



Development of a neutral cementitious material to promote vegetation concrete



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HIGHLIGHTS

- A neutral binder using alpha-calcium sulfate hemihydrate (α -CH) was developed.
- The mass fraction of $\text{Ca}(\text{OH})_2$ was found to be approximately 11% of cement content.
- The microstructural characteristics and strength gain of α -CH were established.
- The use of cement is impractical for reducing the alkalinity of concrete.
- The developed α -CH binders possesses potential for production of vegetation concrete.

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ABSTRACT

To develop a neutral binder for vegetation concrete, various cementitious materials based on alpha-calcium sulfate hemihydrate (α -CH) were compositionally examined as follows: 25% and 50% of the α -CH content were partially replaced by a mineral admixture; and 25%, 50%, 75%, and 100% were replaced by ordinary portland cement (OPC). The hydration products and microstructural characteristics of the prepared binders were investigated using X-ray diffraction analysis and scanning electron microscopy. The portlandite [$\text{Ca}(\text{OH})_2$] phase formed in the hydrated cementitious materials was quantified by thermogravimetric analysis to examine the relationship between $\text{Ca}(\text{OH})_2$ content and pH values of the pastes. The OPC paste without α -CH showed several diffraction peaks for the $\text{Ca}(\text{OH})_2$ phase, whereas the α -CH paste had no diffraction peak at the angles corresponding to $\text{Ca}(\text{OH})_2$. The dehydration mass fraction of $\text{Ca}(\text{OH})_2$ corresponded to approximately 11% of the cement content included in each paste sample. The 28-day compressive strength of α -CH mortar was approximately 55% of that of OPC mortar, whereas the α -CH-based pastes without OPC maintained the pH at the neutral level, between 6.5 and 6.9, showing that the use of cement, even in very low amounts, is impractical for reducing the alkalinity of concrete. The α -CH possesses significant potential for practical production of neutral vegetation concrete.

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1. Introduction

Concrete produced using cement is commonly recognized as a high pH material because the hydrated cement generates a portlandite [$\text{Ca}(\text{OH})_2$] phase that has characteristics of an alkaline plume with a pH of 12.6 [1]. The high pH of cement encourages the enhancement of the durability of concrete elements and the prevention of corrosion of the reinforcing steel bars embedded in concrete [2], whereas the pH liquation from the pore water solution in the concrete is unfavorable to ecological environments, as it affects water purity and plant growth. Although vegetation

restoration concrete techniques [3,4] and ecological porous concrete approaches [5] have been promoted in various areas such as foamed shotcrete for slope protection and green roof gardens on buildings and urban greening industries, there are few attempts to reduce the pH value of hydrated cementitious materials in vegetation concrete. The generally accepted optimum values of the pH for vegetation concrete range between 6 and 8 because plant growth regulating factors such as enzyme histochemistry, microbiological characteristics, and ion exchange catalysts are energetically activated under a neutral condition [6,7].

Most efforts to reduce the pH value in vegetation concrete have focused on porous concrete using mineral admixtures [8–10]. A literature review [11,12] on the pH of cement concrete pointed out that mineral admixtures with a high silica content are effective

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in consuming the $\text{Ca}(\text{OH})_2$ phase in hydrated cement. However, the addition of mineral admixtures for partial replacement of cement is an impractical approach in making concrete with pH below 11 [13–15], though leaching analysis using deionized water measurement method results in low alkalinity in the long term. The pH value of cement-based concrete suspended in distilled water tends to decrease gradually with time at the surface but is in the stationary state at the core, which can lead to distorted pH values. Yang [15] measured pH values using powdered concrete in accordance with ASTM D4972 [16] and demonstrated that concrete produced using high-volume mineral admixtures becomes highly alkaline with a $\text{pH} \geq 11$. Although the pozzolanic reaction of mineral admixtures consumes $\text{Ca}(\text{OH})_2$ in the hydrated cement, the alkali ions such as Na and K gradually bind to calcium-silicate hydrate (C-S-H) gels, which contribute to an increase in the pH in cement concrete [1]. Overall, the use of the conventional cement, even in very low amounts, cannot guarantee the production of vegetation concrete with neutral pH values.

The present study attempted to develop a binder with neutral pH values (ranging between 6 and 8) using an alpha-calcium sulfate hemihydrate (α -CH) as a part of a fundamental investigation to produce artificial soil aggregates for plant growth [15]. Considering decreased cost and strength development of the α -CH-based binder, ordinary portland cement (OPC) and mineral admixtures such as fly ash (FA) and ground granulated blast-furnace slag (GGBS) were used as a replacement for the α -CH. The pH values and compressive strength development of the prepared binders at different ages were measured in accordance with ASTM D 4972 and ASTM C39 [16], respectively. The hydration products and microstructural characteristics of the prepared binders were traced using X-ray diffraction (XRD) analysis and scanning electron microscopy (SEM). Moreover, the portlandite [$\text{Ca}(\text{OH})_2$] phase formed in the hydrated cementitious materials was quantified by thermogravimetric analysis in order to examine the relationship between $\text{Ca}(\text{OH})_2$ content and the pH values of the pastes produced using the prepared binders.

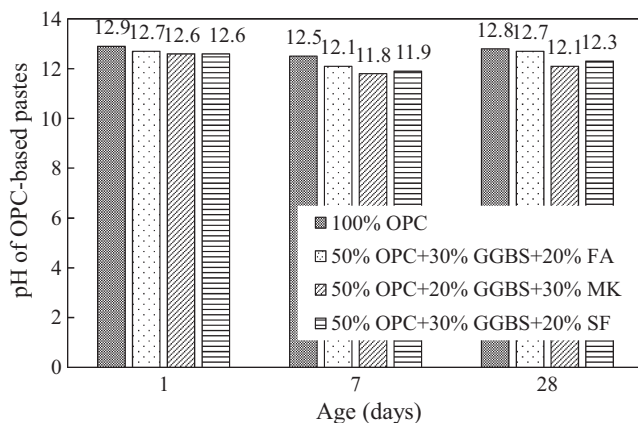


Fig. 1. pH values of OPC-based pastes with different mineral admixtures.

Table 1
Chemical composition of the selected cementitious materials (% by mass).

Materials	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	K_2O	Na_2O	TiO_2	SO_3	LOI [*]
α -CH	0.40	0.24	0.07	54.40	0.13	0.12	–	0.18	44.28	–
OPC	19.60	5.10	2.90	62.80	3.90	0.89	0.26	0.31	2.50	1.74
FA	55.70	26.60	5.08	4.70	0.67	0.57	0.37	1.53	0.68	4.1
GGBS	34.20	14.30	0.05	42.81	6.52	0.26	0.12	0.11	0.48	1.15

* Loss on ignition.

2. Experimental details

2.1. Materials

As a preliminary test, the effectiveness of various mineral admixtures in reducing the pH values in the hydrated cement-based pastes was examined. The pastes were produced using 50% OPC and different mineral admixtures with the following compositions: 30% GGBS + 20% FA, 20% GGBS + 30% metakaoline (MK), and 30% GGBS + 20% silica fume (SF). A control paste with 100% OPC was also prepared. The pH values of the prepared OPC-based pastes at different ages are shown in Fig. 1. The pH value of OPC-based pastes made using various mineral admixtures were mostly equivalent to that of 100% OPC paste, regardless of the age. This indicates that the addition of mineral admixtures with high silica content would be impractical in reducing the pH values of OPC-based concrete, although it is known that the silica ion in the mineral admixtures consumes the $\text{Ca}(\text{OH})_2$ phase in the hydrated cement.

To explore a cement-less binder with neutral pH during the hydration process, α -CH was selected as a source material. To examine the effect in the development of the compressive strength and decreasing the cost of the α -CH-based binders, GGBS and FA were added as a partial replacement of the α -CH. In addition, for examining the incorporation of α -CH and OPC, and for comparison with the hydration product of OPC pastes, α -CH was replaced by OPC at 25%, 50%, 75%, and 100%. The chemical compositions of these materials evaluated by X-ray fluorescence (XRF) are given in Table 1. The α -CH was manufactured from the conversion of gypsum dihydrate ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) by the conventional hydrothermal pressure method conducted under the reaction temperatures of 120–140°C within the autoclave system. Thus, the α -CH was mainly composed of 54.4% CaO and 44.3% SO_3 . The OPC (ASTM Type I) was mainly composed of 62.8% CaO and 19.6% SiO_2 . The used FA, belonging to Class F of ASTM C618 [16], had a low calcium oxide (CaO) content and a silicon oxide (SiO_2)-to-aluminum oxide (Al_2O_3) ratio by mass of 2.1. The GGBS had a high CaO content of 42.8% and SiO_2 -to- Al_2O_3 ratio by mass of 2.4, which conforms to ASTM C989 [16].

The XRD patterns of the virgin cementitious materials in the unreacted state are shown in Fig. 2. The SEM images of these materials are also presented in Fig. 3. The diffraction patterns of α -CH reveal that all peaks correspond to calcium sulfate hemihydrate, showing no peaks for dihydrate gypsum. The α -CH crystal grows longitudinally up, which forms a stick-shaped morphology. The predominant phase of OPC is alite ($3\text{CaO} \cdot \text{SiO}_2$), and the distribution of the peak pattern for belite ($2\text{CaO} \cdot 2\text{SiO}_2$) has a gentle wave shape. The alite peaks at $2\theta = 32^\circ$ overlap with the stronger belite peaks. The microstructure of the OPC can be characterized as an irregular hexagonal alite crystal, as identified by Taylor [17]. The diffraction patterns in unreacted FA are observed at an angle between $16^\circ < (2\theta) < 65^\circ$, attributed to quartz (SiO_2) and mullite ($3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$). The highest peak is observed at a 2θ angle of 27° . FA consisted mainly of spherical particles with a clean surface. The GGBS shows three characteristic peaks at a diffraction angle of $25^\circ < (2\theta) < 32^\circ$, attributed to akermanite ($\text{Ca}_2\text{MgSi}_2\text{O}_7$). Very weak peak intensities corresponding to anhydrite are also observed because a small

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