



Chemo-mechanical modelling of the external sulfate attack in concrete



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ARTICLE INFO

Article history:

Received 29 August 2016
Received in revised form 14 November 2016
Accepted 21 December 2016
Available online xxxx

Keywords:

Concrete
Sulfate attack
Delayed ettringite formation
Damage

ABSTRACT

This paper is focused on the modelling of the mechanical consequences of external sulfate attack in concrete structures under partially or fully saturated conditions. To this purpose a weakly coupled approach is developed: first the moisture content is computed through a simplified diffusion model, then a reactive-diffusion model allows for the computation of the expansive products of the reaction occurring between the aluminates of the cement paste and the incoming sulfate ions, finally the solution of a nonlinear mechanical problem gives the expansion, the stress state and the degradation induced by the reaction. The mechanical problem makes use of a multiphase elasto-damage model, developed in this work and accounting for both chemical and mechanical damage. The model is validated by simulating various experimental tests on concrete specimens subject to external sulfate attack and then applied to the simulation of a reduced scale structure of a tunnel lining.

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1. Introduction

The sulfate attack in concrete consists of a complex set of reactions between sulfate ions (coming from the external environment or released by the cement paste after high heating) and the hydrate calcium aluminates present in the cement paste. In the first case the phenomenon is called External Sulfate Attack (ESA), while in the second case the phenomenon is called Internal Sulfate Attack (ISA) or Delayed Ettringite Formation (DEF). In both cases one of the final reaction products is the secondary ettringite that, forming within the hardened matrix, can generate swelling and microcracks formation inside the material. The mechanical effects on the structural elements can be very serious up to compromise the reliability of the entire construction [1,2]. When a concrete structure is located in a geological site naturally rich in sulfates the phenomenon cannot be avoided and one can only try to mitigate the mechanical consequences of the phenomenon of reducing the permeability, by using low-aluminates cements or by isolating the external surfaces.

In the literature there are many works focused on the description of the chemical mechanisms and of the mechanical consequences of sulfate attack. Although there is no unanimous agreement on the mechanisms that cause the expansion of the material [3], many authors agree on attributing it to the formation of secondary ettringite [4–8]. The rate of reactions and the consequent mechanical

response of material depends on environmental factors (species and concentration of sulfate, pH of the solution, humidity, temperature) and intrinsic material properties (w/c ratio, chemical composition of the cement paste, pore distribution, diffusivity properties). Many experimental campaigns have been conducted on mortar and concrete to characterize specific features of the phenomenon, see e.g. [9–13].

In ESA the sulfates penetrate within the porous network of the material which can be initially in partially saturated conditions. The variation in time of the moisture content influences the diffusion of sulfates. The computation of the amount of the reaction expansive products therefore requires a proper diffusion-reaction model, accounting for the variation of the degree of saturation, while a mechanical model is required for the prediction of swelling and material damage.

The mechanical model can be formulated in different ways. In [5] the volumetric expansion is treated as an eigenstrain and the mechanical response is modeled by a simple uniaxial stress-strain law. In [6] a micromechanical model based on the Eshelby solution of the equivalent inclusion method is implemented to determine the eigenstrain generated by the formation of ettringite crystals in cement paste. In [14] the expansion is related to the crystallization pressure, generated by crystals growing in the pores of the hardened paste. In [15] concrete affected by ESA is modeled at the meso-scale as a two phase composite constituted by aggregates and reactive cement matrix and the degradation is described by cohesive-crack interface elements. Most of the proposed models does not take into account the variation of moisture conditions. Very seldom these models are applied at structural level.

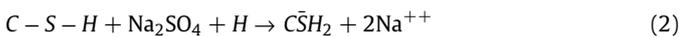
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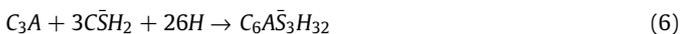
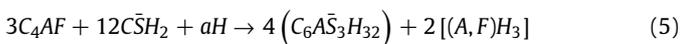
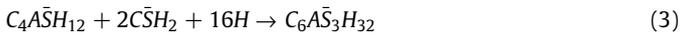
The present work aims to develop a reliable approach to be used in structural analyses to simulate the mechanical effects of ESA, also accounting for partially saturated conditions. To this purpose the moisture content is computed through a simplified diffusion model, [16]. The reactive-diffusion model proposed in [5] and further developed in [15], modified to account for partially saturated conditions, is then used to compute the sulfate molar concentration and the amount of formed ettringite. The ettringite formation implies a volume increase and, once the initial porosity is filled, it induces a volumetric deformation. In the context of the Biot's theory of porous media, and similarly to that proposed in [17–19] in the case of alkali silica reaction, the concrete subject to sulfate attack is represented as a multi-phase medium. Two phenomenological isotropic damage variables describe the chemical degradation and the stress-induced degradation. The model has been validated by simulating the experimental tests in isothermal conditions presented in [13] and [20], and then used to simulate the behaviour of a reduced-scale model of a tunnel lining reported in [1].

2. Chemical framework and mechanical consequences

The sulfate attack of concrete manifests itself through a set of topochemical reactions occurring between the hydrated products of cement and the sulfate. In the most common case the sulfates anions come from the ionic dissociation of sodium sulfate through reactions with the portlandite CH or the gel $C-S-H$ leading to the formation of gypsum $C\bar{S}H_2$ [21], [4], [5], [15]. The initial process can be defined by the reactions:



Then, the reactions between the gypsum and the hydrated and unreacted aluminates lead to the formation of ettringite $C_6A\bar{S}_3H_{32}$:



In reactions (1)–(6) the italic letters refer to the cement notation defined in Table 1.

The amount of secondary ettringite formed through reactions (1)–(6) depends on the composition of the cement and in particular on the amount of tricalcium aluminate initially present. Several

Table 1
Raw materials and clinker components: chemical formula and cement notation.

Principal raw materials	Chemical formula	Cement notation
Water	H ₂ O	<i>H</i>
Sulfur trioxide	SO ₃	<i>S</i>
Calcium oxide	CaO	<i>C</i>
Aluminium oxide	Al ₂ O ₃	<i>A</i>
Silicon dioxide	SiO ₂	<i>S</i>
Iron oxide	Fe ₂ O ₃	<i>F</i>
Clinker components	Chemical formula	Cement notation
Tricalcium silicate	(CaO) ₃ SiO ₂	<i>C₃S</i>
Dicalcium silicate	(CaO) ₂ SiO ₂	<i>C₂S</i>
Tricalcium aluminate	(CaO) ₃ Al ₂ O ₃	<i>C₃A</i>
Tetracalcium aluminoferrite	4CaO • Al ₂ O ₃ • Fe ₂ O ₃	<i>C₄AF</i>

experimental campaigns [11], [10] have shown that cements with lower content of C_3A are significantly less affected by the delayed ettringite formation.

The sulfate attack causes the overall expansion of concrete and the formation of microcracks with progressive reduction of strength and stiffness and possible macro-cracking and spalling of the material.

This phenomenon is mainly due to two interacting effects. The first effect is the dissolution of calcium hydroxide and gel $C-S-H$ caused by the reaction with the sulfates. The gel $C-S-H$ is the main responsible of strength and stiffness in the hardened concrete and its dissolution with calcium leaching causes micro-cracks formation. A similar phenomenon is the decalcification of calcium hydroxide by the interstitial solution poor in calcium. Calcium hydroxide does not contribute to the mechanical strength of concrete but its consumption causes an increase in porosity facilitating the migration of aggressive species inside the material. The second main phenomenon is the formation of expansive ettringite in the pores. This phenomenon is more relevant in ESA where the penetration of sulfate, and the resulting reaction, take place progressively: the outer layers react before and tend to expand. In this region the solid skeleton is subject to a prevailing compression state. The inner layers instead, in which the reaction has not yet occurred, constrain the expansion of the outer layers and are therefore subject to tensile stress which can exceed the material strength and cause cracks formation.

Leaching and expansion due to the ettringite formation are two phenomena strongly interacting. Microscopic observations on mortar samples subject to a combination of leaching and ettringite formation, [22], show that three distinct zones can be identified, as schematically sketched in Fig. 1. The first zone is characterized by diffuse microcracking due to leaching and by ettringite formation. In the second zone there is no leaching and no visible damage is present, but there is delayed ettringite formation. This region tends to expand with respect to the inner part and it is therefore subject to compressive stress. In the third zone the reaction is not yet developed and the material is cracked due to tensile stress arising for compatibility of deformation with the outer reactive layer.

3. Chemo-elastic damage model in fully saturated conditions

3.1. Porous medium

Within the theory of porous materials [23], at the mesoscale the reacting material can be represented by a superposition of two phases: the homogenized concrete skeleton (including the nonreactive material (u) and the reactive part of solid matrix (c)) and the homogenized fluid (including water (w), sulfates (s) and

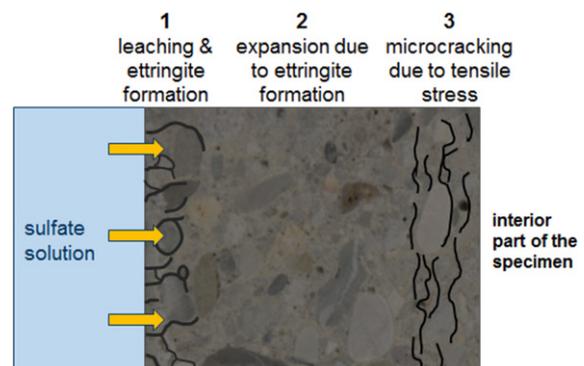


Fig. 1. Schematic view of the degradation and formation of three zones in a specimen affected by ESA.

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