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Influence of cement and aggregate type on thaumasite formation in concrete

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ABSTRACT

In this study the influence of binder type on the formation of thaumasite in mortar prisms made with expanded clay lightweight aggregate (LWA) or quartz sand was examined. For this purpose mortar prisms were made, which after 28 days of curing in deionised water were exposed to a sulphate solution or deionised water. The length and weight change of the prisms was recorded in triplicate as a function of time of exposure to dry–wet cycles at 5 ± 1 °C.

The influence of the binder type on the expansion in the sulphate solution can be ordered from strong to weak as follows: (1) CEM I + limestone filler, (2) CEM I, (3) CEM I + fly ash, and (4) CEM III/A. Because the porosity of the LWA was able to accommodate the growing sulphate crystals, the mortar prisms made with LWA were still largely intact after 3 years of exposure. The only exception being the mortar prisms containing limestone filler. The mortar prisms made with quartz sand and exposed to the sulphate solution were all bent, broken or disintegrated after 24 weeks. The prisms exposed to deionised water showed minimal expansion. Key factors controlling the formation of thaumasite are discussed.

1. Introduction

Conventional sulphate attack in mortars and concretes involves the formation of expansive sulphate phases like ettringite ($Ca_6Al_2(SO_4)_3(OH)_{12}\cdot 26H_2O$) and gypsum ($CaSO_4\cdot 2H_2O$). However, at low temperatures and in concrete containing a source of carbonate, the formation of thaumasite ($Ca_3Si(OH)_6(CO_3)(SO_4)\cdot 12H_2O$) can be observed.

In Portland cement-based materials the thaumasite form of sulphate attack (TSA) requires a source of sulphate, including sulphide that can be oxidised to sulphate, and carbonate (CO_3^{2-}) or bicarbonate (HCO_3^-) in the presence of excess calcium ions and mobile water, at temperatures below 15 °C, and particularly between 0 and 5 °C [1,2]. TSA can be particularly deleterious because the main calcium silicate cementing (C–S–H) phases are affected, which can lead to complete loss of cohesion and strength [3].

The factors affecting thaumasite formation have not yet been fully clarified [2,4–6]. The work of several authors, summarised by Crammond [7], suggest that thaumasite will generally only form at a pH > 10.5. This is also the pH range at which the concentration of CO_3^{2-} dominates over that of HCO_3^- and could possibly point to the importance of CO_3^{2-} in the formation of thaumasite.

In the present work the effects different binders may have on sulphate attack and especially on the formation of thaumasite in mortar prisms made with expanded clay lightweight aggregate (LWA) or quartz sand are examined. Based on these effects the factors controlling thaumasite formation are discussed.

2. Materials and methods

2.1. Materials

Different cements and cement blends were tested: (1) CEM I 52.5 N, (2) 80% CEM I 52.5 N + 20% limestone filler (CALCITEC 2001 S), (3) 80% CEM I + 20% fly ash and (4) CEM III/A 42.5 N LA. The annotation LA specifies that the alkali content of the CEM III/A is lower than 0.9%. The chemical composition of the fly ash is given in Table 1. The chemical characteristics and grain size of the fly ash were analysed following ASTM C618 standards [8]. The chemical parameters and grain size of this fly ash meets the specifications of "class F fly ash" (Table 2).

To examine the role the expanded clay lightweight aggregate (LWA) may have on the formation of thaumasite, prisms were

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Table 1
Chemical composition of fly ash (semi quantitative XRF screening).

Fly ash	CaO	SiO_2	Al_2O_3	Fe_2O_3	MgO	Na_2O	K_2O	SO_3
Weight % dm	4.8	42.9	29.6	5.4	1.0	1.3	1.1	0.7

made with LWA or quartz sand (QS). The LWA used in this study is produced by Argex nv and sold under the name AG 0/4–500 N. AG 0/4–500 N is the sand sized fraction (0–4 mm) of crushed expanded clay aggregates with a density of 518 kg/m^3 dry weight. The quartz sand used is a nitric acid washed pure quartz sand. The grain size distribution of the Argex lightweight aggregate and the quartz sand are presented in Fig. 1.

2.2. Preparation of test samples

The mortars tested in this study were produced following the methods of the European Committee for Standardization EN 196-1 [9], unless stated otherwise. The mortars containing the LWA were made with 375 g cement or cement blend (see Table 3) and 940 g LWA. The water to binder ratio was approximately 0.55. The water used to pre-moisturize the LWA (40 wt.%), was not taken into account. The reference mortars were made with 375 g cement or cement blend and 1500 g quartz sand (see Table 3). The water to binder ratio was approximately 0.55.

After casting of the mortars in moulds with dimensions of $20 \times 20 \times 160$ mm^3, the moulds were covered with plastic wrap to prevent loss of water by evaporation. After approximately 24 h, the mortar prisms were demoulded and water cured in demineralised water to the age of 28 days.

2.3. Methods

2.3.1. Accelerated carbonation

After the 28 day curing step a selection of samples were carbonated. Carbonation occurred by exposing the samples in a batch reactor to a pCO_2 of 0.25 bar at 1 bar total pressure during 1 week. The pressure was monitored manually and drops in the pressure of the reactor due to carbonation of the mortar prisms were regularly compensated by replenishing the atmosphere of the reactor with pure CO_2.

2.3.2. Ageing procedure

After curing (and accelerated carbonation) the mortar prisms were either immersed in a 2.37% sodium sulphate solution or in deionised water at 5 ± 1 °C. The temperature of 5 ± 1 °C was chosen to accelerate the formation of thaumasite. For each formulation 4 mortar prisms were immersed in 1.5 l of solution. The liquid to solid ratio was approximately 3.5.

Wet and dry cycling was applied because the accompanying degradation is normally faster than that associated with continuous immersion. A cycle duration of 3 weeks wet followed by 1 week drying was applied during the first 170 days, thereafter the immersion period was prolonged to 8 weeks followed by a 1 week of drying. After drying, the samples were immersed in fresh solutions of sodium sulphate or deionised water. For the prisms immersed in deionised water the solution was no longer renewed after 34 weeks. This was done since dissolved sulphate (e.g. due to oxidation of sulphides present in the LWA) may be lost during refreshing of the solution.

Table 2
Chemical (and physical) composition of the fly ash analysed according to ASTM C618 standards.

	Fly ash composition (%)	Requirements class F fly ash (%)
Chemical		
$SiO_2 + Al_2O_3 + Fe_2O_3$	78	Min 70
SO_3	0.3	Max 3
Moisture content	0.3	Max 3
Loss on ignition (LOI)	4.7	Max 6
Physical		
Fineness + 325 mesh	13	Max 34

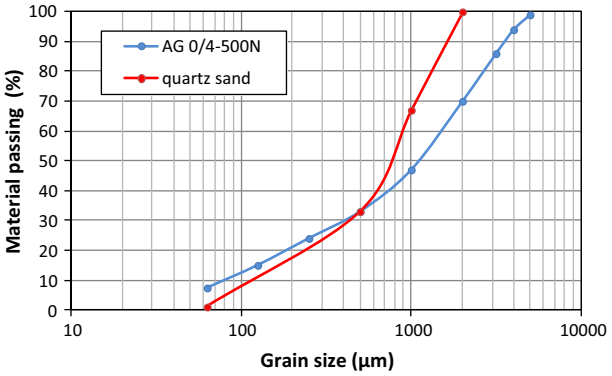

Fig. 1. Grain size distribution of the Argex lightweight aggregate and quartz sand. The grain size distribution was obtained by dry sieving of the aggregates in the lab with standard sieves.

2.3.3. Expansion measurements

The prisms were made with stainless steel inserts cast into their ends to facilitate accurate monitoring of length changes by the use of a length comparator. At the end of each dry cycle the length and weight of the mortar prisms was measured following CUR recommendation 48 [10].

2.3.4. Porosity determination

The porosity was estimated by determination of the water absorption of the mortar prisms. For determination of the water absorption, the samples were oven dried (105 °C). The air in the pores of the mortar prisms was evacuated using a vacuum pump. When vacuum (240 Torr) is reached, water is allowed to fill the pores during 24 h to reach full saturation. The weight of the fully saturated mortar prisms was measured under water (W_{SW}) and in air (W_{SA}). The total porosity (P) was calculated as $P = \frac{W_{SA}-W_d}{W_{SA}-W_{SW}} 100$, with W_{SA} the water saturated weight in air, W_d the dry weight and W_{SW} the water saturated weight under water.

Table 3
Materials and formulations of the mortar prisms (LWA = expanded clay lightweight aggregate, QS = quartz sand, LF = limestone filler, FA = fly ash).

Coding	Binder	Aggregate or sand
CEM I/LWA	375 g CEM I 52.5 N	940 g expanded clay
CEM I/QS	375 g CEM I 52.5 N	1500 g quartz sand
CEM I + LF/LWA	300 g CEM I 52.5 N + 75 g limestone filler(LF)	940 g expanded clay
CEM I + LF/QS	300 g CEM I 52.5 N + 75 g limestone filler(LF)	1500 g quartz sand
CEM I + FA/LWA	300 g CEM I 52.5 N + 75 g fly ash (FA)	940 g expanded clay
CEM I + FA/QS	300 g CEM I 52.5 N + 75 g fly ash (FA)	1500 g quartz sand
CEM III/LWA	375 g CEM III/A 42.5 N LA	940 g expanded clay
CEM III/QS	375 g CEM III/A 42.5 N LA	1500 g quartz sand