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Comparative study on the properties of three hydraulic lime mortar systems: Natural hydraulic lime mortar, cement-aerial lime-based mortar and slag-aerial lime-based mortar

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HIGHLIGHTS

- The differences between natural and artificial hydraulic lime mortars were revealed.
- NHL mortar have preferable fluidity compared to the artificial lime mortars.
- The mechanical properties of artificial lime mortar have advantages over NHL mortar.
- A part of artificial lime mortars is nearby to NHL mortar in environmental resistance.
- Three kinds of lime mortars have big difference in hydration and carbonation reaction.

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ABSTRACT

To evaluate the differences of properties between natural and artificial hydraulic lime mortars, the macro and micro properties of natural hydraulic lime (NHL2 and NHL5) mortars, white Portland cement-aerial lime-based (CL15 and CL25) mortars and slag-aerial lime-based (SL20 and SL40) mortars were studied through the examination of their physical and mechanical properties, environmental resistance properties, hardening mechanism and microstructure of lime mortars or pastes. The results show that the NHL mortars have preferable fluidity compared to the artificial CL and SL mortars, while the mechanical properties of artificial CL and SL mortars are advantageous compared to those of NHL mortar. In terms of environmental resistance, the water resistance of SL mortar is best, closely followed by the CL mortar, and the NHL mortar is relatively weak; moreover, CL mortar shows the best sulphate resistance, and SL mortar has the best alkali resistance. The hydration reaction (or pozzolanic reaction) and carbonization reaction could occur in the three kinds of hydraulic lime pastes, and the hydration reaction of the NHL and CL pastes mainly occur in the early stage of paste hardening (before 28 days), while the pozzolanic reaction of the SL paste reach a high degree at early hardening stage (28 days) and still maintains a certain reaction rate in the later period. The carbonation reaction process of the three kinds of lime pastes continues to the later period of the hardening, and the carbonation rate of SL pastes in the later stage would be obviously enhanced; furthermore, the carbonation degree of NHL paste is obviously higher than that of the CL and SL pastes. The different hydration-hardening process and mechanism of the three lime pastes lead to differences in the morphology and pore structure among them. There are more large-size pores in NHL5 paste (the pore volume in the range of pore diameter greater than 200 nm reaches 63.13%), while the pore size of the SL paste is relatively small (the pore volume in the range of pore diameter less than 100 nm reaches 44.15%). The porosity during the 180 days curing period was as follows: SL40 (27.32%) < CL25 (29.65%) < NHL5 (33.09%).

1. Introduction

Before the emergence of Portland cement, lime was widely used as a major binder material, and a large number of historical sites

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and research records show that lime was widely used in ancient buildings and was an ancient building cementing material [1]. Although the application range of lime is currently reduced due to the heavy use of Portland cement, some peculiar properties of lime cementitious materials such as moderate mechanical properties, good water vapour transmission performance and excellent durability are not available in cement and other cementitious materials [2]; therefore, lime is still being used in many areas and its application, and the attention devoted to lime has been enhanced. Especially, Portland cement and other cementitious materials have poor durability, which makes people re-recognized lime, an excellent cementitious materials, and expands its application fields, such as the application in ancient building restoration and external wall decoration mortar, and has very good prospects for further development.

Currently, lime cementitious materials can be divided into the aerial lime and hydraulic lime categories. Aerial lime generally refers to slaked lime, the essential component of which is $Ca(OH)_2$, and is an air hardening cementitious material usually used as lime mortar. The hardening mechanism is that of the calcium hydroxide reacting with carbon dioxide in the air under the action of water, generating calcium carbonate and releasing moisture. As a building material, traditional lime mortar has advantages of small drying shrinkage and good water vapour permeability but also shows disadvantages of low early strength, slow condensation hardening (24–48 h), poor water resistance and ready dissociation in the wet or water environments. Hydraulic lime, as a hydraulic cementitious material, has both hydraulicity and air hardening property, and is wisely used to prepare bonding, plaster or tick off seam mortar for use in the fields of the ancient building repair materials and external wall decoration mortar engineering. Hydraulic lime shows moderate strength, faster hardening speed (4–12 h), good water resistance, resistance to salt erosion and other advantages.

Currently, three kinds of hydraulic lime material systems are investigated in the research field. One is the natural hydraulic lime (NHL) produced by firing and slaking clay limestone or siliceous limestone, and its mineral phases are mainly dicalcium silicate and calcium hydroxide. The hardening process of NHL consists mainly of the hydraulic component (C_2S) that gives rise to a hydration reaction when combined with water [3] and forms calcium silicate hydrate (C-S-H), while calcium carbonate crystals are formed by the reaction of $Ca(OH)_2$ and CO_2 in the air under the action of water; the hardening mechanism can be expressed by the chemical reaction formulas (1) and (2):

$$2CaO \cdot SiO_2 + nH_2O \rightarrow mCaO \cdot SiO_2 \cdot kH_2O + (2-m)Ca(OH)_2 \quad (1)$$

$$Ca(OH)_2 + CO_2 + nH_2O \rightarrow CaCO_3 + (n+1)H_2O \quad (2)$$

Relevant research demonstrated that during the restoration of ancient buildings, NHL repair mortar is produced by NHL, quartz sand and other raw materials and possesses high compatibility with regards to the physical, chemical and mechanical aspects of ancient buildings, and it was found that soluble salt will not be introduced into the ancient building structure after the use of the NHL repair mortar [4]. The firing temperature of NHL is low (approximately 900–1100 °C), and less carbon dioxide is emitted during its production, so that the use of NHL for the preparation of mortar will greatly reduce the pollution of the environment [5]. Silva studied the early mechanical properties, pore structure, water absorption and water vapour transmission performance of air lime mortar for different mixing amounts of NHL. The results showed that with increase in the NHL mixing amount, the porosity and water absorption of mortar is decreased, while exhibiting a higher early mechanical strength and the water vapour transmission performance is decreased slightly [6]. The cement-lime-based cementitious material system is formed by adding a certain amount of cement to aerial lime and so that it shows both hydraulicity and air hardening. The hardening mechanism is similar to that of cement, mainly generating C-S-H, calcium aluminate hydrate (C-A-H), and $Ca(OH)_2$, as well as a small amount of ettringite through the hydraulic component of cement (C_2S, C_3S and C_3A), giving rise to a hydration reaction when combined with water that is accompanied by the carbonation reaction of $Ca(OH)_2$. The hardening mechanism of the cement-lime-based cementitious materials can be expressed by the following chemical reaction formulas (1)–(5):

$$3CaO \cdot SiO_2 + xH_2O \rightarrow mCaO \cdot SiO_2 \cdot kH_2O + (3-m)Ca(OH)_2 \quad (3)$$

$$3CaO \cdot Al_2O_3 + 6H_2O \rightarrow 3CaO \cdot Al_2O_3 \cdot 6H_2O \quad (4)$$

$$3CaO \cdot Al_2O_3 + 3(CaSO_4 \cdot 2H_2O) + 26H_2O \rightarrow 3CaO \cdot Al_2O_3 \cdot 3CaSO_4 \cdot 32H_2O \quad (5)$$

Ancient buildings require moderate strength and stability of physical, mechanical and structural properties after repair [4,7]. In the field of ancient building restoration, especially in the restoration of historic buildings, great damage is caused to the historic buildings owing to the high hardness, rigidity and impermeability of cement [8,9]. Silva investigated the properties of NHL-lime-based mortar and cement-lime mortar and found that cement-lime mortar show smaller porosity and lower water vapour permeability and higher mechanical strength [10]. Furthermore, supplementary cementing materials (SCMs)-aerial lime-based cementitious material is prepared by mixing an appropriate mineral admixture (fly ash, slag, silica fume, metakaolin, etc.) with pozzolanic activity in aerial lime; the pozzolanic materials contain a large amount of SiO_2 and Al_2O_3 components with hydraulic activity or latent hydraulically, giving rise to pozzolanic reaction with aerial lime under the action of water and generating hydration products such as C-S-H, C-A-H [11–16]. The hardening mechanism can be expressed by the following chemical reaction formulas (2), (6) and (7):

$$xCa(OH)_2 + SiO_2 + nH_2O \rightarrow mCaO \cdot SiO_2 \cdot (x+n)H_2O \quad (6)$$

$$xCa(OH)_2 + Al_2O_3 + nH_2O \rightarrow mCaO \cdot Al_2O_3 \cdot (x+n)H_2O \quad (7)$$

the uncarbonized $Ca(OH)_2$ dehydrate and recrystallize, so that a compact structure is formed with C-S-H to improve the strength. In recent years, researchers have studied that the effects of environmental conditions, curing temperatures and pozzolanic materials on the mechanical properties and durability of lime mortar [17–19] and extensively explored the theoretical basis and methods of the application of pozzolanic materials in cement concrete and hydraulic lime [15,20,21].

The properties and hardening mechanism of three kinds of hydraulic lime mortar systems are complicated and have advantages and disadvantages, but they have not been compared and the application fields have not been evaluated, resulting in a lack of clarity regarding the direction of main research and applications of each hydraulic lime system. Therefore, hydraulic lime mortar of three different systems are prepared from NHL, white Portland cement-aerial lime (WPC-AL) and slag-aerial lime (S-AL) cementitious materials in this work, and a comparative study of the properties of the three kinds of mortar with regard to physical and mechanical properties, environment resistance properties, composition of hydrated and hardened products, microstructure and pore structure was performed, thereby evaluating or revealing the differences in the properties and hardening mechanisms between the two common artificial hydraulic lime mortars and