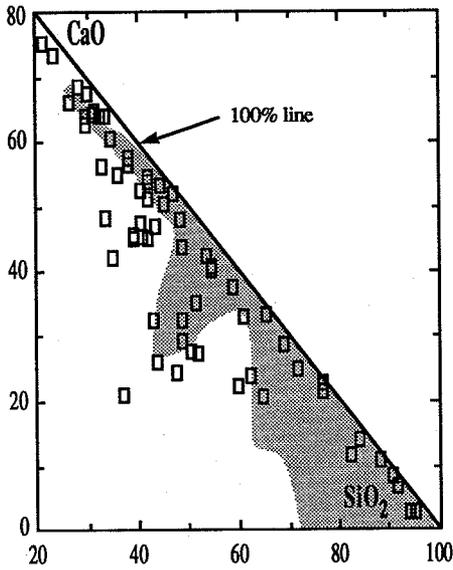


Figure 5 SiO₂ vs CaO graph of all AAR gels
The gray area represents the field of the literature data (see text for details and references).

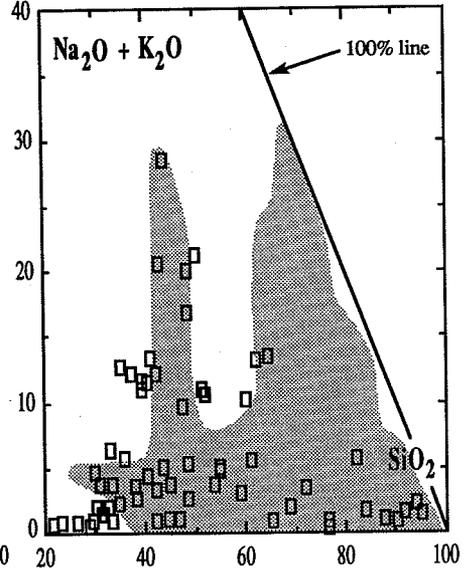


Figure 6 SiO₂ vs Na₂O + K₂O graph of all gels
(see text for details and references).

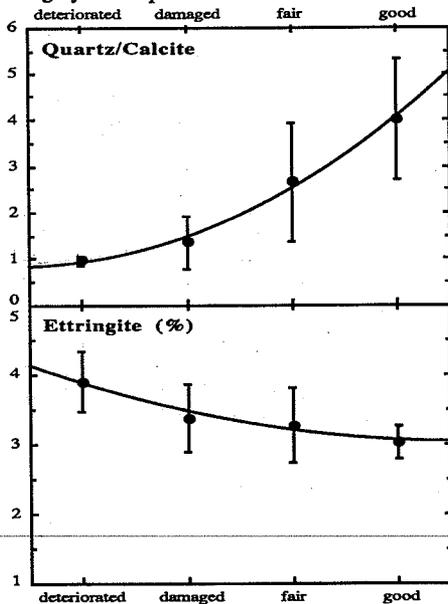


Figure 7 Quartz/Calcite ratio and ettringite % vs the deterioration conditions of the bridges

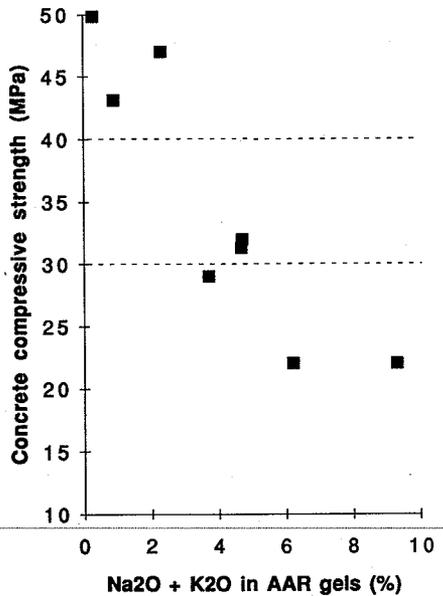


Figure 8 Variation of Na₂O+K₂O in the gels vs the concrete compressive strengths