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Evidence from the highways agency thaumasite investigation in Gloucestershire to support or contradict postulated mechanisms of thaumasite formation (TF) and thaumasite sulfate attack (TSA)

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Abstract

In March 1998 the thaumasite form of sulfate attack (TSA) was identified in the reinforced concrete foundations of Tredington Ashchurch Road Bridge on the M5 Motorway in Gloucestershire, UK and the UK Government's Highways Agency appointed Halcrow to investigate the problem. A total of 28 structures were selected for investigation to encompass sites where bridge works were planned and structures were assessed to be at greatest risk as well as to provide a representative sample of sites and structure types.

Desk study work and peer reviews were continued throughout the project to establish protocols for the site work and facilitate the interpretation of the data. The latter included a dual approach of a systematic examination of the data collected to identify trends coupled with a review of postulated mechanisms for the formation of thaumasite from the desk study work against the data from the investigation. This paper summarises the findings of this review, which examines in detail four proposed stages in the development of TSA.

Keywords: Thaumasite; Highway; Mechanism

1. Introduction

In March 1998 the thaumasite form of sulfate attack (TSA) was identified in the reinforced concrete foundations of Tredington Ashchurch Road Bridge on the M5 Motorway in Gloucestershire, UK. The UK Government's Highways Agency appointed Halcrow to undertake an extensive study of the problem. This was called the thaumasite investigation. The objective of the study was to determine the mechanism, extent and consequences of thaumasite formation (TF) and the thaumasite form of sulfate attack (TSA) at highway structures on the M5 Motorway and on other trunk roads in Gloucestershire. In the course of the study, methods of identifying, monitoring and predicting TF and TSA were to be evaluated.

A total of 28 structures were investigated, working to a staged program and to several different levels of investigation. The structures were selected to encompass sites where bridge works were planned and structures were assessed to be at greater risk as well as to provide representative sample of sites and structure types.

In order to establish protocols for the site work and facilitate the interpretation of the data, desk study work and peer reviews were continued throughout the project.

2. Approach to interpretation

The early stages of the investigation showed that the occurrence of thaumasite was variable within and between structures suggesting thaumasite growth may be complex and dependent on one or a combination of many variables, e.g. concrete mix, backfill type and groundwater chemistry and level.

The chemical and physical complexity of the soil and concrete systems and the number of factors varying across the 28 sites investigated necessitated a dual approach to interpretation:

- The data was systematically examined to identify and interpret trends and other features.

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- A desk study of published or available information was used to identify a range of possible processes for TF and TSA followed by a review of whether the data collected supports or contradicts these postulated processes.

This paper considers the latter of the above approaches when applied to the processes within the concrete. A more comprehensive treatment of the processes, including those in the ground has been reported elsewhere [1]. Published information provided a logical starting point for possible reaction processes. Much of this information is based on laboratory work due to the limited number of published field cases. However, a reliance on laboratory studies alone was considered inappropriate, as this could exclude features and factors of practical significance for highway structures in Gloucestershire. Therefore a peer review was also undertaken to identify possible reaction processes.

Possible mechanisms in the development of TSA are summarised in Section 3. It should be noted that a wide range of postulated (and largely unproven) mechanisms were included for review against the data. All the test data was obtained from structures with an age of approximately 30 years at the time of investigation therefore the stages in the mechanism described below have been based on the condition of the concrete at different depths, with the condition at the surface representing the last stage of the reaction. The evidence for the ground conditions during the earliest stages of the mechanism often relies on an extrapolation back from the time of the investigation and as such is very tentative. Notwithstanding these severe limitations, it is hoped that this speculative approach will prompt others

Fig. 1. Breakout of concrete to column with TSA showing white pasty surface at 5 mm depth and white 'halos' around aggregate at 10 mm depth.

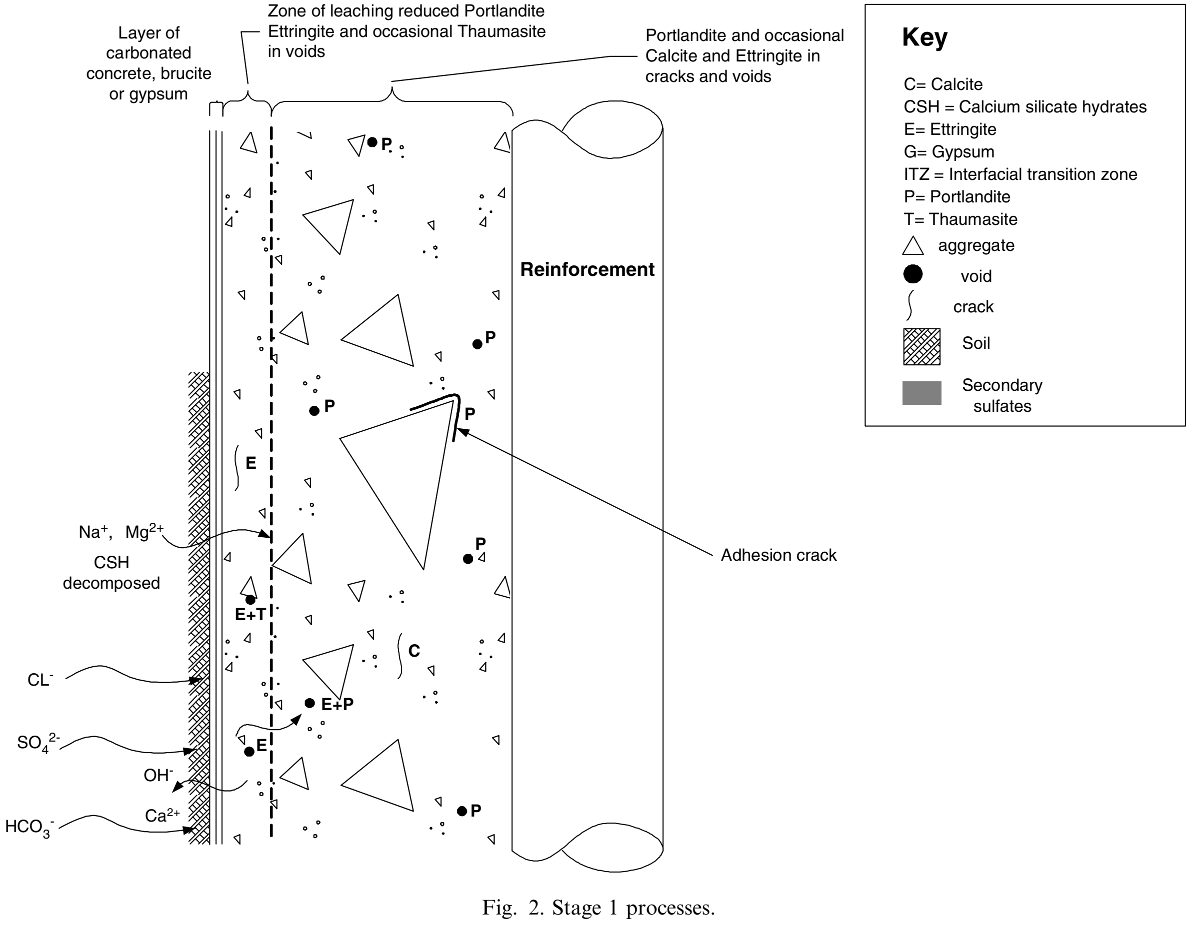

Fig. 2. Stage 1 processes.

to provide a more rigorous evidence for the mechanisms of TF and TSA.

3. Stages in process

3.1. General

The petrographic examination of cores from Tredington Ashchurch Road Bridge, breakout work on site (Fig. 1) and high pressure water jetting trials confirm the presence of four zones of attack, starting at depth in the concrete and moving towards the exposed surface [2]:

- Zone 1: isolated clusters of ettringite or thaumasite within voids with no sign of physical disruption.
- Zone 2: finer cracks infilled with thaumasite, occasional 'halos' of thaumasite around aggregate particles and absence of other sulfate-bearing minerals and portlandite.
- Zone 3: wide sub-parallel cracking infilled with thaumasite and 'halos' of thaumasite around the aggregate particles and absence of other sulfate-bearing minerals and portlandite.
- Zone 4: soft white reaction products predominantly of thaumasite.

These different zones appear to represent different stages in the process, from TF in Zone 1 to extensive TSA in Zone 4. Using these zones as a guide, four stages in the development of TSA have been postulated based on the desk study (Figs. 2–5).

3.2. Postulated stage 1 processes

The groundwater is nearly saturated with calcium sulfate and carbonate. These are brought into contact with the concrete, together with magnesium and sodium sulfates and chlorides (following the application of de-icing salts). Sulfates and chlorides slowly diffuse into the concrete and chlorides are reduced at the surface due to seasonal wash-out. Sulfuric acid developed within the groundwater reacts with the dolomitic limestone aggregate and calcium hydroxide in the concrete to form magnesium sulfate, brucite and gypsum and precipitate calcium carbonate within cracks and pores [3].

The calcium hydroxide is reduced near the surface of the concrete by leaching and there is a corresponding

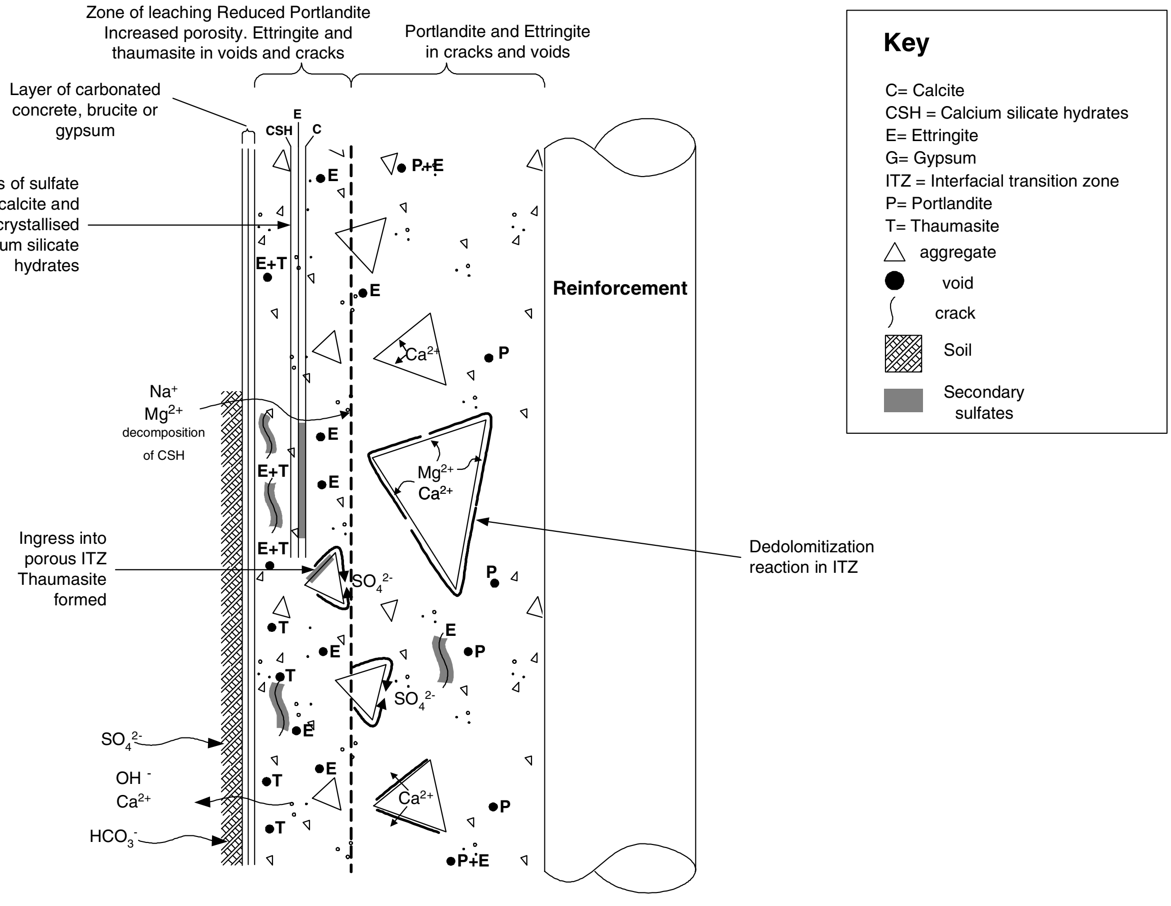

Fig. 3. Stage 2 processes.

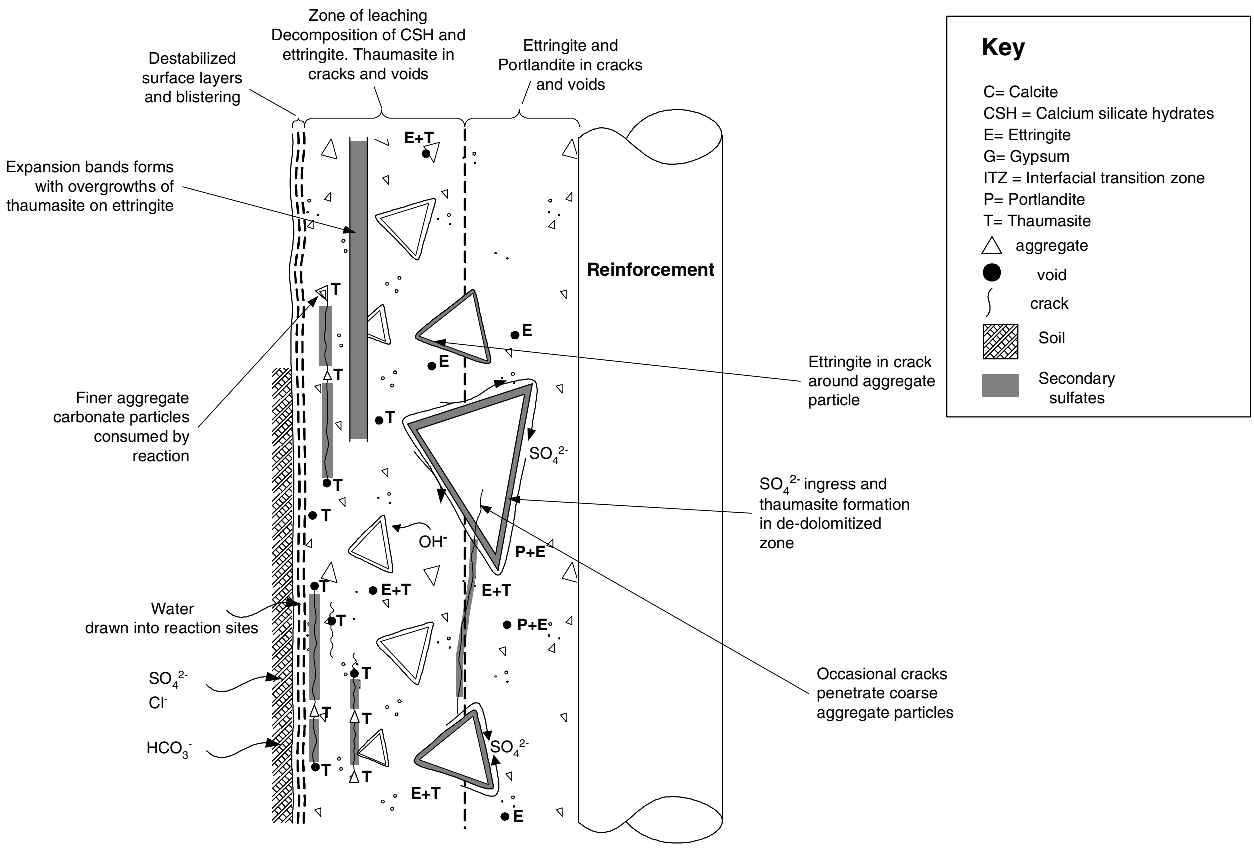

Fig. 4. Stage 3 processes.

increase in porosity. Calcium and pH increase in the adjacent ground leading to the deposition of gypsum and occasionally ettringite in the soil [4].

Enhanced porosity at the surface of the concrete and adjacent to aggregate particles promote the ingress of salts. Ettringite forms in these areas and within voids and cracks due to sulfate attack of the calcium hydroxide and calcium aluminate hydrates and recrystallisation of the cement hydrates associated with water ingress and leaching. As calcium hydroxide is reduced by leaching the other hydration products sequentially decompose as the pH and calcium content fall below critical values [5].

At the surface a relatively stable layer of calcite, brucite and gypsum may form [6]. Ingress of magnesium ions promotes decalcification of the surface of the concrete and can cause a reduction in pH.

The concrete near the exposed surface is weakened due to sulfate attack and leaching of the cement hydrates.

As the sulfate concentration becomes in excess of that to convert the available alumina to ettringite [6] then thaumasite begins to form at nucleation sites such as voids but without evidence of disruption. The thaumasite incorporates high levels of alumina forming a solid solution with ettringite but with limited intermixing.

3.3. Postulated stage 2 processes

Leaching of the concrete and slow diffusion of sulfates and chlorides continues. Chloride diffusion ahead of the sulfates and is promoted by a release of bound chloride due to sulfate attack.

Leaching and decalcification of the concrete by alkali sulfate attack promotes breakdown of the calcium silicate hydrates and weakening and softening of the concrete. Sequential dissolution of the cement hydrates and sulfate species as a result of leaching and decalcification results in diffusion of sulfate and calcium ions into and out of the concrete [5]. A corresponding set of precipitation fronts of secondary sulfates and calcite are generated sub-parallel to the surface. Nucleation of thaumasite occurs preferentially within these mixed layers of reactants within the concrete.

Thaumasite formation at the surface of the aggregate is promoted by consumption of carbonate from the aggregate particles and de-dolomitisation of the dolomitic limestone [4,7]. As the thaumasite develops it becomes more dense and crystalline and incorporates additional ions within its structure [8].

Ettringite and thaumasite continue to form within voids and grow into the available space but without evidence of disruption.

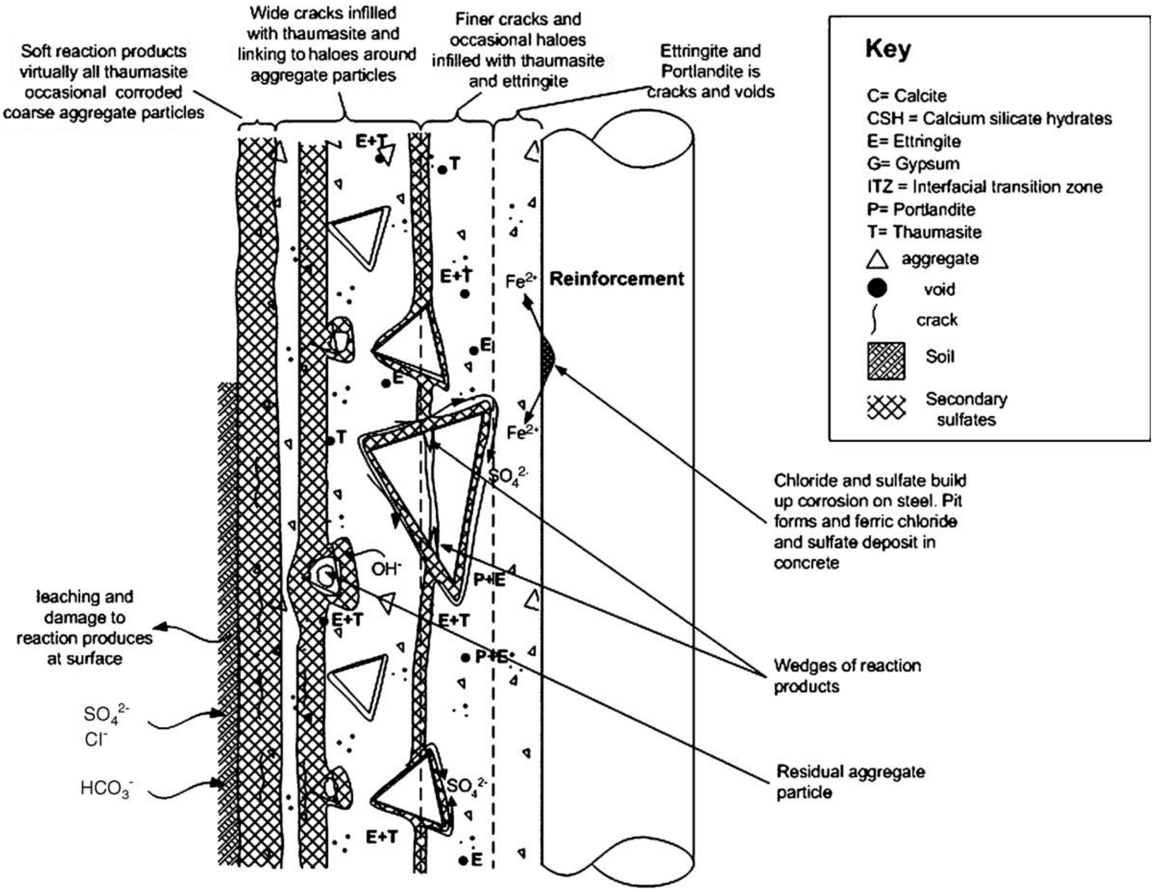

Fig. 5. Stage 4 processes.

3.4. Postulated stage 3 processes

The cement matrix continues to be weakened by loss of portlandite and decalcification and breakdown of the cement hydrates.

Thaumasite crystals grow within the cracks with their axes normal to the walls of the cracks. In large voids no disruption occurs but in small pores and cracks when the available space is used up the cement matrix expands and microcracks form sub-parallel to the concrete surface as the cracks widen in the direction of least restraint [4]. Restraint of expansion due to sulfate attack at the concrete surface by the unaffected concrete below and continued diffusion of reactants from the surface also promote sub-parallel cracking to the surface [3].

The sub-parallel cracking extends exploiting areas of weakness around the aggregate particles and in some cases penetrating the aggregate particles where this is the least path of resistance. As cracking develops a new generation of thaumasite forms filling the available space and creating 'halos' and 'wedges' of reaction product at cracks around and through the aggregate particles. The thaumasite becomes more birefringent under optical microscopy, 'massive' and 'pure' with magnesium and alumina being expelled from the material [7].

Ettringite slowly converts to thaumasite at low temperatures [9]. The proportion of ettringite and thaumasite being influenced by progressive dissolution of the cement hydrates [8] and differences in the solubility of ettringite and thaumasite at low temperature as the pH in the concrete is reduced.

The 'halos' and 'wedges' of reaction product at aggregate particles form expansion 'nuclei' which in turn lead to wide continuous cracks between the aggregate particles and expansion 'bands' [10]. The expansion is promoted by the presence of water and reactants adjacent to the concrete. Where water is available above groundwater level then blistering of the concrete develops at the concrete/ground interface. Below groundwater level, expansion and softening of the concrete is more widespread.

As the reaction proceeds, protective coatings become destabilised due to expansion and softening of the substrate, although residual layers of carbonation and coatings remain.

Additional stresses are introduced by differences in the physical properties of the attacked and unattacked concrete and dissolution and reformation of the reaction products with fluctuations in groundwater chemistry [4,11].

3.5. Postulated stage 4 processes

At the surface of the concrete the increased porosity due to leaching permits silica from the adjacent ground to promote almost full conversion of the cement paste to thaumasite [12]. The layer of reaction products expands and draws in water and reactants becoming a soft porous mass depleted in aggregate particles and susceptible to physical damage. The residual cracks become short and discontinuous as cracks fill with reaction products [13] and the rate of reaction decreases as the new reaction sites are depleted and become remote from sources of the reactants [10].

Deeper within the concrete the expansion continues due to 'wedges' of reaction products forming within cracks in the aggregate and as 'halos' around the aggregate [4]. Disruption of the concrete spreads laterally due to the development of cracking sub-parallel to the surface along the least lines of resistance. Unaffected zones of concrete exist where coarse aggregate particles 'shield' the concrete from sub-parallel cracking [7].

The expansion and crack widths reduce and there is a transition from almost pure thaumasite at the surface of the concrete to ettringite and portlandite with increasing depth.

Table 1
Postulated stage 1 processes

Postulated process	Evidence required	Observations from investigation
Groundwater brings high levels of calcium, magnesium and sodium sulfates, carbonates and chloride into contact with concrete	High levels of sulfates, carbonate and chloride in groundwater Association of sulfate with Ca, Mg and Na	Groundwater nearly saturated in calcium carbonate and calcium sulfate (at 30 years) Strong correlation between sulfate and Ca, Mg and Na in made ground (at 30 years)
Sulfates and chlorides diffuse into concrete and chloride is subject to seasonal wash-out	Sulfate and chloride profiles in the concrete show decrease from surface Chloride reduced at surface	Sulfate values show decrease from surface consistent with diffusion from surface Chloride levels close to surface reduced
Sulfuric acid developed within groundwater reacts with aggregate and cement paste	Low pH in groundwater	pH values >7 and high carbonate content in soil (at 30 years) not consistent with acid conditions
	Consumption of aggregate in concrete	Aggregate margins generally sharp from petrography
Leaching of concrete reduces calcium hydroxide and increases porosity of surface	Reduction in calcium hydroxide and increase in porosity of cement paste	Zone of reduced portlandite present in 75% of cases and significant correlation coefficient +0.72 between depth of porosity increase and reduced portlandite
Calcium and pH increase in adjacent ground and gypsum and ettringite deposited	Trends in Ca and pH in soil and gypsum and ettringite identified	Ca and pH increase towards concrete and ettringite in soil close to concrete surface
Enhanced porosity, ingress and sulfate attack adjacent to aggregate particles Ettrigite forms in these areas and in cracks	Increased porosity and reaction products next to aggregate particles and in cracks	Narrow zones of porosity sometimes associated with aggregate and microcracks Abundant ettringite often coincides with zones of locally porous cement hydrates
As calcium hydroxide is reduced the other hydration products sequentially decompose	Decomposition and recrystallisation of cement hydrates in areas depleted in calcium hydroxide and decalcification of paste	Evidence of recystallisation of cement hydrates and occasional association between reduced portlandite content and increased secondary ettringite formation
Layer of calcite, brucite and gypsum forms at surface	Layer of carbonated concrete, gypsum or brucite at surface	Superficial cabonation present in many cases and gypsum occasionally present in carbonated concrete Magnesium sporadically high consistent with brucite but no direct reference in petrography
Alkali sulfate attack	Porlandite depletion correlates with alkali sulfates	Significant negative correlation between CaO with SO_3, Na_2O and K_2O
Ingress of magnesium sulfate promotes decalcification of concrete	Trends of Ca with Mg from SEM Microprobe work	Significant negative correlation of CaO with MgO
As sulfate levels increase thaumasite forms at nucleation sites without disruption with high levels of alumina by forming a solid solution with ettringite	Thaumasite associated with trigger sulfate level Solid solutions or intermediate solid solutions with ettringite	No direct evidence for trigger level High SO_3 associated with normal SiO_2/Al_2O_3 levels consistent with presence of alumina or solid solutions with ettringite

Table 2
Postulated stage 2 processes

Postulated process	Evidence required	Observations from investigation
Chloride ingress occurs ahead of sulfates promoted by release of bound chlorides by sulfate attack	Reduction in chloride in areas of sulfate attack	No statistically significant difference between chloride content of unattacked concrete and sulfate reaction products
	Trend between chloride diffusion coefficient and sulfate attack	No correlation between depth of thaumasite filled cracks and chloride diffusion coefficient
	Chloride profile show distortion where sulfate attack present	Distortion of chloride profile where thaumasite present but not conclusive
Leaching and decalcification by alkali sulfate attack and carbonation promotes decomposition of the calcium silicate hydrates and softening of the concrete	Association between depth of decalcification and evidence of softening	No unambiguous evidence as softening often associated with sulfate attack
	Decomposition and recrystallisation of cement hydrates in areas depleted in calcium hydroxide from petrography	No direct evidence that reduced calcium hydroxide leads to decomposition of cement hydrates. Some evidence of association between reduced portlandite content and increased secondary ettringite
Sequential dissolution of cement hydrates and sulfate species leads diffusion and precipitation fronts sub-parallel to the concrete surface	Bands of secondary sulfates and recrystallised cement hydrates from petrography	Widespread evidence of recrystallisation of cement hydrates in the form of ettringite but no evidence of interspersed layers except GMRS Report 4570B [4]
Nucleation of thaumasite occurs preferentially within these mixed layers of reactants	Association of thaumasite formation with layers of recrystallised sulfates, carbonates and silicates	No evidence reported and very limited evidence of calcite and thaumasite in association
Thaumasite formation at surface of aggregate promoted by carbonate from the aggregate and de-dolomitization	Thaumasite in association with consumption of carbonate material and de-dolomitization from petrography	No unambiguous evidence of thaumasite in association with consumption of carbonate or de-dolomitzation, except GMRS Report 4570B [4] and BRE Report CR193/98 [7] Generally sharp aggregate margins not consistent with consumption of carbonate
As the thaumasite forms it becomes more dense and crystalline and incorporates additional ions	Changes in thaumasite form with time/depth from petrography	Reference to different forms of thaumasite confined to BRE Report CR193/98 [7]
	Presence of additional ions in thaumasite lattice, eg Al_2O_3,MgO or Cl from SEM microprobe	High Al_2O_3 levels in material identified in petrography as thaumasite
Ettringite and thaumasite form within voids and grow into available space but without evidence of disruption	Thaumasite and ettringite present without disruption in petrography	Ettringite ubiquitous in voids without disruption Very small amounts thaumasite present within voids without macroscopic evidence of cracking

High chloride and sulfate values at the surface of the steel reinforcement initiate corrosion. Pitting corrosion occurs without spalling of the concrete. Soft black corrosion products are deposited in the pits and ferric chlorides, ferric oxides and ferric sulfates form in the adjacent concrete.

4. Summary of findings

For each process of the postulated mechanisms of TF and TSA, the confirmatory evidence needed was established and reviewed against the observations from the investigation. The evidence for stages 1–4 has had to be based on features at different depths within the concrete as all the structures are of similar age. Furthermore, ground conditions are those measured 30 years after construction. Consequently the findings should be regarded as speculative particularly for the early stages of the process.

A summary of this review is given for stages 1–4 of the mechanisms in Tables 1–4 respectively.

5. Conclusions

The review of the evidence from the thaumasite investigation confirmed the following key issues in respect of the postulated mechanism:

- The groundwater brings high levels of chlorides, calcium sulfate and carbonate as well as magnesium and sodium sulfates into contact with the concrete. Chloride and sulfate diffuse into the concrete and chloride is reduced at the surface by seasonal wash-out.

Table 3
Postulated stage 3 processes

Postulated process	Evidence required	Observations from investigation
Thaumasite grows with axis normal to walls of the cracks	Orientation of thaumasite crystals in early stage of reaction from petrography	Evidence of well formed crystals of thaumasite orientated approximately normal to walls of voids and cracks
As space is used up expansion and sub-parallel cracking develops	Voids partially filled with thaumasite without damage	Thaumasite present in very small amount in pores or >70 mm depth in large voids in honeycombed concrete without cracking
Restraint of expansion at the concrete surface by unaffected concrete below promotes sub-parallel cracking to surface	Cracking sub-parallel to concrete surface associated with expansion from defect survey Cracks or microcracks empty of reaction products	Expansion closely associated with sub-parallel cracking Limited evidence of empty microcracks except radial to aggregate particles. Cracks in aggregate particles often empty but 'wedges' of reaction products present
A new generation of thaumasite forms filling available space and extending the sub-parallel cracking around and through aggregate particles	Generations of thaumasite crystals and microcracking from petrography	Evidence of two generations of thaumasite confined to GMRS Report 4570B [4] Limited evidence of microcracking extending from reaction sites but cracks in aggregate coincide with cracking in cement paste
Thaumasite becomes more birefringent, massive and pure with magnesium and alumina being expelled	Massive uniformly crystalline thaumasite with high birefringence from petrography	No direct reference to 'massive' thaumasite but highly birefringent thaumasite is occasionally indicated consistent with the habitat in BRE Report 193/98 [7]
	Changes in levels of alumina, chloride, sodium and magnesium from SEM microprobe work adjacent to massive thaumasite	Thaumasite at surface is pure with low levels of Al_2O_3 and MgO and the adjacent concrete often has high levels of MgO
Ettringite appears to slowly convert to thaumasite at low temperatures	Progressive reduction in ettringite and increase in thaumasite with time	No direct evidence as all structures similar age but ettringite occurs ubiquitously at depth whereas the surface of the concrete is often completely replaced by thaumasite
Expansion 'nuclei' which in turn form wide cracks and expansion bands between the aggregate particles	Expansion associated with reactions at aggregate particles from petrography	Widespread indirect evidence in form of cracking through and around aggregate particles and limited references to cracking originating at surface of aggregate particles
Expansion promoted by water and reactants adjacent to the concrete	Trends in water and reactant levels with degree of cracking and expansion	Correlation between groundwater level and expansion and absence of TSA despite high sulfate levels where water absent
As the reaction proceeds coatings become destabilised due to expansion and softening of the substrate	Reduction in attack where intact coatings present Evidence of carbonation or coatings at surface of attacked concrete	Reduction in sulfate levels and severity of attack where coatings present Fine sub-parallel cracking and thaumasite occasionally below traces of coatings and superficial carbonation at surface of TSA affected concrete
Additional stresses are introduced by differences in the physical properties of the attacked and unattacked concrete	Significant difference in physical and chemical properties of the attacked and unattacked concrete	Rebound hammer in TSA affected concrete typically <10 compared to >30 in unattacked concrete
Reformation of the reaction products with fluctuations in the groundwater chemistry	Evidence of seasonal trends in groundwater level and chemistry and reformation of reaction products	Reaction products and unattacked concrete have statistically significant different levels of SO_3, CaO, Al_2O_3 and MgO Changes in groundwater chemistry (at 30 years) linked to seasonal recharge

- The concrete is subject to leaching leading to a reduction in portlandite, an increase in the porosity of the concrete and the deposition of gypsum and occasionally ettringite in the soil. The porosity of the concrete increases adjacent to aggregate particles promoting ingress and reactions in these areas.
- Ettringite formation gives way to thaumasite formation. Expansion and cracking develops sub-parallel

Table 4
Postulated stage 4 processes

Postulated process	Evidence required	Observations from investigation
Surface of the concrete becomes almost fully converted to thaumasite promoted by silica from the ground	Presence of thaumasite at external surface of concrete from petrography where there is intimate contact with the soil	Widespread evidence from petrography of thaumasite completely or partially replacing the cement paste at the external surface but no conclusive evidence of association with intimate contact with soil
The layer of reaction products expands and draws in water and reactants becoming a soft porous mass	High water and sulfate content in reaction products	Sulfate levels in reaction products exceed 6% by mass of sample and there is qualitative evidence that deposits are soft and have a high moisture content
	Expansion associated with softening from defect survey	Significant correlation coefficient of +0.39 between expansion and depth of softening
The residual cracks become discontinuous as cracks fill with reaction products	Short discontinuous cracks at surface from petrography	Widespread evidence of cracking becoming hidden or discontinuous at surface
The rate of reaction reduces as new reaction sites are depleted and become remote from reactants	Trends in sulfate, moisture content and evidence of reaction with time and depth	Sulfate content generally decreases with depth below external surface although level increases locally at cracks infilled with thaumasite Width and frequency of cracks infilled with thaumasite decreases remote from the concrete surface
'Halo's and 'wedges' of reaction products form where cracks go around or through aggregate particles and the cracking widens	Presence of wedges of reaction products within cracking through aggregate particles from petrography	Halos of reaction products are common around aggregate particles Wedges of reaction products are associated with wide 'empty' cracks through the aggregate particles
Disruption of the concrete spreads laterally due to the development of sub-parallel cracking	Disruption closely associated with continuous sub-parallel cracking	Evidence that reaction largely confined to cracks which extend sub-parallel to the surface rather than propagating from the surface
Unaffected concrete exists where coarse aggregate particles shield the concrete	Unattacked areas associated with aggregate particles from petrography	No evidence for shielding although larger aggregate particles appear to restrict reaction to cracks around and through particles leaving adjacent areas relatively free of cracking and thaumasite
High chloride and sulfate values at the surface of the steel reinforcement initiate corrosion	Association of pitting corrosion with a critical combined level of sulfate and chloride	Chloride values of 1% at steel indicate significant risk of corrosion due to chlorides alone
Pitting corrosion occurs without spalling of the concrete and soft black corrosion products are deposited in the pits and ferric chlorides, oxides and sulfates deposit in the adjacent concrete	Evidence of pitting corrosion from defect survey	Limited incidences of pitting corrosion associated with black, green and white products consistent with ferric oxide, chloride and sulfate
	Highly negative half-cell potentials in areas of pitting	Half-cell potentials not measured

to the surface where available space is filled with reaction products.

- Different generations of thaumasite form as cracking develops and new space becomes available. The thaumasite becomes more birefringent under optical microscopy, massive and pure with magnesium and alumina being expelled into the adjacent concrete.
- Thaumasite forms as 'halos' and 'wedges' at cracks passing around and through the aggregate particles and widens the cracking.
- The degree of softening, cracking and expansion is influenced by the availability of water.
- High chloride levels at the steel initiate pitting corrosion to the reinforcement and ferric chloride, oxide and sulfate are formed in the adjacent concrete.

6. Recommendations

There remain a number of uncertainties regarding the timing and precise sequence of steps in the reaction and the relative importance of different factors. In particular, there is no conclusive evidence of sulfuric acid attack and only limited evidence for the contribution of carbonates from the aggregate, including de-dolomitization.

It is recommended that that following aspects of TF and TSA should be investigated:

- possible contribution of low pH and sulfuric acid developed in the ground,
- relative importance of potential source of carbonates for the reaction,
- precise process by which cracking forms sub-parallel to the surface,
- process of conversion of ettringite to thaumasite,
- occurrence and relevance of different forms of thaumasite,
- factors in the progress of the reaction, eg role of magnesium ions,
- dual effect of sulfate and chloride ions and the combined threshold level for steel corrosion.

Acknowledgement

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