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Preparation and polymeric encapsulation of powder mineral pellets for self-healing cement based materials

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HIGHLIGHTS

- Powder mineral pellets were developed for self-healing cement-based materials.
- Pellets were encapsulated in a PVA based film coating.
- The coated pellets partially replaced the natural sand in mortar mixtures.
- PVA coating retained integrity and stability in concrete environment.
- Pellets showed good compatibility with the mortar specimens.

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ABSTRACT

Using mineral additives and admixtures as self-healing agents in cement-based composites has been extensively researched. However, if the minerals are added directly to the cementitious matrix without any protection, they could immediately react, leading to a decrease in self-healing efficiency with further associated side-effects on the mechanical properties of cementitious composites. Thus, this paper describes the development of coated pellets as a self-healing system in cement based materials. Pan pelletisation was utilised for producing pellets from three different powder minerals as potential healing agents: reactive magnesium oxide (MgO), silica fume and bentonite. Of these materials, two types of developed prototype pellets in addition to another two commercial types of MgO pellets with different pellet sizes were then encapsulated in a polyvinyl alcohol (PVA) based film coating. The PVA coating was evaluated for the apparent solubility in water and in alkaline solution, swelling property, water permeability, dynamic mechanical properties, and shell thickness on pellets. Although PVA coating exhibited a decrease in its mechanical properties in water or in the simulated concrete environment, it retained integrity and stability in both environments. The PVA shell thickness varied from 10 to 50 μm. As the coated pellets partially replaced the natural sand in mortar mixtures, they were characterised in comparison with sand for the particle size distribution, density, porosity, crushing strength and particle shape. Experimental results indicated that the different types of pellets showed higher porosity and lower crushing strength compared to sand. In mortar mixtures, the pellets showed excellent compatibility with minimal influence on the fresh mixture properties and the compressive strength of the hardened specimens. This was accompanied by good distribution inside the concrete matrix. Further investigations on the self-healing performance of these pellets are under way.

1. Introduction

Concrete outperforms other commonly used construction materials as a long-lasting and durable material. However, it is prone to cracking due to its limited tensile strength. The use of reinforcement can improve tensile load carrying capacity but cannot completely prevent crack formation. Although cracks are not considered as a damage, provided a prevailing crack width criterion is not exceeded, they are nevertheless undesirable. These cracks provide pathways for ingress of water, carbon dioxide, acid rain and other aggressive agents. The latter can not only induce corrosion of steel reinforcement but also degrade the concrete and accordingly the service life of reinforced concrete structures is shortened. In addition, cracks cause leakage in concrete structures, such as water reservoirs, roofs and water pipes, negatively affecting their

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functionality [1]. Thus, maintenance and repair work are essential and indispensable. The conventional repair methods increase the associated life-cycle cost of concrete structures, have a significant environmental impact and demand long and intensive labour. This led researchers to develop concrete that had a sufficient healing capability; cracks in concrete could be self-healed after cracking under specific conditions without any human intervention. This paved the way for infrastructures with reduced lifetime costs, improved durability and service life, both from an economic and environmental point of view [2,3].

Cement based materials possess a certain capability to heal cracks intrinsically, which is called autogenous healing. This capability is mainly governed by chemical causes such as further hydration of cement grains and the formation of calcium carbonate [2,4]. However, this autogenous healing is limited to microcracks (preferably less than 50 μm) while the presence of water is essential [5,6]. Using fibres can effectively improve the autogenous healing due to their capacity to control crack width and enhance multiple crack formation [7]. In an important step-forward, many researchers have explored the possibility of developing cement-based materials that heal autonomously. To achieve this, many techniques have been proposed such as the addition of different mineral admixtures, bacterially induced carbonate precipitation, or using microcapsules or hollow fibres as carriers for healing agents.

The direct inclusion of mineral additives and admixtures in the concrete in small dosages has been reported in several studies [8–10]. Despite the capability of these minerals to produce more healing products to fill the cracking area, they face certain disadvantages such as: premature consumption and uncontrolled reactivity in contact with water when directly added in the mix - with other ingredients- without any protection [11]. Moreover in the case of expansive agents, expansion can occur in the interior matrix as well as in the cracked area, which could cause further cracking [1,12]. Thus, researchers have suggested to envelope these minerals into capsules, glass tubes or coatings to be activated only at the time and location of cracking. For instance, Lee & Ryou [13] have investigated a coating encapsulation technique to envelope granulated calcium sulphoaluminate (CSA) into a polyvinyl alcohol (PVA) film. It was found that the PVA film coating could control the time of autonomic healing and prevented water migration via crack closing. Results also indicated an improvement of the healing efficiency of CSA in comparison with the direct application of healing agents in concrete without any coating. A recent study has also proposed to protect water soluble nutrients required for a special type of bacterial spore in a geopolymer coating [14]. Such coating will break whenever a crack occurs in the concrete matrix releasing the nutrients in the crack planes allowing the bacterial spores to germinate and precipitate calcium carbonate to heal the crack.

Three requirements need to be met in order to develop any encapsulation technique for self-healing of cement-based materials. Firstly, the encapsulation system should have compatibility and sufficient bonding with the concrete matrix. Secondly, it should be strong enough to preserve the sequestered healing compounds from unexpected events during the concrete mixing process, but brittle enough to rupture and release the healing agents when necessary. Thirdly, it should be chemically stable in the cement matrix, which is highly alkaline [14,15]. The success of any system depends on the likelihood of two elements: (i) the crack path to coincide with the location of the delivery system and (ii) the volume of healing agent to be released locally, since the latter is not ubiquitous in the matrix but concentrated at specific locations [16]. Similarly, the cargo material, in addition to promoting healing, has to disperse properly within the crack whilst having considerably extensive shelf life [16].

Three different powder minerals were examined in this study as core materials for the prototype pellets. A reactive magnesium oxide (MgO) was selected as the main potential self-healing agent in all formulations. MgO is an expansive mineral, able to yield irreversible and stable hydration products with compatible characteristics with the cement matrix [17]. Silica fume (SF), a silica-based additive was also tested for its potential to enhance self-healing. SF does not possess hydraulic behaviour by itself but can react with free $Ca(OH)_2$ and water, produced during the hydration of cement to form stable insoluble and densified calcium silicate hydrate (C–S–H) [18,19]. Thus many researchers have proposed it for concrete repair and strengthening purposes [20,21]. Bentonite (B) was also used as a dual-purpose material in some formulations. It is commonly used as a binder in the production of pellets [22,23]. Bentonite has adequate binding properties throughout the temperature range of operation; it is also easy to handle, abundantly available, and inexpensive [23]. It can also play another significant role in filling cracks due to its high swelling capacity. It swells 15–18 times its dry size when wetted by water [24]. It is noteworthy here that all of the above minerals have good compatibility with the cementitious matrix.

Polyvinyl alcohol (PVA) was selected as the coating material for the pellets in this study because of its excellent chemical resistance, physical properties, low cost and ease of preparation [25]. As PVA is a water soluble substance, its films are easily prepared by a casting evaporation technique from aqueous polymer solutions, thus avoiding the use of organic solvents. The resultant films are clear, homogenous and resistant to tearing [26]. PVA is produced by the polymerisation of vinyl acetate to polyvinyl acetate, which is subsequently hydrolysed to PVA. The degree of hydrolysation represents the extent to which polyvinyl acetate has been hydrolysed to produce PVA. Commercial PVAs are available as fully hydrolysed grades (degree of hydrolysis $\geq$ 98%) and partially hydrolysed grades (degree of hydrolysis ~86–89%) [27]. The solubility of PVA in water depends on the degree of hydrolysis and the degree of polymerization, with the effect of the former being especially significant. Some PVA grades with higher degrees of hydrolysis (>98%) are only soluble in hot water (50–100 °C); and form films that are insoluble in water at lower temperatures. In contrast, PVA grades with degrees of hydrolysis in the range of 75–98% are easily soluble in water. Molecular weight is another factor affecting the solubility of PVA and the extent of influence is related to the degree of hydrolysis. The solubility of highly hydrolysed PVA increases as the molecular weight decreases while the solubility of less hydrolysed PVA is relatively independent of molecular weight [26]. Thus, PVA with a high molecular weight and degree of hydrolysis of 98–98.8% was used in this study.

This paper aims to provide the basic design and characterisation concepts of developed coated pellets as a self-healing system in cement based materials. To achieve this, pelletisation of potential powder self-healing agents was investigated in this study. Polymer film coating was then employed to envelope the formed pellets. The laboratory-developed pellets were compared with other types of pellets, which are commercially available.

2. Materials

2.1. Healing agents and coating material

Different magnesium oxide (MgO) based pellets were tested in this study. These included two types of commercial pellets (CP) in addition to different formulations of laboratory prepared pellets (prototype pellets, referred to as PP). The two CP types were LUVOMAG MgO pellets supplied by Lehmann & Voss, Germany, and MagChem, hard-burnt MgO pellets, supplied by Martin Marietta