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Water depercolation of setting cement paste evaluated by diffuse ultrasound

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ABSTRACT

Microstructural evolution during setting of cement-based materials explains its solidification. While various studies have attempted to reveal the microstructural evolution, many questions still remain. The setting process refers to the phase change from a cement suspension to a poroelastic solid. This study proposes a diffuse ultrasound method to investigate microstructural evolution and to determine the setting of cement paste. The diffuse ultrasound refers the propagation of an incoherent component on P-waves through-transmission. Its velocity and attenuation reflect the tortuous paths in early-age cement paste. Closing the tortuous paths by cement hydration and coagulation causes the diffuse ultrasound to combine with the propagated P-wave. The combined time is then an indicator of the setting point. The diffusivity, dissipation, and permeability evaluated by the diffuse ultrasound also shed light on the microstructural evolution of fresh cement paste.

1. Introduction

The fresh state of cement-based material determines the strength and durability of ts hardened state. After a cement-based material is at rest, is microstructural evolution starts with the first step: Setting. The initial setting time represents the condition where the cement-based mixture cannot undergo further mixing or handling. The final setting time is then the condition where the mixture is no longer workable and the beginning of mechanical strength. Even though the time of setting is conventionally measured by a Vicat needle test [1], the result hardly provides information to reveal the microstructural evolution. Various studies have also evaluated the evolution of mechanical properties of early-age cement-based material. The evolution is the result of cement hydration, a chemical phenomenon involving the binding of water in minerals. Measurement of isothermal calorimetry can be used to monitor the progress of cement hydration. However, the technique is reportedly inappropriate to identify the setting process because their results are hardly sensitive to the effect of the water-to-cement ratio on the setting process [2]. The setting was discussed on the basis of the measurement of Vicat needle penetration. The setting process is also evaluated by chemical shrinkage and the development of yield stress [3]. The setting process is affected by the water-to-cement weight ratio (*w/cm*), dispersed particle size, temperature, and environmental condition [3,4]. The influence of cement fineness of early-age properties of cement-based materials [5] and fluid-to-solid transition of setting in cement paste based on comparison between experimental and numerical technique [6] were investigated, and recently the influence of cement replaced limestone powder on hydration with developed mechanical properties [7], rheological properties [8], and its measurement method [9], also have been reported.

Advanced techniques of cement-based materials during setting and hardening were reviewed by RILEM TC 185-ATC [10]. The hardening of concrete was usually monitored using ultrasonic methods such as wave through-transmission [10–12] and reflection [13–16]. Robeyst et al. [17,18] reported the correlation between the P-wave velocity (P-wave modulus) and the setting time by the use of an ultrasonic through-transmission. The research was practically the expanded work done by Sayers and Grenfell [19]. Previous studies attempts to correlate between setting time and the P-wave velocity based on the propagating velocity of P-wave is

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determined by P-wave elastic modulus of matrix [4,20]. However, setting of cement paste can be defined a percolation of solid network to have a mechanical strength by hardening. Therefore, change of P-wave modulus can correlate with setting time but it hardly represents a setting process than diffuse ultrasound proposed by this study.

On the other hand, the methods using ultrasonic wave exploit the coherent response such as phase delay and amplitude decrease. The coherent response has been widely used to evaluate the material properties or the degree of damage based on the measurement of wave velocity and attenuation [21–24]. In contrast, diffuse ultrasound caused by wave scattering propagates incoherently in a heterogeneous material. The measurement of the diffuse ultrasound was reported in metallic materials [25–27] and in a solid beads-in-water suspension [28]. Diffuse ultrasound can follow a tortuous path in a medium. Changes in tortuosity have been used in soil structures to observe the tortuosity phenomena in pore spaces that influence the transport of water, solutes, and gases in soil [29]. Additionally, macropores in soils resulting from air and water transportation were recently measured by X-ray computed tomography. The 3D visualizations show a decrease in macroporosity of the soil sample [30]. In hardened cement-based materials, the method was applied to detect and evaluate damage [31–36]. The measurement evaluated the scattering parameters: Diffusivity and dissipation. Properties of aggregates affect the diffusivity in hardened concrete, which include their shape, distribution, and density. Viscoelastic properties of hardened cement paste govern the energy dissipation. A parameter indicating the arrival time of diffused-wave energy was also used to demonstrate the energy concentration of diffuse ultrasound [36].

This paper adopts a diffuse ultrasound method to characterize the setting process of early-age cement paste. Experimental measurement of the diffuse ultrasound on through-transmission P-waves was performed using a laser Doppler vibrometer. The measured arrival time of the diffuse ultrasound is expected to explain the percolating evolution of microstructure in early-age cement paste. The setting point determined by the diffuse ultrasound was compared with that by the conventional ultrasonic pulse velocity technique, where both P- and S-waves were used. The setting point by the diffuse ultrasound is slightly later than the birth time of S-wave propagation related to the final setting time by a Vicat needle test. Finally, the diffusivity, dissipation, and permeability of the diffuse ultrasound were also evaluated, and their variation shows the microstructural evolution of fresh cement paste.

2. Experiment

2.1. Sample preparation

Cement paste samples were prepared to apply the diffuse ultrasound technique. Type I Portland cement was used, and its specific gravity was 3.15. Oxide composition is 62.9% CaO, 21.1% SiO_2, 4.8% Al_2O_3, 3.5% Fe_2O_3, 2.6% MgO, 2.8% SO_3, 1.2% K_2O, 0.3% TiO_2, and 0.2% Na_2O. The diffuse ultrasound was measured for samples having water-to-cement ratios of 0.35, 0.40 and 0.50 by weight. Mixing time was 10 min. Relative humidity and temperature during the experiment were maintained at 55% and 20 °C, respectively.

2.2. Vicat needle test for setting time measurement

ASTM C191-08 proposed Vicat needle test for setting time measurement of cement paste based on the measurement of penetrated depth of needle [1]. The stress resistance to the axial force (about 2.94 N) by a penetrated needle (weight 300 g) is recorded as the penetrated depth. The penetrated depth will decrease according to the evolution of cement hydration. The time of each initial or final setting is defined when the penetration depth is 25 mm or less than 0.5 mm, respectively. In this study, the results are averaged values of two tests as shown in Fig. 1. As a result, the initial and final setting for $w/cm = 0.40$ are about 6 h and 7 h, respectively.

2.3. Measurement of phase velocity

The phase velocities for P- and S-waves were measured in early-age cement paste using the conventional ultrasonic through-transmission method. The phase velocities converted into the evolution of P-wave and shear modulus, respectively. The experimental setup is schematically described in Fig. 2, where a sample mold (300 mm × 200 mm × 20 mm) for mounting transducers were devised following that proposed by Reinhardt and Grosse [11]. A sample was placed in the mold. A sinusoidal tone-burst signal was then generated by a function generator (NI PXI-5142; National Instruments Corp.) with frequency from 150 kHz to 350 kHz with a sampling rate of 100 MS/s. A power amplifier (BA4825; NF Corp.) amplifies the generated signal. Contact-type transducers were used for excitation of P-waves (V192; Panametrics Inc.) and S-waves (V150; Panametrics Inc.), which were directly attached to the sample through a hole at the center of the mold. The measured voltage signals on the opposite transducer were amplified with a pre-amplifier (SR560; Stanford Research Systems, Inc.). The signals were then digitized with an analog-to-digital converter (NI PXI-5105; National Instruments Corp.) with a sampling rate of 60 MS/s. The contact-type transducers were reattached to the sample using a viscous couplant for each measurement due to drying of the sample surface for 3 h. With an average of 10 measured output signals to increase a signal-to-noise ratio, a rectangular window was used at the center of the tone-burst signal to compute the phase and its delay.

The generating and propagating of wave can be determined by used equipment and experimental setup. For example, low input voltage hardly generates and propagates wave through early-age cement paste. This study prepared an optimized experimental setup to measure wave propagation of cement paste during hardening. An example of computed complex phase of propagated S-waves is depicted in Fig. 3, which were measured 5 h and 10 h after mixing. The water-to-cement ratio of the sample was 0.40. It is difficult to precisely observe the phase delay of S-wave in early

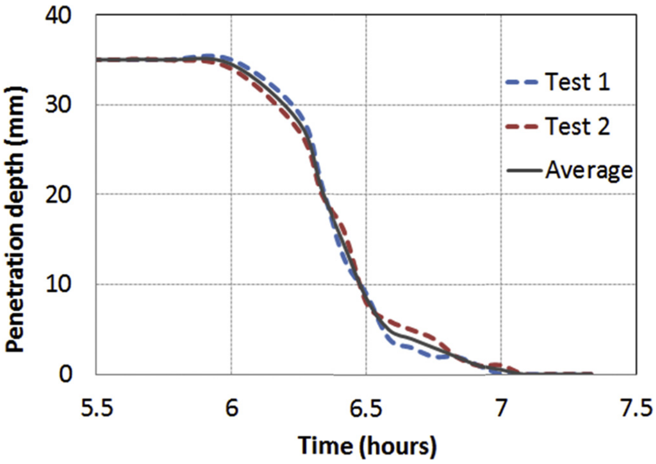

Fig. 1. Results of Vicat needle test.