



Pore structure and permeability of concrete with high volume of limestone powder addition

Chenzhi Li ^a, Linhua Jiang ^{a,*}, Ning Xu ^b, Shaobo Jiang ^a

^a College of Mechanics and Materials, Hohai University, Nanjing 210098, PR China

^b Nanjing Hydraulic Research Institute, Nanjing 210098, PR China

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ABSTRACT

Limestone is one of the preferred admixtures used in concrete due to its wide availability and environmental advantages. The durability of concrete is strongly dependent on its pore structure and permeability. To understand the microscopic mechanism underlying this behaviour, mercury intrusion porosimetry (MIP) and ultrasound waveform analysis are employed to evaluate concrete porosity, pore size distribution and the fractal dimension of pores. The results show that the porosity and mean pore diameter of concrete decrease slightly when up to 30 wt% limestone powder is added, though further increase in the limestone content reverses the trend. For the same limestone content, concrete porosity and mean pore diameter increase as the specific surface area of the powder decreases, or as the water-to-binder ratio increases. The results confirm close correlations between concrete permeability, porosity and mean pore diameter. The diffusion coefficient of chloride ions, measured by the rapid chloride migration (RCM) method, is found to decrease with increasing fractal dimension of the pore volume. Based on the percolation theory, the existence of a critical porosity is predicted, below which concrete permeability approaches zero. Three regimes, i.e. subcritical, critical and conventional diffusion regimes, are identified with increasing porosity. The implications of the percolation threshold on concrete permeability and corrosion propagation are discussed.

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1. Introduction

Concrete is widely used in civil engineering construction because of its adaptability, low cost, durability and other advantages [1]. One of the hallmarks of technological advances in modern concrete is the widespread application of mineral admixtures. The purposes of using admixtures are to replace a portion of cement and improve the performance of the concrete. Fly ash and slag are the most widely used mineral admixtures [2]. However, in recent decades, with increasing public awareness for water conservancy and rapid development of transportation and building construction, fly ash, slag and other admixtures have gradually become short in supply in some parts of China. In addition, fly ash and slag for infrastructure construction in northwestern China are transported over long distances, resulting in a significant increase in the cost of concrete production and construction. Therefore, it is imperative to find a mineral admixture that is well suited for the application, easy to use and cost-effective.

Limestone powder is a mineral admixture made from grinding of naturally occurring limestone. Compared with slag and fly ash, it is secured in availability and less expensive [3, 4]. The use of limestone

powder as an alternative concrete admixture can reduce the carbon footprint of concrete by approximately 15%, saving about 10% of cement raw materials [5, 6]. Addition of limestone powder in cement-based materials has a number of effects, including accelerated hydration [7], activation effect [8] and changes in particle surface morphology [9]. Limestone powder, when added to an appropriate portion, can act as nucleation matrix of C-S-H and accelerate cement hydration, which is beneficial to the early-age strength of concrete. The activation effect results from the reaction of limestone and the aluminate phase in the cement to produce a carbo-aluminate composite, which enhances the late-age strength of concrete. Particle morphology effect refers to the dispersion of dense and smooth limestone powder particles on the surfaces of cement particles. Due to their smaller size, the limestone particles can fill the interstices between the cement particles that form the skeleton, thereby improving the strength and workability of concrete.

The properties of Portland-limestone cement have been studied by several researchers. Kakali et al. [10] studied the hydration products of C₃A minerals with 0–30 wt% CaCO₃ at different ages. They concluded that CaCO₃ limits the transition from calcium carbonate (AFt) to mono-aluminate (AFm), while producing carbo-aluminates (Ca₄Al₂O₆·CO₃·11H₂O) to replace mono-aluminate. The addition of CaCO₃ does not change the type of hydration products, but it does affect

* Corresponding author.

E-mail address: hhulhjiang@163.com (L. Jiang).

the rate of formation of some hydration products. Nehdi et al. [11] believe that Portland-limestone cement has a higher early-age strength because the limestone powder accelerates the hydration of C_3S , especially when the limestone is finely ground. Bonavetti et al. [12] added limestone powder to concrete with low water-cement ratio and found that the hydration conversion of concrete could be as high as 80–85% at 28 d. When the water-cement ratio was 0.3 and the limestone powder was 10 wt%, the compressive strength was higher than that of concrete without limestone in the first 7 days before it decreased again. They attributed the increase in the hydration conversion and compressive strength to the effect of limestone powder on the early-age hydration of cement and the increase in the effective water cement ratio. Zhang et al. [13] found that concrete with 30 wt% limestone powder showed a lower compressive strength compared with concrete with the same content of fly ash at the same age, regardless of the curing method (standard curing or steam curing). Europe Self-compacting Concrete Project concluded that the self-compacting concrete containing limestone powder exhibits more preferable mixing behaviour because the powder improves the rheological properties and flow ability of self-compacting concrete [14, 15].

However, previous studies on limestone addition are far from comprehensive: (1) These works have mostly focused on the effects of limestone mix proportion, curing method and age on concrete properties and performance of concrete. Research on the effects of particle size on the pore structure of limestone-added concrete has rarely been reported. (2) In previous studies, limestone powder was used as an admixture to replace up to 35 wt% of cement. Little work has been done on pore structure when even higher portions of limestone powder are added. Researches along this direction are expected to pave the way towards a solution to the shortage of resources of fly ash, slag and other admixtures in remote areas, with significant economic and environmental impacts. (3) Previous studies have mostly focused on the hydration mechanism and the effect of limestone powder as binder on the mechanical properties of concrete, whereas the influence of limestone addition on concrete durability is scarcely investigated. Research on durability lags behind that on the safety and applicability of reinforced concrete structures.

In the present work, the porosity and mean pore diameter of concrete are determined by mercury intrusion porosimetry (MIP), and the effects of limestone content, specific surface area of the powder and water-to-binder ratio on the resulting concrete porosity and mean pore diameter are studied. The permeability of concrete with limestone addition is characterized by measuring the diffusion coefficient of chloride ions using the rapid chloride migration (RCM) method. The term permeability is often used interchangeably with ion diffusion coefficient hereinafter in this article. A fractal dimension is calculated to characterize the fractal structure of the pore network. The variation of concrete permeability with its porosity and fractal dimension is examined.

2. Experimental

2.1. Materials

P·O 42.5 (Chinese standard GB175–2007) ordinary Portland cement was used in the tests, the chemical composition of which is provided in Table 1. River sand with a fineness modulus of 2.8 was used as the fine aggregate and crushed stone, continuously graded from 5 to 20 mm, was used as the coarse aggregate. The specific surface area of the limestone powder, used as a mineral admixture, varied from 335 to

Table 1
Composition of cement.

Oxides	CaO	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	MgO	K ₂ O	SO ₃	LOI
wt%	61.89	22.95	6.35	4.10	1.02	0.75	1.58	1.36

Table 2
Composition of limestone.

Oxides	SiO ₂	Fe ₂ O ₃	CaO	MgO	SO ₃	K ₂ O	LOI
wt%	0.20	0.15	55.11	1.12	0.09	0.02	43.31

1028 m²/kg. The chemical composition of the limestone is provided in Table 2. The XRD analysis of the limestone is shown in Fig. 1.

2.2. Mix proportions of concrete

Limestone powders with three different specific surface areas, i.e. 335 m²/kg (Group A), 732 m²/kg (Group B) and 1028 m²/kg (Group C), respectively, were used in the tests to replace a portion of the cement. The content of limestone powder varied from 30 wt% to 50 wt%. The water-to-binder ratios were 0.3, 0.4 and 0.5 respectively.

The mixing proportions and properties of the concrete samples are summarized in Table 3. In the sample code “AX-Y”, for example, A stands for Group A; X represents limestone powder content, Y represents water-to-binder ratio, and so forth for Groups B and C samples. Groups A, B and C represent limestone powder with a specific surface area of 335 m²/kg, 732 m²/kg and 1028 m²/kg, respectively.

2.3. Porosity and mean pore diameter

The porosity of the sample was determined using an AutoPore IV9500 automatic mercury intrusion porosimeter (Micromeritics, USA). The MIP method employed in this study is based on functional relation between the volume of mercury intruding into a porous medium and the applied pressure to overcome the mercury surface tension. Since microcracks could be generated when hammering the test piece during sampling and affect the measurement results, a small sample of the concrete paste was poured into a separate container. The sample was conditioned together with the test specimen following the same curing procedure. The sample were cured for 28 days before they were moved into a container, where hydration was terminated using anhydrous ethanol. The sample was then dried in an oven for 4–5 h at 90 °C and cooled down to room temperature.

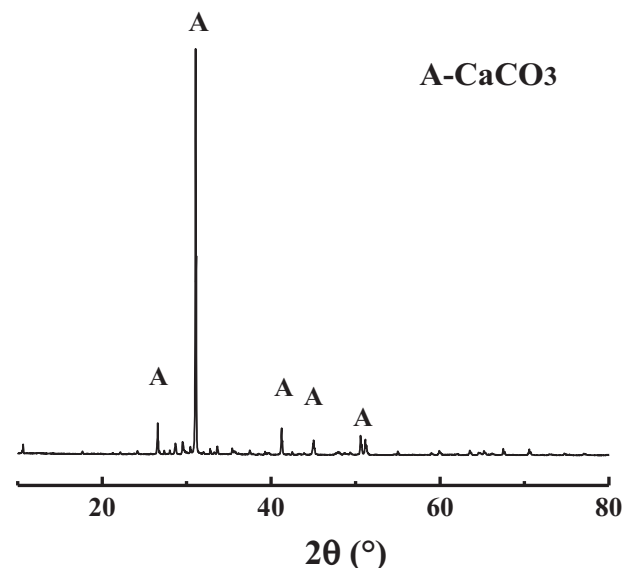


Fig. 1. XRD analysis of limestone.

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