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## Durability of self-compacting concrete containing pumice and zeolite against acid attack, carbonation and marine environment



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#### HIGHLIGHTS

- $\bullet$  The substitution of 10% of OPC by pumice increases compressive strength ( $f_c$ ) at both early and long term ages.
- SCC with 15% of pumice and zeolite exhibit higher resistance to sulfuric acid attack.
- The load loss by splitting can be a good indicator for evolution of chemical resistance.
- The SCC mixture based on the other mixture zeolite is much more vulnerable to carbonation compared to other mixtures.

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#### ABSTRACT

Due to their fluidity and simplicity of use, self-compacting concretes (SCCs) have undeniable advantages. Since the study on the behavior of SCC containing pumice or zeolite with respect to aggressive media as acid attack and carbonation is very rare in literature, the purpose of this study is to focus on this subject. In this study, the influence of the partial substitution of 10% and 15% of Portland cement by pumice and zeolite admixtures on the fresh state, compressive strength of self-compacting concrete (SCC) is investigated. For durability, resistance to acid attack, carbonation and marine environment is studied. The results show different behaviors depending on the nature of the pozzolan. SCCs containing pumice exhibit a compressive strength at least equal to that of control concrete at early ages and higher than the ones of the control mix at long term ages. In contrast, SCC based on zeolite has a lower resistance compared to the control concrete due to the presence of a large porosity formed during the preparation of the mixture. This study shows and quantifies the positive effect of the partial substitution of Portland cement by 10% and 15% of pumice and 15% of zeolite on improving chemical resistance in acid attack. The carbonation study showed a lower resistance of SCCs containing pozzolan, this effect is more important for SCC with zeolite. Additional studies on the formulation and the properties at the fresh state of concrete containing zeolite in order to improve the mechanical strength will surely make it possible to obtain better performance and thus better profits from this natural pozzolan.

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#### 1. Introduction

Concrete is the most commonly used building material throughout the world. Due to the difficult placement and vibration between the steel bars of reinforced concrete and the defects it may cause, the use of self-compacting concrete (SCC) is becoming more and more widespread.

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Concrete infrastructure members can be degraded by the attack of sulfuric acid present in groundwater, such as chemical wastes generated by the oxidation of sulfur compounds (e.g., pyrite) in the embankment. The effect of sulfuric acid on concrete is more harmful than that of sulfate attack because in addition to attack by sulfate ions, there is a dissolving effect caused by hydrogen ions [1,2]. The deterioration of concrete due to sulfuric acid can generally be characterized by the following reactions [3]:

$$Ca(OH)_2 + H_2SO_4 \rightarrow CaSO_4 \cdot 2H_2O \tag{1}$$

$$CaSiO_2 \cdot 2H_2O + H_2SO_4 \to \ \ CaSO_4 + Si(OH)_4 + H_2O \eqno(2)$$

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$$3\text{CaO} \cdot \text{Al}_2\text{O}_3 \cdot 12\text{H}_2\text{O} + 3(\text{CaSO}_4 \cdot 2\text{H}_2\text{O}) + 14\text{H}_2\text{O}$$

$$\rightarrow 3\text{CaO} \cdot \text{Al}_2\text{O}_3 \cdot 3\text{CaSO}_4 \cdot 32\text{H}_2\text{O}$$
(3)

According to these reactions, gypsum and ettringite are produced. Consequently, the following effects can be created:

- Dissolution of portlandite initiates for a pH below 12.5.
- The expansion of concrete (considering that the volume increase by the ettringite is greater than that of the gypsum).
- Tensile stresses are induced in the concrete (cracking in concrete).

One of the possible reactions in concrete is the conversion of the calcium present in the soluble products. This reaction leads to the decalcification of calcium silicate hydrate (CSH) in the concrete, which then causes a progressive opening of the structure and reduction of pH. In this way the surface of the concrete is mechanically weakened thus, the passivation and consequently the corrosion of the reinforcing steel bars is quite possible [4]. Effectively, decalcification contributes to softening and loss of overall strength [3]. The aggressiveness of the attack depends on the different elements such as the solubility of the salts formed due to the production of the acid-base reaction such as calcium, aluminum, iron and magnesium salts, the aggressive acid species and the pH of the solution attack [5].

The mechanism induced by degradation is due to the concentration gradient between the aggressive waters and the pore solution of the concrete. The flux of transported ions will cause, on one hand the dissolution of the hydrates previously formed and, on the other, the precipitation of the new compounds leading to the progressive degradation of the concrete [6]. The main leached species are therefore calcium Ca<sup>2+</sup> and hydroxide OH<sup>-</sup> ions [7]. Their transport to the external environment will allow the modeling of the diffusion phenomenon [8]. Indeed, leaching is a slow reaction which begins with the dissolution of portlandite, followed by decalcification of monosulfoaluminates, ettringite and C-S-H [9]. It should be noted that the leaching of concrete from structures can be carried out according to different processes [6]:

- Surface leaching.
- Dissolution of solid phases.
- Transport of dissolved species under the effect of a concentration gradient.

Bassuoni and Nehdi have worked on the chemical resistance of variable SCCs against sulfuric acid attack in two variable phases with the same initial concentration of 5% [1]. In Phase I, the pH was set to an initial value of 0.9 and the maximum threshold value of 2.5 thereafter during six immersion weeks. The solution was renewed at the beginning of phase II, but the maximum pH threshold was set at 1.0 until the end of the test (twelve weeks of immersion). According to their results, in the Phase II exhibition the role of calcium hydroxide content decreased, besides the important contribution of decalcification of C-S-H seemed to be the determining factor in weight loss. In the Phase II exhibition, the increase in the volume fraction of coarse aggregates is effective to decrease the weight loss of SCC mixtures exposed to sulfuric acid attack due to reducing available cement surface for reaction with the acid. There is a lack of literature concerning the effect of mineral admixtures on the resistance of cementitious materials in acidic media, some essential works are cited in the following:

Roy et al. studied the effect of silica fume, metakaolin, and fly ash on the resistance of mortars to acid attack [10]. According to this study, chemical resistance is significantly improved with silica fume than with metakaolin. In turn, the latter gives better resistance than the fly ash and decreases as the level of substitution

increases from the 0–10% level to the 15–30% level. In the study conducted by Oueslati [6], the admixture of silica fume and fly ash did not present a notable improvement in resistance to acid media. According to the literature review, silica fume generally showed contradictory results with respect to H<sub>2</sub>SO<sub>4</sub> attack. The admixture of metakaolin makes it possible to increase the compressive strength of concrete [11,12] and to improve the chemical resistance in front of acid attack [13]. The reaction products in mix containing metakaolin are much more chemically stable [14] and the ITZ (Interfacial Transitional Zone) is even denser [15]. Their higher chemical resistance among other admixtures is due to the fineness of the grains, the consistency of the mixtures and the refinement of the pores within the cementitious matrix [6].

According to Rahmani and Ramezanian pour the binary addition (limestone filler + natural pozzolan) maintains the best resistance in a sulfuric acid environment [16]. On the other hand, Ghrici et al. found that the limestone filler had better resistance in the sulfuric acid solution compared to natural pozzolan addition and binary addition (pozzolan natural + limestone filler) [17]. In keeping with the research carried out by Bassuoni and Nehdi, the quaternary mixture prepared with 50% OPC, 15% limestone filler, 20% slag and 15% fly ash has the greatest reduction in weight loss with respect to acid medium. This can be attributed to the incorporation of limestone filler in the binder of mixture [1]. Because, in an acidic environment, the limestone filler in the cementitious matrix has a role in chemical neutralization (consumption of acid). According to Bassuoni and Nehdi, the existence of strong calcium/magnesium carbonate peaks on the surface of quaternary mixture samples suggests that limestone can create a local buffer on the surface of the concrete. The acid concentration will be low and pH is high near the surface, leading to a slower rate of degradation [1]. According to Bertron et al., the weight loss in slag cement is lower, contrary to when compared to ordinary cements [18]. According to them, the origin of durability of blast furnace slag could be due to residual anhydrous grains widely present in the degraded zone of the pastes with slag.

The carbonation process proceeds slowly due to the low concentration of carbon dioxide. The carbonation of the concretes corresponds to a set of physico-chemical mechanisms resulting from the instability of the hydrated and anhydrous phases of the cement paste in contact with carbon dioxide ( $CO_2$ ). Carbon dioxide is highly reactive with cement hydrates such as calcium hydroxide [ $Ca(OH)_2$ ] and calcium silicate hydrate (CSH), as well as non-hydrated cement phases such as tricalcium silicate ( $C_3S$ ) and dicalcium silicate ( $C_2S$ ). Among these phases,  $CO_2$  has a greater affinity for the reaction with  $Ca(OH)_2$  [19]. The carbonation of the cementitious matrix can be described in a simplified way by the following three chemical reactions consisting of precipitation of calcite by dissolution of portlandite and decalcification of C-S-H and ettringite (See equations below).

$$Ca (OH)_2 + CO_2 + H_2O \rightarrow CaCO_3 + 2H_2O$$
 (4)

$$C-S-H + 2CO_2 \rightarrow SiO_2 + 2CaCO_3 + H_2O$$
 (5)

$$\begin{split} 3\text{CaO} \cdot \text{Al}_2\text{O}_3 \cdot 3\text{CaSO}_4 \cdot 32\text{H}_2\text{O} + 3\text{H}_2\text{CO}_3 \\ &\rightarrow 3\text{CaCO}_3 + 3(\text{CaSO}_4 \cdot \text{H}_2\text{O}) + \text{Al}_2\text{O}_3 \cdot \text{xH}_2\text{O} + (29 - \text{x})\text{H}_2\text{O} \end{aligned} \tag{6}$$

In fact, carbon dioxide is diffused by the porous networks and the zone of concrete previously carbonated. In this reaction, an initial pH between 12 and 13 evolves towards neutrality (pH  $\sim$  9). Consequently, the vulnerability of reinforcements and concrete increases. Hence, the phenomenon of corrosion which causes degradation of the concrete and, especially, the swelling of the steel.

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