



# Producing C-S-H gel by reaction between silica oligomers and portlandite: A promising approach to repair cementitious materials

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## 1. Introduction

The technological advances brought by the procedure to obtain Portland cement and the use of reinforced concrete shaped late 19th and 20th century architecture, giving rise to the architectural styles of the modern era (modernism, rationalism, brutalism...). As a defining characteristic of said styles, these new reinforced structures were often left in fair-faced condition [1], without coatings or painting, increasing its susceptibility to suffer degradation under aggressive environments. The earlier manifestation of most degradation processes is the onset of nano- and micro-cracks that is usually followed by increased porosity [2], material loss and decreased mechanical performance [3]. In addition to these phenomena, reinforced concrete is specially affected by the ingress of soluble CO<sub>2</sub> and Cl<sup>-</sup> ions, which may cause de-passivation and corrosion of the reinforcements, eventually resulting in delamination and structural failure. Nowadays, the current manufacturing processes and construction techniques are designed to minimize these issues. However, in the case of older concrete-based structures the limited knowledge about the concrete degradation mechanisms has given rise to a sizeable number of buildings that require repair interventions and preventive maintenance treatments to combat their damages.

Nowadays, the application of surface treatments, which prevent the water ingress and fill the cracks produced by degradation, is the preferred choice for preserving historic concrete [4]. Although coatings such as epoxy and acrylic resins are commonly employed with this aim, they present significant drawbacks associated to thick layer formation (0.1–1 mm) on concrete [5]. These layers can modify the aesthetic conditions of the surface and, especially, they present low durability due to low penetration and poor adhesion to the substrate. Impregnation treatments, on the other hand, present a low viscosity, and consequently have the ability to penetrate deeper (1–20 mm) and react in situ within the pore structure of the damaged substrate, promoting long-lasting effectiveness [5]. Different treatments based on nano-silica dispersions, available in the market, are employed for concrete protection due to their ability to undergo pozzolanic reaction with the portlandite and fill the pores with C-S-H. However, despite the small particle size, these treatments have difficulties penetrating into the smaller pores [5,6].

Silica oligomer/monomer-based sols (e.g., tetraethoxysilane, TEOS) are widely used in the stone conservation market [7] due to their advantages and their economical cost comparable to other alternatives. Their low viscosity allows them to penetrate deeply into the porous structure and after polymerization, which occurs upon reaction with environmental moisture, a stable gel with a silicon-oxygen backbone is formed. This  $\text{SiO}_2$  structure has a similar composition to the quartz and silicate minerals present in a wide variety of building materials, including the hydrated calcium silicate (C-S-H) and aluminosilicates (e.g. strätlingite) present in concrete or the siliceous sands used as fine aggregate. As the main drawbacks for these products, xerogel cracking is produced due to the high capillary pressures inside the dense microporous gel network and their effectiveness tends to be limited on carbonate-rich materials, such as limestone and concrete surfaces subjected to intense carbonation.