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Antifungal performance of BFS mortar with various natural antifungal substances and their physical properties

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ABSTRACT

This study discusses the basic physical properties, as well as the durability and antifungal performance of blast furnace slag (BFS) mortar with various natural antifungal substances. As a series of basic experiments, the antifungal performances of various natural antifungal substances extracted from Marjoram, Phytoncide, Thyme, Ginkgo leaves, and Chitosan were investigated on five types of mold: *Chaetomium globosum*, *Aspergillus niger*, *Aureobasidium pullulans*, *Gliocladium virens*, and *Penicillium pinophilum*. Marjoram and phytoncide extracts, which showed excellent antifungal performance in the basic experiment, were selected and applied to the antifungal mortars. The physical properties in addition to the durability and antifungal performance of the antifungal mortars were investigated. The results clearly showed that the antifungal mortars with the marjoram extract had excellent antifungal performance compared with other antifungal mortars with natural antifungal substances. Their antifungal performance was at an equivalent level to that of the mortars with organic chemical antifungal agents. It was also shown that the optimum addition rate of marjoram extract in the mortar was 3% by mass of binder, considering antifungal performance and durability. The adverse effect of marjoram and phytoncide extract on the consistency and strength of antifungal mortars was insignificant.

1. Introduction

Since the 1970s, airtight and sealed structural systems in buildings have been actively applied to increase the energy conservation effects in accordance with policy in Korea. This has led to the reduction of indoor ventilation rates, and in turn to an increase in the likelihood of dew condensation in buildings. This contributes to the development of favorable environments for the growth and germination of fungi due to the increase in the relative indoor humidity [1]. Sufficient evidence of associations with exposure to damp indoor environments and with the presence of fungi or other agents in damp indoor environments was shown with four outcomes: upper respiratory (nose and throat) tract symptoms, coughing, wheezing, and asthma symptoms in sensitized people [2]. Health effects associated with indoor dampness and fungi have been observed in many studies conducted in many geographical regions, and some evidence was found for dose–response relations [3,4]. However, the mechanisms remain unknown. Recently, healthy indoor environments and the improvement of indoor air quality have been highlighted as important issues in the health and life of residents, especially considering the contemporary living habits of people who tend to stay indoors more than 80% of the day. In Korea, the enforcement regulations on indoor air quality management law in multiple-use facilities was completely revised in May 2004, and construction companies were then obliged to reveal the indoor air quality of their apartment buildings. The microbial contamination on the surface of interior materials could significantly reduce the performance and efficiency of the materials (for example, the insulation performance in insulators), and accelerate their degradation and decay as well as contribute to very poor living environments due to odor [5]. However, studies on the biological contaminants (bacteria and fungi, etc.) that affect the health of residents have attracted little interest compared to chemical toxicity substances such as volatile organic compounds (VOCs) and formaldehyde emitted from interior materials, in spite of the increasing number of cases of residents' poor health and damage to interior materials due to fungi, bacteria, and insects.

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Only a few investigations on the biological contaminants of building materials have been performed. In addition, most of the investigations have focused on the development and improvement of a model to predict mold growth in building materials. Viitanen et al. [6] reported a numerical mold growth model for the evaluation of the response of humidity, temperature, and exposure time for mold growth on wood material. This model was based on comprehensive laboratory studies with Northern wood species [7]. Other types of models were also presented by Adan [8] and Clarke et al. [9]. Research on the further development of Viitanen's mold growth model was conducted by Vinha et al. [10] in order to adapt the model for analysis of structures made of various building materials for different climatic conditions.

On the other hand, experiments or investigations on antimicrobial building materials as an effective solution for inhibiting the growth and germination of harmful microbes are relatively scarce, especially on materials such as mortar or concrete. Ramachandran [11] briefly introduced the subject of fungicidal and insecticidal admixtures in cement mortar and concrete in his book, *Concrete Admixture Handbook*. Some effective fungicidal and insecticidal admixtures are listed in ACI 212.3R-91 [12], which provides the range of addition rates required from 0.1% to 10% by weight of cement. In his book *Properties of Concrete*, Nevile [13] also found that some microbes such as fungi, bacteria or insects can adversely affect the properties of concrete, and a number of special admixtures that are toxic to the attacking microbes need to be incorporated in the mix.

In this study, the antifungal performances of various natural antifungal substances on the five types of mold, which might be easily detected in the indoors and outdoors of buildings, were widely investigated. The durability and antifungal performance of blast furnace slag (BFS) mortars mixed with various natural antifungal substances were also investigated and compared with those of mortars with existing chemical antifungal agents, typically used in various fields such as painting or in interior finishing materials. The influence of natural antifungal substances on the basic physical properties of antifungal mortar was investigated.

Table 1
Properties of binder.

Composition	Blast furnace slag	Calcium hydroxide
Blaine fineness	8000 cm^2/g	Particle size: 25–43 μm
Density (20 °C)	2.9 g/cm^2	2.8 g/cm^2
Chemical composition	SiO_2: 33.1%, CaO: 41.0%	CaO: >70%
pH (20 °C)	–	>12.4
Appearance	White powder	White powder

2. Experimental methods

2.1. Overview

This study was carried out in three steps. In step 1, an investigation was carried out of the antifungal performances of various natural antifungal substances extracted from Marjoram, Phytoncide, Thyme, Ginkgo leaves, and Chitosan (oligosaccharide), which in many studies were found to have high antibacterial performance [14,15]. In step 2, various antifungal mortars were made using natural antifungal substances selected from the experiments of step 1. The influences of the addition rates and types of natural substance on the antifungal performance of the mortars were then investigated in detail. Finally, in step 3, the durability of the antifungal performance of the mortars was investigated using two methods: the long-term indoors exposure test and the water-immersion test. Moreover, the influences of the addition rates and types of natural antifungal substance on the basic physical properties of antifungal mortar were also investigated.

2.2. Materials

(1) Binder and aggregate

To verify the observations of mold germination and growth, and the development of eco-friendly antifungal building materials without cement, antifungal mortars were made using blast furnace slag (BFS) powder with calcium hydroxide ($Ca(OH)_2$) as a binder and standard sand as a fine aggregate. The properties of the binder are shown in Table 1. Standard sand was used as specified in the Korean standard, L 5100, predominantly graded to pass No. 30 (600 μm) and remain on No. 50 (300 μm).

(2) Antifungal agents

Two types of organic and inorganic chemical antifungal agents were used, which are typically used in various fields such as painting or in interior finishing materials [16], and five types of natural antifungal substances extracted from Marjoram (*Origanum majorana*), Phytoncide, Thyme (*Thymus vulgaris*), Ginkgo (*Ginkgo biloba*) leaves, and Chitosan oligosaccharide were used, employing a steam distillation method, as described in Table 2.

(3) Fungal species

Based on ASTM G-21:96 [17], five types of mold were used: *Chaetomium globosum* (ATCC 6205), *Aspergillus niger* (ATCC 9642), *Aureobasidium pullulans* (ATCC 15233), *Gliocladium virens* (ATCC 9645) and *Penicillium pinophilum* (ATCC 11797). These molds are easily detected in the indoors and outdoors of buildings in damp environments [18]. Freeze-dried strains from each of the fungi were provided by the American Type Culture Collection (ATCC, Manassas, USA).

2.3. Preparation and curing of antifungal mortars

The antifungal mortars were manufactured using the ratio of fine aggregate to binder by mass of 2.45:1, and the water-binder rate of the mortars was fixed at 0.50 in order to observe the effect on the fluidity during the addition of an antifungal agent. The binder was composed of a ratio of blast furnace slag to calcium hydroxide by mass of 0.9:0.1. All antifungal agents were directly added to the mortar at six concentrations of 0%, 1%, 3%, 5%, 10%, and 15% by mass of binder. Details of the mixes in the test program are given in Table 3. The disk-typed specimens of ∅ 40 mm × 5 mm were prepared for testing the antifungal performance of the

Table 2
Properties of antifungal agents.

Type (botanical name)		Component	Appearance	Efficacy	Application
Natural antifungal substances	Marjoram (*Origanum majorana*)	Sabinene, a-terpineol, terpinene	Extract (liquid state)	Antivirus, sterilization	Fiber, food and pharmacon industry
	Phytoncide	Sabinene, bornyl acetate, α-terpinyl acetate	Extract (liquid state)	Antibacterial, antifungal	Fiber and cosmetic industry
	Thyme (*Thymus vulgaris*)	Carvacrol, thymol, linalool	Hydrosol (liquid state)	Antibacterial, antioxidant, sterilization	Fiber, food and pharmacon industry
	Ginkgo leaves (*Ginkgo biloba*)	Flavonoid, ginkgolide	Extract (liquid state)	Antibacterial, antifungal, deodorization	Food and pharmacon industry
	Chitosan	D-Glucosamine, N-acetyl-D-glucosamine	Oligosaccharide (liquid state)	Antibacterial, antifungal	Fiber, food and cosmetic industry
Chemical antifungal agents	Organic	Isothiazoline-carbamate	White aqueous solution	Antibacterial and antifungal	Painting and interior finishing materials
	Inorganic	Zeolite	White powder (0.1–10 μm)	Antibacterial and antifungal	Painting and interior finishing materials