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# Development and experimental validation of an overlay mortar with biocide activity

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Biodeterioration of concrete by microorganism colonisation may be a problem in several structures, especially in irrigation and hydroelectric canals. The main problem in such structures is the proliferation of algae and cyanobacteria that affect the performance of the structure, increase the maintenance costs and affects its durability. A research was conducted to develop a novel cement-based material with biocide activity that can be used as an overlay mortar in existing structures, such as canals and pipes. With this aim, ten commercial biocides were evaluated in a laboratory campaign to assess the effectiveness of the compounds against the microbial colonisation of concrete. Both mono- and multicomponent formulations were designed from the commercial products, to increase their antimicrobial effect obtaining a set of biocide formulations. The formulations were submitted to a flowchart process to determine their influence on the physical properties of the concrete, evaluate the release of the actives, and their antimicrobial efficiency both before and after accelerated aging processes. During the campaign, some formulations were observed to diminish the strength of the concrete. Such behaviour was normally due to the interaction of the active with the cement hydration process. Other formulations showed a high release of active from the concrete in water, compromising the durability of the treatment. In general, monocomponent formulations did not succeed to fulfill all the requirements, thus multicomponent formulations were analysed. One studied multicomponent formulation presented particularly good results in all properties analysed. This product did not significantly change the properties of concrete and the release of active in water from the concrete was low, while the antimicrobial effects were long lasting.



## 1. Introduction

Concrete is one of the most used construction materials because it has high structural performance, long durability, and a relatively low cost. The surface of concrete is resistant to corrosion and provides a physically and chemically stable environment that can be exposed to potable water or wastewater; for this reason, it is commonly used in pipes and canals [7]. In these structures, microorganisms can easily colonise concrete and cause biodeterioration of the material. The main problem in irrigation and hydroelectric canals is the growth of algae and cyanobacteria among other microorganisms on the canal walls. This biological growth creates many problems that are reflected in increased maintenance costs. The effective section of the canal is decreased and the roughness coefficient of the canal wall is increased, which diminishes the water flow [14]. Some studies reflect a decrease of 10% in the hydroelectric generation capacity because of algal growth [8,19]. Furthermore, it is worth noting that presence of some types of fungi can have detrimental effects in concrete walls as well as can affect production of certain cereals, as Fusarium [12]. Moreover, the detachment of filamentous organisms such as green algae carries the risk of clogging and plugging filters. Both the growth of these organisms on concrete and the required cleaning activities to remove them from the walls cause erosion to the surface of the concrete. This intensifies the porosity and roughness of

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the material and makes it more vulnerable to re-colonisation.

Algal growth is usually preceded by concrete biofouling. This phenomenon is often grouped in the literature into several stages that include an initial accumulation of adsorbed organics, the settlement and growth of pioneering bacteria creating a biofilm matrix and the subsequent succession of micro and macrofoulers [9]. Usually, bacteria play a dominant role in the formation of this biofilm and further colonisation of larger microorganisms [22].

Several antimicrobial treatments have been used in concrete and cement-based materials. Conventional studied procedures involve the usage of water repellents, biocides, or both. Water repellents slow the adsorption of water by reducing the surface energy and capillary forces of the concrete, thus decreasing the bioreceptivity of the material. Biocides focus on decreasing biological activity. While the use of hydrophobic compounds alone was shown to be insufficient in inhibiting microbial growth, the application of a combination of both treatments has been reported as effective [10,20]. The order of application of the different products influences the efficiency of the final treatment; this must be considered to avoid negative effects [15–17].

Biocide treatments have one of two mechanisms: the elimination of the microorganisms already present on the material, and the prevention and control of microbial re-colonisation on clean surfaces. Many compounds have been analysed and tested over the years, including zeolites supporting heavy metal ions (such as copper and silver), zinc oxide, silver nanoparticles, pyridine biocides, silver nitrate, and quaternary ammonium compounds, or Quats [7,10,11,20]. Quats are most frequently used because they have good efficacy as algaecides [18]. Nevertheless, this issue is far from being solved from a scientifically and technically point of view.

The main aim of this paper is to present the results obtained in a research devoted to the development of cement-based materials with biocide activity to be used as a biocide mortar layout in existing concrete structures [6,21]. Therefore, different commercially available antimicrobial products were evaluated for effectiveness against microbial colonisation in concrete. These products were chosen for cost-effectiveness, non-toxicity, and capacity as disinfectants in other applications, such as floor and wall coverings. Different combinations of the commercial biocides were considered to increase their antimicrobial effect obtaining a set of biocide formulations. The experimental campaign was designed to evaluate the influence of the incorporation of the biocide formulations in concrete properties and their antimicrobial activity. Furthermore, the durability of the biocide activity was also evaluated through accelerated aging processes. The performances of the different combinations studied are shown and analysed, with special attention to the reasons responsible for the failure of the formulations.

## 2. Materials and methods

### 2.1. Concrete specimens

Concrete specimens were fabricated with the dosage shown in Table 1. The cement selected to produce the different mixtures was a CEM II A/V 42.5 R. All aggregates were siliceous with a low fraction of particles under 0.125 mm sieve. The largest aggregate size was selected according to the size of the Petri dishes utilized for the microbiological tests. Furthermore, a large water-to-cement (w/c) ratio of the mixes (0.7) was used to obtain samples with high porosities that favoured microbiological colonisation. Lastly, a plasticiser additive (Pozzolith 475N, BASF Construction Chemicals) was added to each mixture based on percentage over cement weight (% ocw). All concrete samples were made according to EN 12390-2 [1]. The samples were cured in a curing chamber (20° ± 2 °C; 95± 5% relative humidity) for 28 days.

**Table 1**
Concrete dosages for reference and biocide mixtures.

| Compound | Dosage (kg/m$^3$) |
|---|---|
| Cement | 350 |
| Fine aggregate (0–5 mm) | 1074 |
| Coarse aggregate (5–12 mm) | 724 |
| Plasticiser | 3.5 (1%) |
| Water | 244 |

### 2.2. Design of biocide formulations

Ten commercially available antimicrobial products (Table 2) were tested during the experimental campaign. These products were chosen for cost-effectiveness, non-toxicity, and capacity as disinfectants in other applications, such as floor and wall coverings. The antimicrobial products shown in Table 2 were used to develop different formulations for cement-based biocides. These biocide formulations were composed of one (monocomponent) or several antimicrobial products (multicomponent) as well as incorporating different additives in some cases (see Table 3). Multicomponent formulations were designed to achieve as wide a spectrum of antimicrobial action as possible. The content of each component of the formulations is given between brackets, expressed as percentage of the formulation.

As Table 3 shows, some additives were incorporated to improve the physical properties of the mixture. MOUSSEX®, BUBLEX®, and tri-isobutyl phosphate (TIBP) are used as defoamer compounds to avoid air entrapment in the concrete mix; propylene and polyethylene glycol (PG and PEG, respectively) are used as solvents instead of water to allow the solubilisation of the antimicrobial products and the collection of liquid admixtures; OPTIGEL CR® is an activated bentonite product used for anti-settling and stabilizing in water-based systems. Finally, calcium filler is used as a dispersive matrix for the biostatic agents. The biocide formulations were incorporated in the concrete mix in different amounts, as listed in Table 4; the dosage is expressed as percentage over dry weight of the mix (% odw). Concrete specimens were fabricated for each formulation dosage. Furthermore, reference samples were made for each biocide formulation without since cement samples may vary over time. These quantities were chosen based on the recommendations of the manufacturers of the products. Table 4 also provides information of the content of active principle in the formulations; in case of multicomponent formulations, individual and total contents are given for each antimicrobial product.

### 2.3. Experimental campaign

The experimental campaign was designed as a flowchart, as described in Fig. 1 with three consecutive main phases. Firstly, the influence of the different biocide formulations on physical properties of concrete was determined. Secondly, those biocide concretes that passed the first phase were subjected to a release test to determine the amount of active released from the sample. Finally, microbiological tests were conducted in those samples that passed the previous phases. The evaluation of antimicrobial properties was conducted twice, before and after an accelerated aging test, to simulate adverse conditions and thus estimate the durability of the tested treatment.

#### 2.3.1. Physical characterization of concrete specimens

Concrete specimens were characterised to determine the influence of the biostatic formulations on the physical properties of