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Low-pressure silica injection for porosity reduction in cementitious materials

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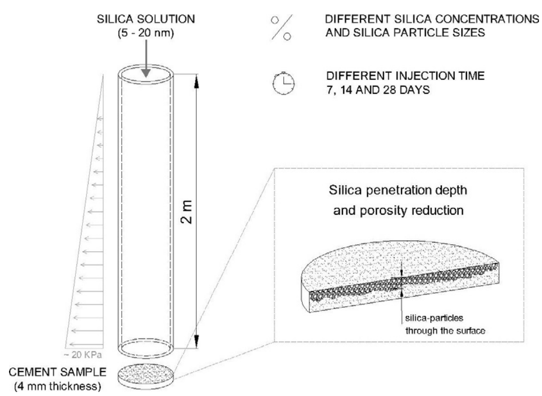
HIGHLIGHTS

- A novel non-destructive technique for cement surface treatment has been developed and proven effective under laboratory conditions.
- Nano-silica and silica fume can successfully penetrate the cement surface within 14 days and create extra C-S-H.
- Silica injections are carried out at low-pressure, ca. 20 kPa and this is the first demonstration of a simply applied and effective technique.

GRAPHICAL ABSTRACT



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ABSTRACT

The durability of building materials is related to the presence of cracks as they provide a fast pathway for the transport of liquid and gases through the structure. Restoration and preservation of historic buildings are the potential applications of this novel technique which uses nano-silica and silica fume particles for consolidation. The small particle size range and the high reactivity of nanoparticles allow them to interact with calcium sources naturally present in cement and concrete, forming binding and strengthening compounds such as calcium silicate hydrate. Nanoparticles act as a crack-filling agent, reducing the porosity and increasing the durability of existing materials. In this study we describe the injection of nano-silica, under low water pressure, into hydrated cement paste. This novel technique can tailor the mechanical and hydraulic properties of existing building materials using a simple and non-destructive procedure.

1. Introduction

Most of the built environment uses cement or concrete in some way, and many iconic buildings constructed in 1920s and later suffer from crack formation, water penetration and damage mechanisms such as alkali-silica reaction. Cracking in concrete and mortar is an inevitable phenomenon of ageing and erosion. Thus, material characteristics such as porosity, permeability and strength are altered during ageing. Hardened concrete and cement contain two important mineral phases: calcium hydroxide (portlandite) and calcium silicate hydrate (C-S-H), the former has a defined crystalline structure, the latter is semi-crystalline [14]. C-S-H is the phase responsible for strength development in concrete

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and can form up to 70% of the total volume of hardened concrete [4]. C-S-H is produced by hydration of alite and belite (impure tricalcium silicate and dicalcium silicate respectively) which are present in cement clinker. Pozzolanic materials such as fly ash, rice husk ash and silica fume can also be added, resulting in the production of more C-S-H and improved mechanical performance [18,16]. The formation of cracks and increased porosity from leaching in concrete and cement paste presents an easy pathway for the ingress of moisture. Gaps and cracks can be reduced by application of nanoparticle consolidants. In the work presented here, the injected silica reacts with portlandite naturally present in hydrated cement paste to form new C-S-H and reduce the porosity of the system. The result is decreased permeability and potentially increased durability of the cement [3,8,17,9]. Research on partial replacement of cement clinker with nano-silica [13] found that increasing the quantity of nano-silica replacing cement from 3 vol.% to 5 vol.%, produced a mortar with higher mechanical strength by acceleration of the hydration reaction and the filler effect of nano-particles. In addition, the hydrated paste had a dense and compact texture and an absence of portlandite crystals was observed, suggesting that most of the calcium hydroxide reacted with the nano-silica added [13,10,15]. Nano-silica addition to cement paste increases C-S-H formation and accelerates hydration of unreacted alite (C_3S), due to the high reactivity of small particles [2]. An average water penetration depth of 14.6 cm in concrete made with fly ash and cement under low applied pressure was observed, whereas a water penetration depth of 8.1 cm in the same concrete mixed with nano-silica under high applied pressure was recorded, confirming the improvement in water penetration resistance with nano-silica addition [10]. It was concluded that the pozzolanic reaction of fly ash in the presence of nano-silica produces C-S-H faster and earlier compared to ordinary Portland cement (OPC) mixed with fly ash but no nano-silica. Varying the nano-silica content (3 wt.%, 6 wt.%, 10 wt.%, and 12 wt.%) in mortar produces an increase in strength correlated with a decrease in calcium hydroxide content. The heat of hydration is also increased by addition of nano-silica due to the rapid hydration of silicates [11]. Nano-silica surface treatments have been investigated using electro-kinetic deposition, nanoparticle coating, brushing, etc. A reduction in permeability was observed by Cardenas et al., for low alkali cement paste with 0.8 w/c ratio and impregnated with colloidal alumina by electro-phoresis [3]. Pore size refinement by reduction in the pore volume of treated samples with higher w/c ratio was also observed. The effect of curing temperature on hardened cement paste treated with nano-silica and tetraethoxysilane (TEOS) under sealed and unsealed conditions was studied [8]. Hou et al. found that mortar samples cured at 50 °C and treated with nanosilica/TEOS show a reduction in water absorption compared to samples treated in the same way but cured at 20 °C. High temperature curing contributes to the production of additional C-S-H gel and reduction of calcium hydroxide, which results in smaller capillary pores and finer gel pores. The transport properties of cement pastes with varying w/c ratio and surface treated with nano-silica and TEOS were also investigated. The water absorption and water vapour permeability are decreased by incorporation of nano-silica and TEOS in mortar with higher w/c ratio. Hardened mortar, surface treated by nano-silica using electro-migration, showed reduced cumulative porosity, and a higher rate of pozzolanic reaction was confirmed by the reduction in portlandite content [17]. While the application of nano-particles to cement and concrete surfaces has been shown to have beneficial effects on cement durability, very little research has been conducted on developing low cost and non-destructive techniques for concrete surface treatment. The aim of this work was to investigate a non-destructive and easily applied conservation treatment for cracked or friable concrete which is relevant to infrastructure conservation, ranging from buildings to bridges and more specialist applications in nuclear waste containment ponds. In this study the effect of nano-silica and silica fume injection in hardened cement paste was investigated by quantitative analysis of the resulting hydration products (C-S-H and portlandite) present.

2. Materials and methods

2.1. Materials

All experiments were carried out on pure hardened cement paste, made using ordinary Portland cement CEM II/A-L, class 42.5 N (physicochemical properties are listed in Table 1) and deionized water. Samples were treated with nano-silica (NS) suspension, LUDOX T-50, or silica fume (SF), ELKEM microsilica. Their chemical properties are detailed in Table 2.

2.2. Sample preparation

Cement samples were prepared by mixing Portland cement and deionized water at a water to cement (w/c) ratio of 0.41. Cement paste was mixed in a rotary mixer according to BS EN 196-1:2005 before being cast into plastic moulds (35 mm ø and 4 mm thickness) and cured under controlled conditions (relative humidity of 98 ± 2% and temperature of 21 ± 2 °C). After 28 days, cement discs were oven-dried at 60 °C for ca. 100 h, or until mass change was negligible. The aim of this experiment was to measure silica entrainment through the pore structure, rather than conduct accurate micro-structural analyses. Therefore, relatively gentle, oven drying at 60 °C was considered adequate for a comparative study of silica imbibition.

2.3. Experimental design

Nano-silica injection was carried out by varying three parameters: injection period, concentration of silica suspension injected, and silica particle size (NS or SF), using a constant applied pressure head. Silica solutions were prepared using nano-silica stock suspension or solid silica fume, mixed with deionized water. In order to investigate how the penetration depth in the disc varies with nano-silica content, three different suspension concentrations (10 wt.%, 15 wt.% and 20 wt.%) were used, for a total injection time of 14 days. The effect of injection time was determined by keeping cement discs under constant hydrostatic injection for 7, 14 and 28 days using 10 wt.% nano-silica colloidal suspension. To compare the reactivity and effect of particle size on penetration depth, samples were injected with 10 wt.% and 20 wt.% suspensions of silica fume or nano-silica for a period of 14 days (Table 3). The cement

Table 1
Characteristic of CEM II/A-L (Class 42.5 N) Portland cement (according to the certificate of conformity, test method BS EN 196-2).

Components	CEM II (%)
Clinker	80–94
Gypsum added	4.0
Limestone	6–20
Chemical composition (>0.2%)	
SiO_2	18.2
Al_2O_3	4.5
Fe_2O_3	2.6
CaO	64.0
MgO	1.3
SO_3	2.3
Na_2O	0.23
Solid density (kg/m^3)	3100
Specific area (m^2/g)	0.41
Compressive strength at 28 day (MPa)	57.5