



Influence of chlorides on magnesium sulphate attack for mortars with Portland cement and slag based binders



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HIGHLIGHTS

- Influence of Cl^- on MgSO_4 attack is temperature and binder type dependent.
- Cl^- aggravates MgSO_4 attack when blast-furnace slag is used.
- At 5 °C chlorides in MgSO_4 environment increase deterioration regardless binder type.
- At 5 °C thaumasite degradation in presence of Cl^- and MgSO_4 is severe in all cases.

ARTICLE INFO

Article history:

Received 2 February 2017

Received in revised form 7 July 2017

Accepted 29 July 2017

Keywords:

Concrete
Mortar
Chloride
Sulphate
Magnesium
Combined attack
Blast-furnace slag

ABSTRACT

Sulphate containing environments are aggressive and cause damage to cementitious materials by means of cracking, spalling and strength loss. In realistic conditions, sulphates may be found in combination with e.g. chlorides. Therefore, to predict concrete's durability, it is important to understand the (combined) attack mechanisms and estimate their effect.

This study aimed to elucidate especially the influence of chlorides on magnesium sulphate attack at two different relevant temperatures (5 °C and 20 °C) and for mortars with different binders, including ordinary Portland cement (OPC), high-sulphate resistant Portland cement (HSR) and a binder composed of 50% blast-furnace slag (BFS) and 50% ordinary Portland cement. Mass change measurements were performed to examine the influence of Cl^- on MgSO_4 attack, and XRD-analyses to identify phase changes.

It can be concluded that the influence of Cl^- on MgSO_4 attack is temperature and binder type dependent. The presence of chlorides does not affect deterioration due to magnesium sulphate for OPC and HSR at an environmental temperature of 20 °C, however, it increases the degradation when BFS is used as partial binder and/or when temperature decreases to 5 °C. The presence of BFS decreases the formation of a protecting brucite layer and favours decomposition of calcium silicate hydrates to magnesium silicate hydrates. At 5 °C the degradation involves thaumasite formation, and is more equal for the different binders and quite severe.

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1. Introduction

When water contaminated with sulphates penetrates into concrete by means of diffusion or capillary suction, severe deterioration such as cracking, spalling, increased permeability and strength loss occurs. Nevertheless, it is a long-term process. Quite some literature is found about single sulphate attack, mainly tested by means of sodium sulphate or magnesium sulphate. Notwithstanding, in realistic environments sulphate attack occurs con-

comitantly with other deterioration mechanisms such as chloride ingress. It is remarkable that few literature is found about combined attack mechanisms. This paper focuses on the influence of chlorides on magnesium sulphate attack for different types of mortar at different environmental temperatures. In a previous paper by the authors [1], the influence of chlorides on sodium sulphate attack is described.

1.1. Single sulphate attack

Sulphates (SO_4^{2-}) will chemically bind with the hydration products of the binder, namely calcium silicate hydrate (CSH), calcium hydroxide (CH) and calcium aluminate hydrates (C_3A). These

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chemical reactions yield the following reaction products: secondary gypsum, secondary ettringite, thaumasite and brucite [2].

Gypsum is a reaction product from CH or CSH, SO_4^{2-} and water. It is generally assumed that its formation leads to reduction of stiffness and strength, expansion and cracking and eventually to transformation of the material into a mushy and non-cohesive mass [11]. In addition, secondary ettringite yields from the reaction between SO_4^{2-} , Ca^{2+} and monosulphate ($\text{C}_4\text{ASH}_{12}$). It has the ability to swell strongly, which results in a densification of the microstructure followed by internal stresses that lead to cracking and destruction of the concrete [3]. Whether gypsum formation results in expansion is disputed in literature [4]. Roziere et al. [5] and Santhanam et al. [6] mention that ettringite as well as gypsum have an expansive and destructive character, while Schmidt et al. [7] claim that the contribution of gypsum is limited while the expansion of ettringite dominates. As described by Kunther et al. [8], deterioration due to expansion of the reaction products will only occur when there is a coexistence of ettringite and gypsum. Next to the volume increase, which is prerequisite for expansion, also supersaturation in the pore solution and the stresses exerted by the formed minerals are decisive. In addition, thaumasite formation leads to strength loss and decomposition of the microstructure. Thaumasite is formed as sulphates or ettringite react with CSH, CO_3^{2-} , Ca^{2+} and excess water. It is generally assumed that thaumasite is formed at temperatures below 15 °C [11].

In the specific case of magnesium sulphate attack, as schematically shown in Fig. 1, the interaction between hydrated cement paste and a MgSO_4 solution is a reaction between this salt and CH of the paste, yielding gypsum as well as brucite, $\text{Mg}(\text{OH})_2$ [3,9]. Brucite is practically insoluble and its formation lowers the pH of the pore solution which destabilizes CSH. Ultimately, this phase converts to an amorphous hydrous silica (SiO_2 – silica gel) or to a magnesium silicate hydrate (MSH) [2,10,11]. In addition, CH is released in the solution. CH reacts again with MgSO_4 and the process proceeds [9]. Mg^{2+} can also replace Ca^{2+} from CSH and form MSH [3,9,10]. MSH causes strength loss and expansion while brucite forms a protective layer, which slows down the degradation process [12–14].

The rate of external sulphate attack is largely influenced by the accessible porosity, porous microstructure and permeability of the concrete, the sulphate concentration in the external medium in contact with the concrete, the C_3A content of the cement and the

use of additions. In turn, these factors are influenced by the type of cation, pH, and temperature of the medium.

Concerning the cation type, it seems that this also has an influence on the OH^- concentration. E.g. Na_2SO_4 and MgSO_4 have opposite effects on the pore solution alkalinity. Na_2SO_4 increases the alkalinity while MgSO_4 reduces it. The latter has been shown recently by De Weerd et al. [15].

With regard to the effect of the binder type, Gollop and Taylor [17–21] showed that the replacement of cement by slag reduces the degradation due to sulphate attack and attributed this positive effect mainly to the dense microstructure. The latter is confirmed by other researchers [2,16]. Brown et al. [16] showed the effectiveness of BFS in case of Na_2SO_4 and MgSO_4 solutions, other researchers e.g. Higgins [13] and Taylor [22] only mentioned a higher effectiveness of BFS in case of exposure to Na_2SO_4 solutions. Contrarily, the effect of partial replacement of OPC is not always positive. According to Rasheeduzzafar et al. [23] deterioration in BFS samples exposed to MgSO_4 solutions exceeds the deterioration observed in OPC samples. Because the CH is consumed by the pozzolanic reaction, the sulphates and the magnesium ions will react directly with the CSH resulting in a cohesionless MSH [24]. Furthermore, it is clear from literature that thaumasite formation can be favoured in case of partial cement replacement by BFS [25]. The damage to concrete due to thaumasite formation is usually profound and is mainly due to the decomposition of the CSH phase, resulting in a loss of strength of the material.

1.2. Combined attack of chlorides and sulphates

According to Al-Amoudi et al. [26] the presence of chlorides may intensify or mitigate sulphate attack, or it may have an insignificant effect. They have added high volumes of sodium chloride (15.7% Cl^-) to mixed sodium and magnesium sulphate solutions, with sulphate concentrations of 2.1% SO_4^{2-} in which half of the sulphates came from sodium sulphate and the other half from magnesium sulphate. While monitoring expansion and strength reduction, they noticed that the deterioration was reduced by the addition of chlorides to the sulphate solution. The cement type did not have a major effect for cements with C_3A contents varying between 3.5% and 8.5%. Replacement of part of the OPC by BFS had a marginal beneficial effect. Abdalkader et al. [27] performed tests on mortars at 5 °C immersed in solutions containing 6 g/l SO_4^{2-}

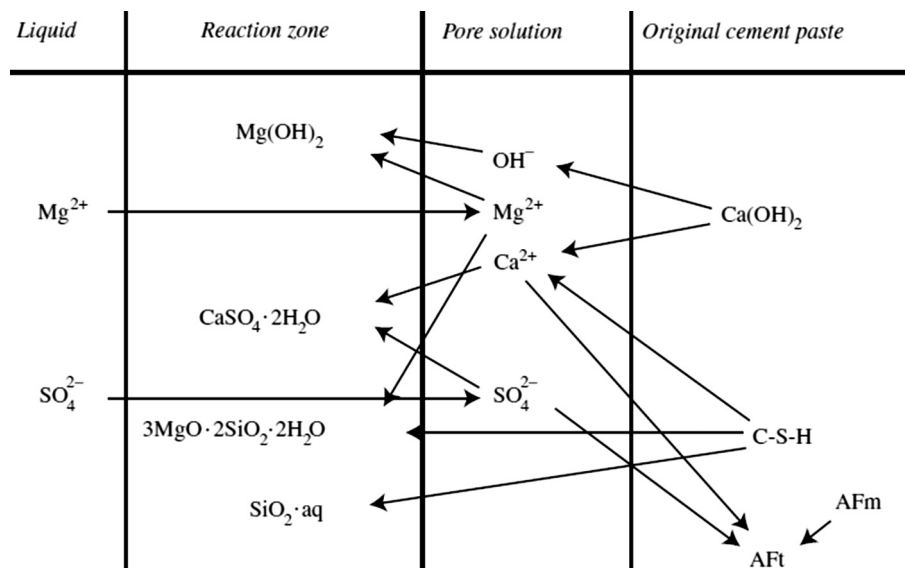


Fig. 1. Reactions taking place between Portland cement components and magnesium sulphate solution [12].

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