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Modelling of irregular-shaped cement particles and microstructural development of Portland cement

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HIGHLIGHTS

- Irregular-shaped cement particles are reconstructed using a discrete-based method.
- Irregular-shaped cement particles can well represent real ones in terms of geometry and packing.
- Cement hydration model is built based on the reconstructed irregular-shaped cement particles.
- The effect of cement particle shapes on hydration process is significant in the early period.

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ABSTRACT

The shape of cement powder particles plays a crucial role in particle packing and hydration process of cement. However, cement powder is generally regarded as sphere for simplification in most cement hydration models. This paper aims to investigate the influence of cement particle shapes on hydration of Portland cement and microstructure of cement paste. A novel central growth model along with a particle packing algorithm is developed to generate cement particles with irregular shapes and reconstruct initial 3D microstructures of Portland cement. Afterwards, the hydration process of cement with different shapes are simulated using CEMHYD3D model. The results indicate that the generated irregular-shaped particles precisely reproduce the real cement particles in terms of surface area, particle size distribution and geometry. The effect of particle shapes on hydration is significant due to the difference in surface area and geometric discrepancy, whereas this effect becomes less obvious with decreasing water-to-cement ratio.

1. Introduction

Many natural and man-made materials composed of particles typically have random irregular shapes rather than an inherent shape, which leads to an increasing number of interests in the investigation of the influence of particle shapes on microscopic and macroscopic properties of granular media [1]. With regard to cement-based materials, the shapes of cement powder particles and aggregates have been shown to play a critical role in their workability, mechanical performance and durability [2–5]. Cement powders sharing similar clinker, gypsum and fineness have different water demands, setting behaviour and viscosity as different grinding techniques are used [2]. These discrepancies are attributed to the different particle shapes of cement. Therefore, an accurate characterisation and reconstruction of real irregular particle shapes is a crucial aspect in better understanding of hydration process, microstructural evolution and mechanical properties development of cement-based materials.

In recent years, different advanced techniques including 3D laser ranging [6,7] and X-ray computed tomography (XCT) [8] have been used to characterise and analyse the 3D shapes of coarse aggregates in concrete. Unlike coarse aggregates, the size of cement powder particles ranges from a few tenths of a micrometer to dozens of micrometers. It is still quite challenging to accurately capture the individual cement particle shapes in 3D using these techniques due to their limited resolution as well as the complicated and prohibitive sample preparation process. For example, there should be at least 10 voxels in any one dimension of a particle in order to obtain this particle's shape that can only be achieved using the best XCT synchrotron sources [9]. Computer simulation may provide a suitable methodology to reconstruct and analyse the 3D shapes of cement powder particles without above-mentioned drawbacks. Meanwhile, the only

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way to overcome the conundrum of investigating the influence of specific cement particle shapes on hydration process and microstructural evolution of cement pastes is numerical simulation to date.

Over the past few decades, several cement hydration models validated by experiments have been proposed, such as HYMOSTRUC3D [10], μic [11], DuCOM [12] and CEMHYD3D [13,14], which gives the possibility to investigate the relationship between cement particle shapes and cement hydration process in a quantitative manner. These models can not only capture the cement hydration process including the evolution of each phase (e.g., hydration products, anhydrous cement and capillary pore) but also the long-term properties including mechanical strength and transport properties. Nevertheless, in almost all these hydration models, the cement particles are simply assumed spherical in shape, which is far from the real situation that cement particle shapes are irregular in general. As such, the accuracy of simulated results using these models would be significantly affected. Therefore, it is essential to take irregular particle shapes into consideration in order to provide more accurate simulation of hydration process and microstructural development of cement-based materials. Although an increasing number of studies have been undertaken to explore the effect of irregular shapes of aggregate on properties of cement-based materials, unfortunately, to the best of authors' knowledge, there are only very few studies related to the influence of cement particle shapes on cement hydration. Researchers from National Institute of Standard and Technology (NIST) are the pioneers to capture the 3D shapes of cement particles of a standard reference cement, CCRL-133, using XCT techniques and reconstruct 3D cement particles with irregular shapes by spherical harmonic function for replacing the spherical cement powders in CEMHYD3D [2,9,15]. The spherical harmonic function method could successfully reproduce the real cement particles, while it is only suitable for medium and large cement particles due to the limitation of resolution of XCT images, i.e., 0.95 μm/voxel. Moreover, the reconstructed particle library is only available for a specific reference cement due to the complex, time-consuming and expensive experiments. Thus, further studies are necessary to reconstruct and analyse real irregular shapes of cement particles and complement our knowledge in the field.

The main purpose of this paper is to reconstruct the irregular-shaped particles and investigate the influence of shapes of cement particles on Portland cement hydration process. A novel central growth model (CGM) based on the discrete method is proposed to rebuild cement particles with random irregular shapes. The theory of CGM and relevant procedures of particle packing for generating 3D representative volume element (RVE) of irregular-shaped cement particles are described in detail. The obtained initial microstructure of cement is subsequently incorporated into the CEMHYD3D model to simulate the hydration and setting of irregular-shaped cement. A series of experiments are carried out to acquire the input parameters for simulations and measure some properties associated with hydration of a Chinese Portland cement (similar to ASTM Type I Portland cement) for validation of simulations including hydration heat, degree of hydration and setting behaviour. Based on the input parameters obtained from experiments and the reconstructed RVE accounting for irregular-shaped particles, parametric analysis and sensitivity analysis are undertaken to estimate the effect of particle shape on particle packing such as surface area, particle size distribution (PSD) and geometry, and cement hydration process including hydration heat, degree of hydration and connectivity of solid phases.

2. Initial microstructure model of irregular-shaped cement

2.1. General

In order to introduce irregular-shaped cement particles into cement hydration model, it is necessary to produce 3D irregular shapes of cement powder particles and then carry out random packing of these particles in a RVE. Herein, a central growth model based on the discrete method and cellular automata theory is proposed and developed to achieve these purposes.

The discrete-based method is an important approach for reconstructing objects in imaging techniques. 2D and 3D images are pixel and voxel based respectively. No matter how complex the single object is in shape, its shape can be represented using discrete basic elements. In turn, the quality of pixels or voxels can affect the similarity of the reconstructed shape and the shape of real object. Although the discrete-based method has a noticeable resolution-dependent limitation, as compared to the vector-based method for generating irregular-shaped particles, it can easily determine the geometric attributes and obtain an effective random packing process. For example, it is difficult to estimate the surface area of a vector-based random-shaped particle using traditional algorithms [16]. Regarding the packing algorithm of irregular-shaped particles, the mathematical decision procedure of overlapping between particles using vector-based method is too complicated and time-consuming [17,18]. Furthermore, the discrete-based cement hydration model has been successfully employed to simulate the hydration process and microstructural development of cement-based materials [14]. In this work, the procedure used to simulate cement hydration process follows the similar rules that have been applied to the existing discrete-based hydration model, e.g., CEMHYD3D. Thus, the discrete-based method for producing cement particles with irregular shapes can directly meet the rules of the cement hydration model without digitizing the reconstructed particles again, if the spatial resolution of particles is identical to that of RVE. In addition, the obtained virtual microstructure of cement-based materials from the vector-based cement hydration models, e.g., HYMOSTRUC3D and μic, also needs to be digitized or meshed prior to its incorporation into other numerical models for determining physical and mechanical properties of cement-based materials [11,19,20]. Therefore, the discrete-based method is chosen and used to produce irregular-shaped cement particles in this work because of its attractive advantages.

Cellular automation is a discrete method that has been widely used in microstructure modelling, mathematics, physics, complexity science and other areas [21]. Cellular automation consists of three basic elements, i.e., cells, evolution rules and relationship between adjacent cells. Cellular automata algorithm has been successfully implemented in the CEMHYD3D cement hydration model [13,14]. It was found that the voxel size and different relationship between neighbouring voxels have strong influences on the simulated pore structure, solid percolation and hydration rate of cement paste using CEMHYD3D [22]. As shown in Fig. 1, there exists three relationships between neighbouring voxels of the target voxel in 3D that are referred to as face-to-face, edge-to-edge and point-to-point neighbour models, respectively. In face-to-face neighbour model, the voxels are identified as neighbours of the target voxel if they have sharing faces with the target voxel. Similarly, the neighbours are defined as the voxels with sharing faces and edges in edge-to-edge neighbour model, and sharing faces, edges and points in point-to-point neighbour model, respectively. For face-to-face, edge-to-edge and point-to-point neighbour models, the number of neighbours is 6, 18 and 26, respectively.