Biomineral Sealing of Cracks in Degraded Concrete Structures - 22138

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ABSTRACT

Civil Nuclear sites contain significant volumes of concrete infrastructure, including both external and internal structures. As a consequence, different concretes are exposed to differing environmental conditions resulting in variable mechanisms and rates of concrete degradation. For example, external structures may be exposed to salt water and freeze-thaw cycles, while internal structures may be exposed to high temperatures and/or high levels of radiation.

Degradation can take the form of spalling, mineral dissolution and ultimately, cracking. These mechanisms increase concrete permeability, which can lead to two significant issues for decommissioning operations. First, cracking of the concrete increases water ingress, accelerating rebar corrosion and reducing the structural integrity of the buildings. This increases the complexity of decommissioning operations, which frequently involve the use of heavy plant, particularly for waste retrieval operations. Second, cracking of concrete waste containment structures can lead to contamination of the subsurface, increasing the complexity of environmental remediation operations.

Microbially-induced calcite precipitation (MICP) may provide a low-cost, durable method for the reduction of permeability in aged or damaged concrete infrastructure. The method relies upon the ureolytic capacities of the bacterial strain *Sporosarcina pasteurii* to metabolise urea in the presence of calcium, resulting in precipitation of calcium carbonate. Bacteria are injected into the fractured concrete, alongside urea and calcium in solution, and layers of calcium carbonate are precipitated. In the research presented here, we treat fractured concrete cores in the laboratory, reducing the hydraulic conductivity by 2-3 orders of magnitude in concrete samples collected from UK Civil Nuclear sites. We then utilise X-CT imaging to quantify and visualise the calcite deposited within the fracture network. Our research indicates this treatment protocol can significantly reduce concrete permeability as well as improving structural integrity, thus increasing the longevity of degraded concrete nuclear assets.

Introduction

Civil Nuclear sites contain significant volumes of concrete infrastructure, most of which is long past its original design life (generally 50-60 years). UK decommissioning programmes are planned to take place over several decades, hence, these structures must continue to operate safely for years to come. Concrete structures on nuclear sites are exposed to differing environmental conditions depending on their use and location, resulting in variable mechanisms and rates of concrete degradation. For example, external structures may be exposed to salt water and freeze-thaw cycles, while internal structures may be exposed to high temperatures and/or high levels of radiation. These harsh environmental conditions lead to the formation of cracks or fractures; and consequently to increased permeability, ultimately resulting in corrosion of the reinforcement. Traditionally, the repair method used depends upon the purpose of the concrete structure, but includes patching with cement or bitumen, the injection of new cement and the injection of epoxy resins [1], [2]. Use of cement repairs is limited by penetrability; cement is viscous and typically has a large grain size (30-50 microns), while epoxy resins have low durability (~10 years) and have associated negative environmental impacts.

Microbially-induced calcite precipitation (MICP) may provide a low-cost, low-carbon, durable method for the reduction of permeability in aged or damaged concrete infrastructure. The method relies upon the ureolytic capacities of the bacterial strain *Sporosarcina pasteurii* to metabolise urea in the presence of calcium, resulting in precipitation of calcium carbonate. Bacteria are injected into the fractured concrete, alongside urea and calcium in solution, and layers of calcium carbonate are precipitated. The MICP process occurs via the enzymatic breakdown of urea to produce ammonium and carbonate ions. These carbonate ions will bond to any free calcium ions present in the system, and in a high pH environment the formation of calcium carbonate (calcite) will be promoted.

Most laboratory studies of the treatment of concrete have relied on pouring/dripping treatment solutions onto concrete blocks or immersing blocks fully in treatment solutions [5]. Neither of these approaches are practical for most in situ applications since they rely on gravity and the top of the crack is generally not exposed.

In this research, we inject fluids to enable MICP repair of non-vertical cracks and to ensure treatment of the full fracture network. We inject the MICP treatment solutions into a fractured concrete core under controlled flow conditions. We visualise and quantify the resulting deposition of calcite within the fracture using X-μCT. We demonstrate that the MICP reduces fracture permeability and results in some strength recovery in an initially fragmented concrete core.

Materials and methods

Concrete Sample Collection and Preparation

Concrete blocks were provided by Babcock Marine Ltd, taken from concrete caissons/dock blocks used as part of a dry dock structure at Devonport Royal Dockyard facility, within HMNB Devonport. The dimensions of each block were W: 100 cm, L: 181 cm, H: 80 cm, weighing approximately 3.3 tonnes.

From these blocks, a 36 mm diameter by 72 mm length core was cut to produce aged concrete samples suitable for laboratory scale testing. An unconfined compressive strength (UCS) test was conducted to determine the initial UCS value of the concrete and also to artificially induce fracturing. Once visible fracturing was observed, the core was then split in two along the predominant fracture through impact with a chisel.

The fractured core was reassembled, wrapped in heat-shrink tubing and confined at 1000 kPa in a core holder for 1 hour to compress the two halves of the core firmly together. After which the core was vacuum-saturated with tap water and scanned to image the initial open fracture network via X-μCT. Finally, the core was remounted in the core holder and a confining pressure of 1000 kPa was applied to ensure no by-pass of MICP treatment fluids around the core during treatment.

Bacterial growth and preparation for injection

S. pasteurii were grown from cryopreserved stock cultures in a solid medium consisting of 5.5 gL^{-1} Yeast Extract (Sigma-Aldrich), 5 gL^{-1} sodium chloride (Fisher scientific), 0.4 gL^{-1} D-glucose (Sigma-Aldrich), 0.4 gL^{-1} K_2HPO_4 (Sigma-Aldrich), 20 gL^{-1} urea, and 15 gL^{-1} agar (Sigma-aldrich). Urea was added aseptically after autoclaving. A single bacterial colony was then transferred into a liquid growth media consisting of 5.5 gL^{-1} Yeast Extract (Sigma-Aldrich), 5gL^{-1} sodium chloride (Fisher scientific), 0.4 gL^{-1} D-glucose (Sigma-Aldrich), 0.4 gL^{-1} K_2HPO_4 (Sigma-Aldrich), and 20 gL^{-1} urea (Sigma-Aldrich). Urea was added aseptically after autoclaving. The culture was incubated overnight at 30 °C. The culture was then centrifuged at 6000 G for 7 minutes. The supernatant was discarded, and the bacterial cell pellet resuspended in mains tap water to an OD_{600} of 1.0. This solution was prepared immediately prior to injection into the core.

MICP Treatment and Permeability Measurements

A HPLC pump was used to inject water and treatment fluids through the core. Initial absolute permeability (units m^2) was determined during injection of tap water by controlling the flow rate at the pump and measuring the differential pressure across the core. This calculation utilised Darcy's law to measure permeability (k):

$$q = -\frac{k}{\mu}\nabla p\,.$$

Where k = permeability (m^2), μ = dynamic viscosity of the fluid (Pa.S), ∇p = pressure drop (Pa), and q = instantaneous flux (m^3/s).

Treatment cycles consisted of seven main injection stages through the core, interspersed with water pulses to prevent blockage of the pump and the tubing. For each bacterial and cementing solution injection stage, 5 ml of fluid was injected per cycle at a flow rate of 0.1 ml/min. Cementing solution consisted of 111 gL^{-1} calcium chloride (Sigma-Aldrich), and 60 gL^{-1} urea (Sigma-Aldrich).

The order of these injection steps, for a single treatment cycle are shown in Table 1. Permeability measurements were taken, by injecting tap water at three different flow rates, after each treatment cycle.

Table 1: Treatment Cycle Steps for Cycles 1-6

Treatment Step	Treatment Solution	Flow Rate (ml/min)	Duration (minutes)	Total Volume (ml)
1	Bacterial Injection	0.1	50	5
2	Static Period	N/A	120	N/A
3	Water Injection	0.1	20	2
4	Cementing Injection	0.1	50	5
5	Static Period	N/A	960 (overnight)	N/A
6	Water Injection	0.1	20	2

Between treatment cycles, the tubing lines and pump were flushed thoroughly with tap water to prevent clogging. Six treatment cycles (Table 1) were completed.

After 6 treatment cycles, permeability had decreased significantly. Hence, to lower the inlet pressure, the flow rate for all injection stages was halved to 0.05 ml/min, with the treatment duration doubled to maintain the same volume of treatment fluid as in Cycles 1-6 (see **Table 2**). Three further treatment cycles were completed with this flow rate.

Table 2: Treatment Cycle Steps for Cycles 7-9

Treatment Step	Treatment Solution	Flow Rate (ml/min)	Duration (minutes)	Total Volume (ml)
1	Bacterial Injection	0.05	100	5
2	Static Period	N/A	120	N/A
3	Water Injection	0.05	40	2
4	Cementing Injection	0.05	100	5
5	Static Period	N/A	960 (overnight)	N/A
6	Water Injection	0.05	40	2

Tomography (X-μCT) Method

X-ray micro computed tomography of the concrete core was carried out using a Nikon XT H 225 LC X-ray computed tomography system. This generated a 2D stack of projections from the scan. The core was scanned before MICP treatment, and afterwards (after cycle 9). Following reconstruction, pre- and post-treatment stacks were aligned using the registration software Elastix [4]. Thresholding and processing of the stacks was performed using the FIJI distribution of ImageJ [5] which allowed the solid components (concrete: cement matrix or aggregates) to be distinguished from the void or fracture spaces. The data was binarized based on grey values (255/White or 0/Black), with 255 representing concrete/calcite, and 0 representing void/fracture space.

By subtracting the pre- and post-treatment binarized stacks, it was possible to visualise where calcite was deposited within the fracture network. It was also possible to count the number of voxels in the scan, within which calcite had precipitated, this corresponds to an estimate of the volume of calcite precipitated.

Results and discussion

Permeability was observed to continually decrease with each treatment cycle completed (Fig 1). After 9 treatment cycles, a permeability reduction of 3 orders of magnitude was achieved. After 9 treatment cycles, the core was removed from the core holder and imaged under X-μCT. X-μCT analysis revealed that the initial fracture network within the core was now coated with a new solid phase, calcite, see Fig 2. Based on X-μCT data, the measured volume of calcite precipitated was 46.42 mm^3 within the fracture network.

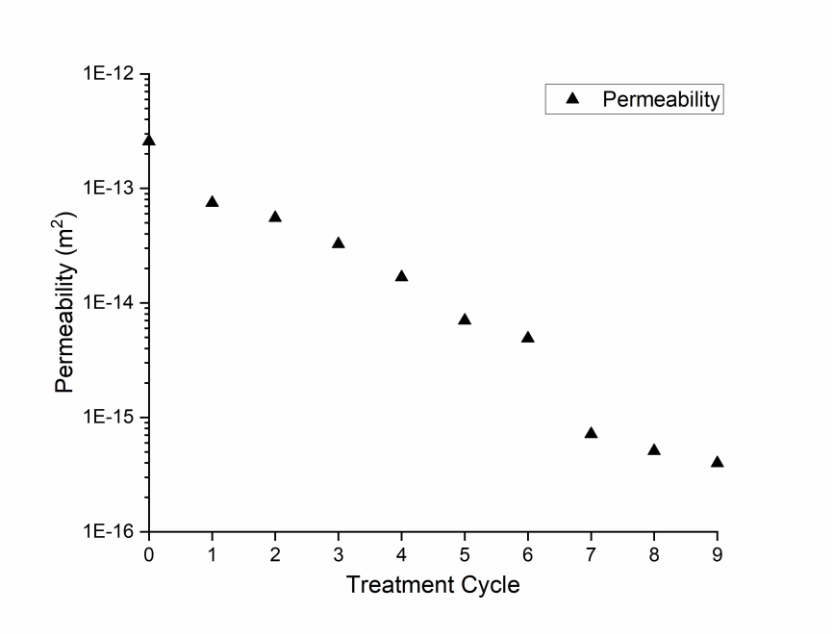
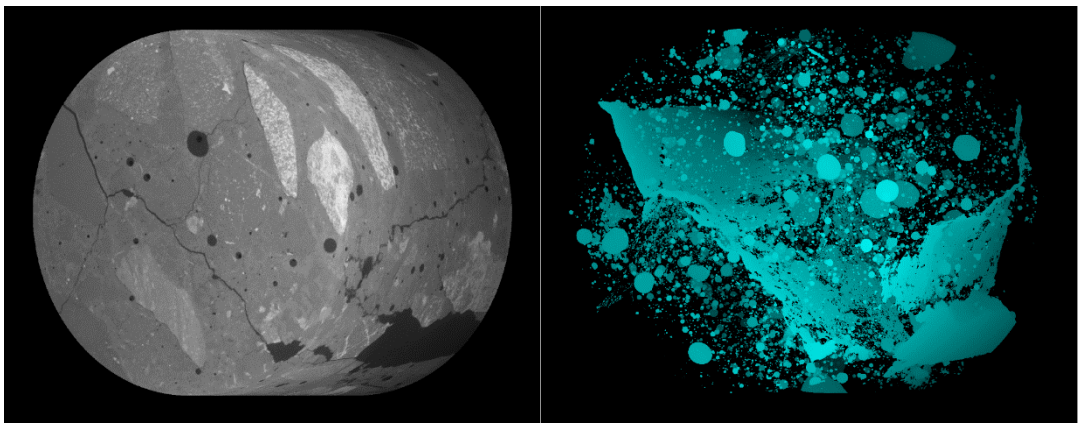


Figure 1: Permeability change vs. Treatment Cycle Number

a) b)



c)

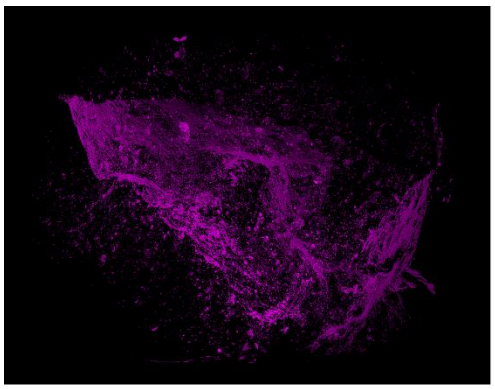


Figure 2: X-CT visualisations of the concrete core, showing front ¾ view relative to the base of core. (a) 3D reconstruction of the scan shows that fractures are present in the concrete, along with voids and aggregate pieces. (b) Initial void and fracture space prior to treatment (cyan). (c) the location of calcite in the fracture network after 9 cycles of MICP treatment (pink).

The initial measured compressive strength of the core was 14.41 MPa. After the 9 cycles of MICP treatment a second UCS test was conducted, and the repaired compressive strength was measured as 1.58 MPa, indicating that ~10% of the initial strength of the concrete core had been regained via MICP the treatment.

Conclusion

In this study we demonstrated that MICP treatment via controlled injection can be used to reduce the permeability of a fractured concrete core by three orders of magnitude. The final strength of the MICP treated core was 1.58 MPa (recovered from an initial UCS value of zero when the core was fragmented prior to treatment). Using X-μCT we show that the calcite precipitation has occurred throughout the fracture network, along the full length of the core (72 mm length). Hence, penetration of treatment fluids has occurred throughout the fracture network and damage repair has been achieved over the entire fracture length.

Acknowledgments

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