CIVIL
ENGINEERING
JOURNAL

Article no. 34

THE CIVIL ENGINEERING JOURNAL 4-2024

APLICABILITY OF MAGNESIUM PHOSPHATE CEMENT FOR BIORECEPTIVE CONCRETE TILES

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ABSTRACT

The pressing need for innovative green solutions in urban environments is evident. This study explores a novel concept of integrating green areas directly onto structural surfaces to enhance urban infrastructure with real floral life. Specifically, the research focuses on the application of newly developed bioreceptive concrete, which is engineered to promote the growth of microorganisms, including mosses, lichens, and algae, on its surface. To achieve optimal bioreceptivity, the concrete's properties were carefully modified by changing the mix design and using Magnesium Phosphate Cement instead of traditional Ordinary Portland Cement. This substitution resulted in a significant reduction in pH, creating a more favorable environment for the life of plants and microorganisms. Additionally, an optimal formulation was developed with a suitable grain size distribution to achieve the desired porosity, which is critical for water retention and microbial establishment. This bioreceptive concrete was tested as an additional layer on top of normal OPC concrete which was used to manufacture to the supporting structure. The concrete properties were carefully optimized to enhance bioreceptivity by altering the mix design and employing a different type of hydraulic binder to meet the conditions necessary for biological growth. The study primarily examines two key properties of bioreceptive concrete: pH and porosity.

KEYWORDS

Bioreceptivity, Bio-active concrete, Low-pH cementitious material, Magnesium phosphate cement, Porosity

INTRODUCTION

Cities and the city life today face significant pressure from the increasing urbanization. Rapid population growth leaves a profound impact on the environment with the urban development transforming the natural areas into engineered infrastructure. This shift presents enormous challenges for maintaining urban ecosystems. The built environment has led to extensive impermeable paved surfaces, resulting in vegetation loss, increased surface runoff, and enhanced solar energy retention. Consequently, there is an urgent need to rethink and rebuild urban infrastructure to address these issues, and many cities are beginning to recognize the importance of green infrastructure. Incorporating greenery into densely populated urban areas is essential for environmental enhancement. It is well known that public green spaces positively affect biodiversity, climate, wellness, and air quality. They promote physical activity, relaxation, and social interactions. Plants produce oxygen and filter out polluted air particles, while also playing a critical role in cooling urban areas. These benefits ensure that cities become better places to live and work, with a positive influence on mental health and well-being. Therefore, natural green assets, such as parks and water systems, are in high demand. Unfortunately, they require substantial ground space, which is often

DOI 10.14311/CEJ.2024.04.0034

scarce in dense profit-oriented urban centers, which consequently poses a significant challenge to their implementation [1].

Then, the green roofs, walls, and facades offer a viable solution to this problem, as they require minimal or no profit-generating space. There are numerous unused spaces, such as bare facades and retaining walls, whose surfaces could be better utilized from the green-solution perspective. Architects and designers should consider these green solutions when designing new urban infrastructure. Green walls and roofs have been applied successfully worldwide. However, green walls are more spatially efficient since vertical areas provide more space than roofs, leading to a primary focus on integrating plants into wall systems. Green walls, also known as vertical gardens or living walls, encompass all forms of vegetated vertical surfaces (Figure 1). They can be incorporated into both new and existing buildings [2].

Fig. 1 - Museum Caixa Forum, Madrid [5]

Another option for greening the facades is the use of pots and planters which are placed around the perimeter of the facade. This relatively new construction method is mentioned in [3] as the so-called perimeter flowerpots, it means an intensive system of green facades, because the planters are placed in front of the facade and are not connected to the soil in the ground. These planters are designed for larger plants, or even trees, giving buildings a unique aesthetic touch. However, unlike other structures, they impose significant extra loads and thus have a major impact on the building's structure (Figures 2 and 3) [4].

At a small building scale, green walls offer multiple benefits in terms of protection for the buildings. They enhance the sustainable performance of structures and also improve the local microclimate, as the plants contribute to air quality by producing oxygen and reducing atmospheric CO_2. Additionally, dense foliage captures pollutants. Recent studies confirm that green wall systems can influence building heat gain and loss, thereby reducing energy demand and improving indoor thermal comfort [8]. Furthermore, living walls serve as passive acoustic insulators.

Fig. 2 - Tower-Flower [6]

Fig. 3 - Bosco Verticale [7]

However, green walls also present several challenges. They require significant maintenance and an elaborate watering system, which may lead to high additional cost. Thus, there is a need for more efficient technical solutions. With advancements in building and architectural technologies, there is a push for more innovative approaches to green wall systems that combine environmental and structural benefits. Observations have shown that there is typically low integration between vegetation and structural elements. This has led to the development of a new concept: integrating plants directly into the structure by incorporating them into construction materials. [9]

One promising approach is the use of bioreceptive concrete, which allows for the integration of microflora directly onto the building structure. This concrete encourages and sustain the growth of the microorganisms such as mosses lichens and algae directly on its surface and thus increases the cryptogamic cover of the material. One of the essential characteristics of bioreceptive concrete is its significantly lower pH compared to conventional concrete, which typically ranges from 12 to 13. In contrast, the pH of bioreceptive concrete is between 7 and 9, creating a more suitable environment for the establishment of small plants and microorganisms shortly after construction is completed. Although this solution is still in the development phase, the ongoing effort to create greener and more sustainable cities highlights the potential of these new types of bio-active cementitious materials to reduce the ecological footprint of concrete-based infrastructure.

BIORECEPTIVITY OF MATERIALS

As efforts to integrate plants into structural materials increase, it is crucial to understand the relationship between living organisms and building materials. Traditionally, the natural colonization of building materials has been studied from a negative perspective, with a prevailing belief that microorganisms harm structural materials through biodegradation or biodeterioration. These terms refer to the undesirable chemical and physical changes in materials caused by living organisms. However, colonization does not necessarily lead to the degradation of structures. It can primarily result in color changes that may not only be harmless but also aesthetically pleasing, positively impacting the environment. Guillitte´s study [10] of bio-colonization effects on materials introduces the term "bioreceptivity," which refers to the ability of materials to be colonized by living organisms. This concept encompasses the impact of colonization on the material without necessarily causing its deterioration. It also implies the material properties necessary for the attachment and development of vegetation on material surfaces, such as porosity, roughness, moisture, and the chemical composition of the surface layer.

Guillitte [10] categorized bioreceptivity into three types: primary, secondary, and tertiary. The primary bioreceptivity refers to the initial state of colonization when the material properties remain unchanged or only slightly altered. Over time, bio-colonization can lead to changes in material properties, establishing the secondary bioreceptivity. Further modifications of these secondary characteristics by human activities, such as particle consolidation or surface polishing, are termed the tertiary bioreceptivity. This new perspective on biological colonization in civil engineering introduces a novel concept for building and ecology.

To be colonized by living organisms such as algae, fungi, and lichen, certain conditions must be met for the reception and development of these organisms. Crucial factors for colonization include environmental conditions and the chemical and physical properties of the material. Bioreceptivity encompasses material properties conducive to colonization. However, the degree of bio-colonization depends not only on material properties but also on environmental factors.

The importance of environmental conditions such as temperature, light, water, and material exposure was discussed in Miller's [11] studies on the bioactivity of stone materials. Natural stone, widely used in monuments, is a preferred object for colonization due to its surface roughness, pore space structure, and permeability, which are favorable for bioreceptivity regardless of environmental conditions. Another study suggests that environmental factors, particularly water availability and light regime, play an even more significant role in bio-colonization [12]. Despite ongoing debates about the relative importance of environmental versus material properties, it is evident that both are crucial for biological colonization. While water availability depends on surrounding conditions, the ability to capture and retain water depends on the material's porosity and roughness [11].

In summary, bio-colonization of stone materials depends primarily on environmental conditions, microclimatic parameters, and the material bioreceptivity. Further investigations are needed to fully understand the extent to which material properties are affected under certain conditions. However, knowledge of natural stone bioreceptivity can inform us about other materials' susceptibility to colonization, such as concrete.

BIO-ACTIVE CONCRETE

Concrete is the most widely used building material. In the second half of the 20*th* century, the construction industry focused primarily on using of Ordinary Portland Cement (OPC) [13]. With the increasing demand for environmentally friendly and sustainable solutions in the construction industry, alternatives to conventional concrete are being sought. These alternatives aim to not only to decrease the environmental impact of OPC production but also to implement greener solutions within urban infrastructure. Consequently, a new concept has been developed: integrating microflora directly onto concrete structures by enhancing the bio-receptive properties of concrete.

To achieve this, the chemical and physical properties of concrete must be modified, specifically its pH, porosity, and roughness. Traditional concrete has very high initial alkalinity and a low porous structure, which are not suitable conditions for bioreceptivity. Its pH ranges from 12 to 13, while the suitable pH value for the growth of microorganisms is between 5.5 and 8.5 [14]. Typically, biological colonization can only occur after the pH decreases due to carbonation. Several methods exist to lowe the alkalinity of concrete. Additives such as silica fume and fly ash can reduce alkalinity, although the pH still fluctuates around 10, depending on the amount of cement replacement. Another method involves adding an acid solution to the mixture, which can negatively affect material properties [2]. This has led to the development of alternative hydraulic binders with naturally low pH. These types of cement, typically composed of oxides and phosphate acids, offer a viable alternative to Ordinary Portland Cement. The most widely used acid-based cement is Magnesium Phosphate Cement.

Magnesium phosphate cement (MPC)

Magnesium Phosphate Cement (MPC) is a relatively new type of binder. It was first used at the end of the 19th century as dental cement and later applied mainly for construction repairs due to its excellent mechanical properties [15]. Compared to Ordinary Portland Cement (OPC), MPC offers several advantages, including very quick setting time, high early strength, low drying shrinkage, and very high bonding strength with old concrete. These properties are crucial in repair construction, which is why MPC is mainly used as mortar for rapid repairs of concrete structures, such as pavements, airport runways, and bridge decks [16]. MPC is derived from the reaction between phosphate and metal oxide. Three types of phosphate salts are generally used: ammonium, potassium, and sodium. However, ammonium is restricted to outdoor use due to the release of ammonia during the reaction [15]. The reaction between oxide and phosphate in the presence of water is very rapid, necessitating the addition of a retarder to control the setting time.

The mechanical properties and setting time of MPC depend on the water/cement (w/c) ratio, the addition of a retarder, and the ratio of phosphate to magnesium (P/M) [17]. The strength is highly influenced by the P/M ratio; a decrease in the ratio leads to increased strength. However, a high amount of magnesium causes very quick hydration and high heat release, potentially damaging the product. Therefore, an optimal P/M ratio must be chosen. According to an experimental study [18], the maximum compressive and flexural strength are achieved with a P/M ratio of 0.2−0.25. Similarly to OPC, the w/c ratio is an important parameter, with decreasing w/c ratio resulting in increased compressive strength [19]. MPC develops early strength very rapidly, reaching 70% of its strength within 3 hours. This rapid early strength development is positively affected by the high hydration heat of MPC [16]. Additives such as fly ash can be used up to 50% to improve properties, adjust color, and reduce overall cost [19].

Many research efforts have focused on phosphate cement-based materials, primarily examining the chemical compositions and mechanical properties of mortar used for concrete repair. However, there are only few studies on the bioreceptivity of this concrete substrate, a critical property for the successful application of green walls [20]. Bioreceptivity is expected to become one of the most important properties of these materials, and further studies are being conducted to optimize this new cementitious material.

OPTIMIZATION OF BIO-ACTIVE CONCRETE MIXTURE

Development of low pH concrete

For development of low-pH cementitious materials, Magnesium Phosphate Cement (MPC) was chosen as a hydraulic binder to improve bio-receptive properties of concrete. [21]

Materials

MPC is prepared by mixing MgO (M) and $NH_4H_2PO_4$ (P) with a retarder in a given proportion. Dead-burned magnesia (MgO) calcinated in temperature over 1400 ℃ with low reactivity was used. The content of MgO was at least 89 % with the particle size 0−0.1 mm. For the retarder, 6 % of Borax was used as a weight of a total cement mix. The reaction of MPC is acid-based neutralization and it is strongly exothermic. The main reaction product is Struvite [22].

$$MgO + NH_4H_2PO_4 + 5H_2O \rightarrow MgNH_4PO_4 \cdot 6H_2O$$

However, the reaction is still not well understood.

Samples preparation

In order to obtain the best pH values, the samples of different P/M ratios were tested ranging from 1:1 − 1:1.75. The mixes are summarized in Table 1.

Tab. 1 - pH value for various MPC mixes tested in a range of 28 days.

Sample mix: 60 g of MPC, 18 g / 15 g water, 6 % of retarder					
P/M	B (%)	w/c	pH at 1 d	pH at 4 d	pH at 28 d
1:1	6	0.25	6-7	7	7
1:1.5	6	0.3	6-7	8	8
1:1.75	6	0.3	6-7	8	8-9

The cement paste was prepared by mixing the solid components first in a dry form and then with water to create a cement paste. For the good workability, the optimum w/c ratios of 0.25 and 0.3 were used for the specimens. The content was then poured into the molds of circular shape with a diameter of 50 mm. All the samples were demolded after two hours (Figure 4).

Fig. 4- Samples of different MPC mixes, from left P/M: 1:1, 1:1.5 and 1:1.75.

These samples were then tested for their pH value. The surface of the samples was cleaned with dry cotton and then the fresh water was dropped on it with a small plastic squirrel. After the 60 s the pH strip was inserted into the water on the surface. The color of the strip was then compared to the color chart. The pH was measured after 1 day, 4 days and 28 days.

Evaluation of pH

While all results ranged from 6−9 pH over a period of time, the general trend observed is that the pH value increases with the decreasing P/M ratio. It was observed that with time up to 4 days, the pH gradually increased, reaching a stable value between 4 and 28 days. The resulting tests indicate that all mixes are suitable for the targeted pH of 5.5−8.5 with a slightly more alkaline solution for the ratio of 1:1.75, see Table 1.

Alteration of physical properties of concrete

Although microorganisms are capable of adapting to their environments, certain conditions and material properties can be optimized to enhance the desired biological development. High surface roughness and macroporous textures increase the wall ability to retain water, creating a moist environment that supports floral growth. Consequently, porosity was the primary physical property investigated in this study.

Porosity

The pore structure of concrete is a crucial characteristic that significantly influences its mechanical properties, including strength, elasticity, and creep strain [23]. Typically, efforts are made to minimize pores in concrete to create a well-compacted structure with low porosity. However, for our purposes, the goal was to create a porous structure to better accumulate and retain water on the surface. Porosity in concrete is influenced by factors such as the water-cement ratio, aggregate size distribution, and the degree of compaction of the cement-based material. In this study, the focus was on using different aggregate sizes in the concrete mixture to characterize porosity.

Usually, Mercury Intrusion Porosimetry is used to characterize pore structure. A major limitation of this technique is that interpretation of results involves assumptions about pore shape that might lead to underestimation of the fraction of larger pores. As thoroughly analyzed by Lange et al. [24], image processing techniques might be more suitable for quantification of larger pores. As for bioreceptive concrete the number of larger pores (able to retain water and moist environment that supports floral growth) is crucial, we used image processing to determine optimal concrete mixture.

Five samples with different grading curves were prepared (see Table 2 and Figure 5). Each 1-liter concrete mixture was prepared from 500 grams of cement, 1600 grams of aggregates, and a water-cement ratio (w/c) of 0.3 for all samples. Aggregate sizes ranged from 0−0.25 mm to 3−4 mm. Ordinary Portland Cement (OPC) was used instead of Magnesium Phosphate Cement (MPC) in these initial tests to save on materials and costs.

Tab.2 - Variation of aggregates for concrete samples made from OPC.

Mixture (1l): Cement I 42,5 R- 500g, water -175g, Stachema 2000- 15g					
Aggregate Size (mm)	Weight (g)	Aggregate Size (mm)	Weight (g)	Aggregate Size (mm)	Weight (g)
Sample 1		Sample 2		Sample 3	
3-4	-	3-4	-	3-4	-
2-3	160	2-3	320	2-3	480
1-2	480	1-2	-	1-2	-
0.5-1	480	0.5-1	640	0.5-1	640
0.25-0.5	320	0.25-0.5	400	0.25-0.5	320
0-0.25	160	0-0.25	240	0-0.25	160
Total	1600	Total	1600	Total	1600
Sample 4		Sample 5			
3-4	160	3-4	160		
2-3	480	2-3	480		
1-2	-	1-2	240		
0.5-1	480	0.5-1	320		
0.25-0.5	320	0.25-0.5	240		
0-0.25	160	0-0.25	160		
Total	1600	Total	1600		

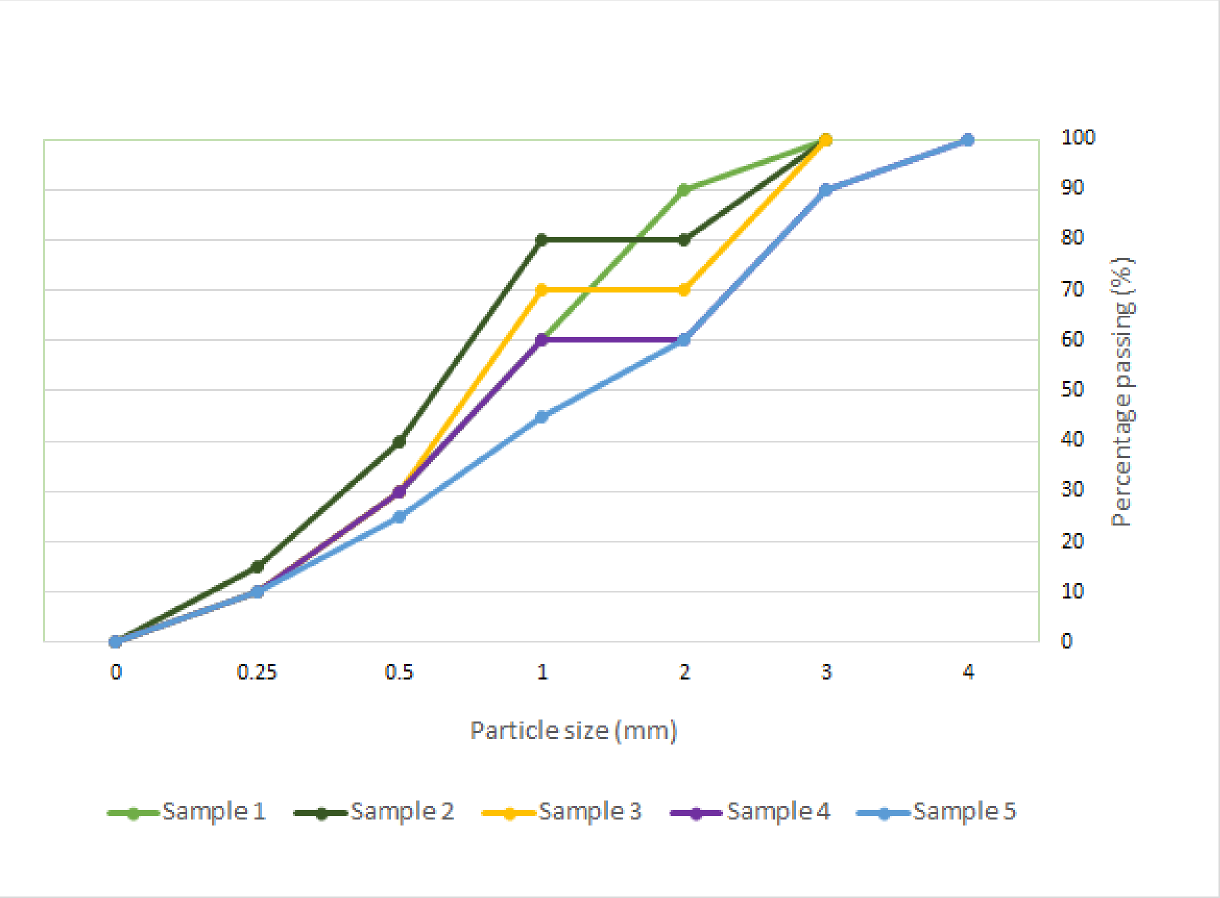

Fig. 5 - Aggregate grading curves

The images of surfaces of individual samples (see Figure 6) were first transferred to greyscale, then Gaussian filtering [25] was applied. For thresholding, high-value cutoff threshold was applied first, then Otsu method was used [26]. Flood fill [27] was used to analyze each pore separately, so that pore size distribution can be determined.

Due to limitations of image resolution pores smaller then 0.25mm could not be reliably analyzed. While these might contribute to overall porosity significantly [28], for bio-active concrete these are less relevant, as not accessible by organisms. Pore size distribution for pores larger than 0.25mm obtained by image analysis is displayed in Fig 7. Fig 8 shows cumulative porosity (when pores > 0.25mm considered). It can be seen that mixture 4 is coming with largest proportion of pores greater than 0.25mm. Therefore mixture 4 was then selected as the most favorable option for preparation of MPC sample tiles.

To confirm that porosity results obtained with OPC are applicable also for MPC an MPC sample was prepared using mixture 4. Figure 9 shows that the porosity of MPC and OPC samples is indeed comparable.

Fig. 6 - Samples (4 x 3 cm) of concrete with different aggregate grading curves

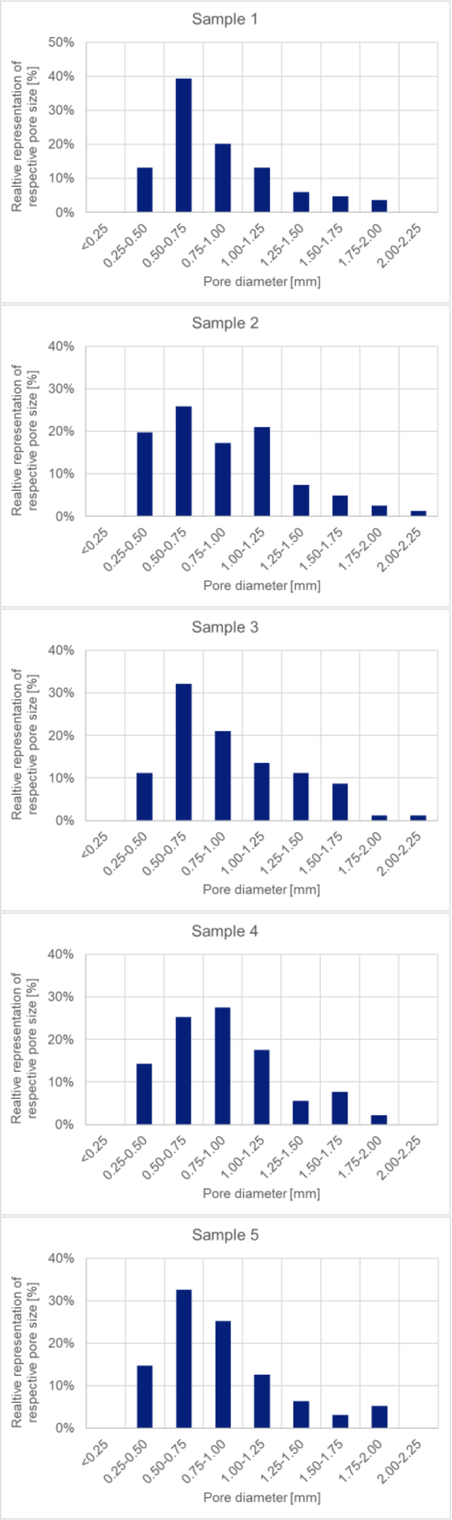

Fig. 7 - Pore size distribution for pores larger than 0.25mm obtained by image analysis for samples 1 - 5

DOI 10.14311/CEJ.2024.04.0034

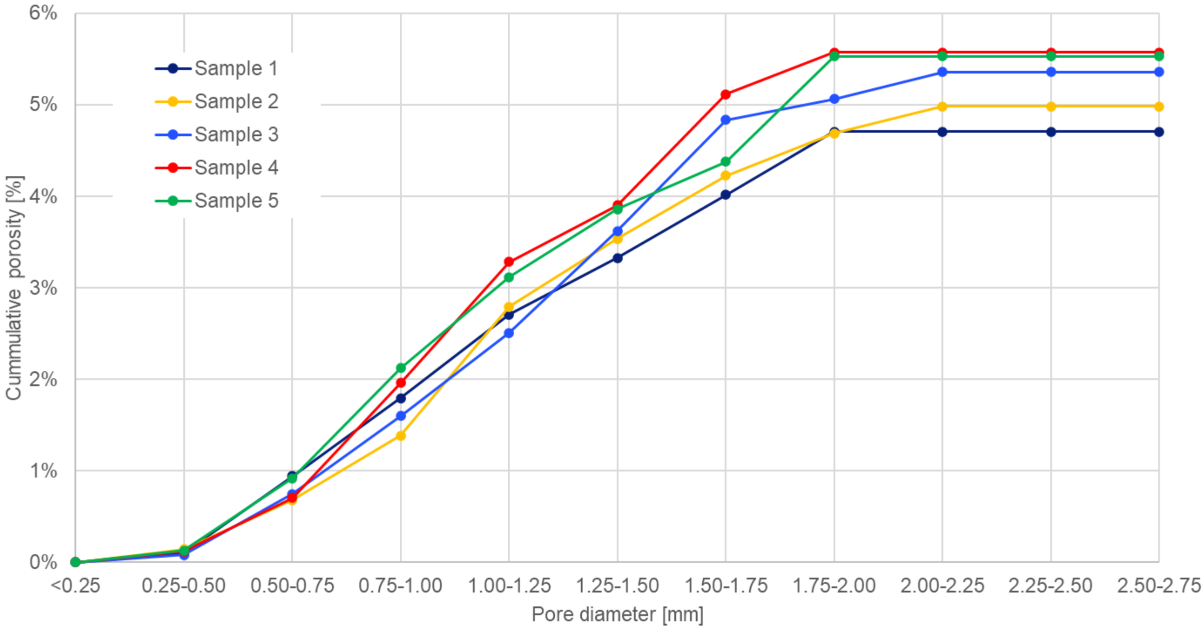

Fig. 8 - Cumulative porosity for pores > 0.25mm

Fig. 9 - MPC sample (left) and OPC sample (right)

APPLICATION OF THE OPTIMIZED MIXTURE

Building on the optimization presented above, two types of bio-active concrete tiles (250 x 350 mm) were fabricated using digital fabrication and CNC milling, as described in [29]. One type of tile produced is a Standard tile that can be used as a wall tile, and the other type with an integrated pocket in which soil can be placed and planted with a smaller plant and is called a Planter tile. The two prototype tiles produced are shown in Figure 12.

The concreting of the tiles was conducted in two sequential phases. Initially, a layer of magnesium phosphate cement concrete, prepared with a dry consistency, was applied. This mixture was spread manually across the tile mold without extensive consolidation to preserve its porous structure. To enhance the bonding between the two layers, the surface of the magnesium phosphate cement was deliberately roughened. Subsequently, a second layer consisting of ordinary Portland cement concrete was poured atop the initial layer. A close-up view illustrating the interface between the two distinct concrete layers is presented in Figure 10. The anchors required for the eventual installation of the tiles can be placed in the OPC concrete bearing layer.

Fig. 10 - Detail view of the contact between two different concrete layers

Although the manufactured element is not load-bearing and the mechanical properties of the concrete such as compressive strength are not critical to its functionality, three MPC samples were fabricated to test the compressive strength of the concrete during the production of the tile prototypes. The compressive strength of our specimens after three days averaged 21.6 MPa, with a maximum deviation from this average of 3.8 MPa. Conventional OPC concrete with a defined strength of C20/25 was used for the bottom layer of the tile.

Figure 11 clearly shows the porosity and roughness of the bioreceptive concrete layer, prepared according to the grading curve of sample 4 (see Table 2). The load-bearing part of the tile was made from non-porous OPC concrete.

Fig. 11 - Prototype tile produced using the optimized mixture - detail of rough and porous surface of tile.

DOI 10.14311/CEJ.2024.04.0034

The pH values of the concrete tiles reduced as expected. A P/M ratio of 1:1.5 was used for the cement mixture with a 6% addition of Borax. The pH reached 6−7 after one day and increased to 8−8.5 after four days, stabilizing at this level thereafter.

The tiles were installed in an outdoor environment to observe plant coverage development and analyze the long-term performance of the tiles (see Figure 12). Further investigation is necessary to evaluate the benefits of an entire wall composed of bio-active concrete tiles.

Fig. 12 - Prototype tile produced using the optimized mixture - outdoor installation

CONCLUSION

A novel way of greening our cities has been proposed and is being tested. A surface layer of bioreceptive concrete was added to the supporting concrete, when the added layer can be shaped in different ways. In order to make this layer bioreceptive, it was made with a lower pH than that for normal concrete and with a higher porosity for easier plant attachment.

To produce a low-pH cementitious material, Magnesium Phosphate Cement, as a special type of concrete binder, was utilized. The P/M ratios employed in the samples met the pH conditions necessary for microorganism growth. The decrease in the P/M ratio resulted in an increase in the pH value. Among the concrete mixes and mortars tested, although a P/M ratio of 1:1.75 gave a slightly higher pH value than the 1:1.5 sample after 28 days, the effect of increasing pH was not as pronounced. Considering the cost effectiveness, the final mixture with a P/M ratio of 1:1.5 was selected for the bioreceptive layer with the guaranteed pH value within the suitable range for flora of 5.5 to 8.5.

The addition of larger aggregates, particularly aggregates of 2−3 mm and 3−4 mm, resulted in increased surface porosity and roughness. This enhancement in terms of increased porosity, textured surface and modified chemical composition created an environment favorable for the embedment and growth of microflora. By facilitating vegetation growth and water retention on concrete surface, the tile presents a solution for improving both the initial and maintenance cost of green walls.

The negative effect of the increased porosity of the bioreceptive layer on its durability, especially due to the freezing of the retained water, is corrected by the fact that the material used for the bioreceptive layer is commonly used for quick repairs. Therefore, the expected damage of the bioreceptive layer, when the flora will hide the damaged parts initially, can be repaired with the same material, which will turn green quickly again. It should be noted that the load bearing layer, which is also often reinforced with steel, and thus requires a higher pH, will remain intact.

ACKNOWLEDGEMENTS

This work was financially supported by the Czech Technical University in Prague, project no. SGS24/041/OHK1/1T/11, which is gratefully acknowledged.

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