Wave mechanism of structure formation in cement compositions

N P Gorlenko¹, Yu S Sarkisov¹, V I Syryamkin², L B Naumova², A N Pavlova¹ and B I Laptev³

¹Tomsk State University of Architecture and Building, 2, Solyanaya Sq., Tomsk, 634003, Russia
²National Research Tomsk State University, 36, Lenin Ave., Tomsk, 634050, Russia
³Nove Technologije, D. O. O. Ljbljana 1000, Sloveniya

E-mail: gorlen52@mail.ru

Abstract. The development of new methods of physicochemical analysis opens vast opportunities for studying the kinetics and mechanisms of the structure formation in cement systems. A study of the structure formation and cement hardening processes will allow widening a range of methods of their non-destructive testing and developing production techniques for advanced composite materials. The X-ray microtomography system is used to analyze the formation of alternating dark and bright concentric rings both on the surface and in the bulk of the specimens. It is supposed that the formation of concentric rings is caused by the generation of acoustic vibrations in the cement-water system at early stages of the structure formation.

1. Introduction
Today, digital technologies are applied in investigations of physicochemical and service properties of construction materials allowing users to estimate their non-uniformity, detect structural defects, pore size and pore distribution in the bulk of the material [1-4]. As is known, the inner structure of the cement-water system depends on many factors and is determined mostly by the formation of the capillary-porous, three-phase system solid phase-water-entrained air. The volume of unreacted cement particles, gel and capillary pores significantly changes during its curing. Control for their development and, as a consequence, the properties of construction materials can be performed only using new experimental data and detailed analysis of this process development. This will provide the digital technology-based online research in the field cement compositions [5].

2. Materials and Methods
PC-500 cement bricks 2×2×2 cm in size were used in the experiment. The water-cement ratio was 0.34 for humid curing conditions.

The X-ray microtomography (XMT) system used in this work has the following specifications: 1–13 µm image segmentation; soft X-ray source with energy range from 20 to 160 kV; 0–250 mA anode current; < 5 µm focal spot size (at 4 W). Image reconstruction time is 10 min/cm³. The X-ray detector has a resolution of 4872×3248 pixels, with the pixel size of not over 7.4×7.4 µm. The time of three-dimensional image reconstruction is 10 min. The system allows studying objects at any angle with ±1 µm accuracy of location without destroying the original object.
3. Results and Discussion
Figures 1 and 2 present a chain of the XMT images of the cement brick morphology with the increasing scanning depth.

![XMT scans of cement brick layers (a, b, c, d, e, f, g, h) at a 2 µm scan depth. Magnification: ×5.](image)

The XMT scans reveal the detailed morphology of the cement specimen, which contains dark formations (pores) and alternating dark and bright concentric rings. The following conclusions can be made after the analysis of the XMT images.

1. The pore distribution over the area and the bulk of specimens is random, and the total areal porosity at different depth is not a constant value.
2. The pore size ranges mostly from $10^{-2}$ to $10^{-3}$ µm at a depth varying from several units to hundreds of micrometers.
3. Entrapped air is both of symmetrical and asymmetrical configurations, which may be caused by the alternating density gradients at the cement-air interface at the end of the cement setting time.

4. The configuration of the observed alternating dark and bright concentric rings changes with the scanning depth. At the same time, the distance between two minima of the holes (dark rings) or between two maxima of the shoulders (bright rings) is constant and amounts on average to 0.006 m.

![XMT scans of cement brick layers](image)

**Figure 2.** XMT scans of cement brick layers (a, b, c, d, e, f) at a 4 µm scan step. Magnification: ×5.

The formation of these concentric rings is probably caused by the process development of the electromagnetic wave generation in the bulk of the hardening cement-water system. It is well known that acoustic vibrations accompany the processes of cement hydration and curing with the frequency ranging from 100 Hz to 20 kHz and intensity of 3–10 dB observed at the dispersion of cement particles [6]. It is possible to assume that the higher frequency vibrations, including ultrasonic, occur in the cement-water system especially at a time of cement mixing. Probably, ultrasonic vibrations is one of driving forces of the cement particle dispersion, whereas the concentric rings result from the formation of standing waves in the cement-water system (see Figure 3).

![Standing wave formation](image)

**Figure 3.** Schematic of standing wave formation: B – wave nodes; A – vibrational amplitude; \( \lambda \) – wavelength.
It is known that standing waves appear when two waves with the similar amplitude and length travel in opposite directions. This can occur if the electromagnetic vibrations are generated by particles or their new formations constituting the bulk of particles with similar size and properties. In other words, the system represents the totality of particles with more or less evenly distributed parameters, such as, for example, weight, elasticity, size close to that respective parameters in each elementary volume of the system. It is also known that due to the ultrasonic waves, the smaller particles and the liquid phase shift toward the standing wave node, whereas coarse particles shift toward the larger amplitude of vibration. As a result, the cement paste compacts nearby the contacting area of the electromagnetic source (dark rings). Outside this source, the formation of the lower density area is observed (bright rings). At the same time, the wavelength for a standing wave is determined by the double distance between the dark and bright rings and amounts on average to 0.012 m as presented in Figure 2.

The vibration frequency of ultrasonic waves in the cement-water system can be found from

$$f = \frac{c}{\lambda},$$

where $c$ is the velocity of the ultrasound propagation in the curing system; $\lambda$ is the wavelength.

Given that at the initial point of curing the system is fluid, the ultrasound propagation velocity can be conventionally set to 1490 m/s, i.e. the propagation velocity in water. In the course of formation of the cement brick the ultrasound propagation velocity increases and ranges between 1930 and 3000 m/s.

Let us calculate the frequency of standing waves according the given above equation:

$$f = \frac{(1490 – 3000)/0.012 = (124.2 – 250.0)}{0.012} \text{kHz}.$$.

Actually, as follows from Figure 1, these standing waves are characterized by a range of frequencies which include multiple harmonics due to which a wider frequency band can be expected, namely from acoustic to megahertz range observed in experiments.

In our view, the electromagnetic source is the eigen-frequency vibrations of cement particles which can be calculated as

$$f = \frac{1}{2\pi} \sqrt{\frac{F}{l}} m,$$

where $f$ is the eigen-frequency; $k = F/l$ is the proportionality constant representing the elastic force $F$ acting on the bond length unit $l$; $m$ is the particle mass.

At the initial point of curing, the cement paste is a colloidal system with the particle size ranging between $10^6$ and $10^8$ m. Assuming that the density of the cement paste is 1400 kg/m³, let us find the mass of the individual particles:

$$m = V\rho = (10^6 – 10^8)^3 \times 1400 \approx (10^{13} – 10^{21}) \text{ kg}.$$

For particle which manifest the dipole-dipole interaction, $k = (0.9 – 1.8) \times 10^7$ kg/m [7]. Therefore,

$$f = 0.16 \sqrt{\frac{(0.9 – 1.8) \times 10^{-7}}{(10^{-13} – 10^{-21})}} = (2 \times 10^7 – 1.5 \times 10^8) \text{ Hz}.$$

Accordingly, if the system contains a population of particles with the similar size and mass, the formation of standing waves having the acoustic and ultrasonic frequency bands is possible during the generation of free electromagnetic vibrations. The energy conversion from kinetic to potential and vice versa occurs within the section of the oscillating system (Figure 3), namely from the amplitude to the neighboring wave node. In this case, there are no energy losses because each of such section does not exchange energy with neighboring sections that is characteristic to standing waves. The absence of energy losses can lead to the intensification of the dispersion, hydration and structure formation processes in the cement paste. Hence, preconditions for maintaining standing waves in the system can additionally enhance the operating parameters of cement compositions.
Circumstantial evidence that ultrasound exerts an effect on the intensification of the dispersion, hydration and structure formation processes, is the literature data [8, 9]. One can suppose that along, for example, with a thermal effect, the generation of the internal electromagnetic waves in the cement-water system at the initial point of curing is the additional factor that affects the curing process.

It is important to note that the formation of concentric rings can be also observed at a travelling spherical wave. This is conditioned by different propagation rates of transverse and longitudinal waves. The ratio between these waves is determined by the Poisson ratio:

\[
\frac{V_1}{V_2} = \sqrt{\frac{1-2v}{2-2v}}
\]

where \(V_1\), \(V_2\) are the propagation velocities of the longitudinal and transverse waves, respectively; \(v\) is the Poisson ratio.

Given that \(v = 0.2\) for concrete, we get:

\[
\frac{V_1}{V_2} = \sqrt{\frac{1-2\times0.2}{2-2\times0.2}} = 0.375.
\]

Taking all these calculations into account, the curing cement-water system can be considered as a medium with the electromagnetic field impedance, where \(V_1 < V_2\), that results in the formation of visible concentric rings.

4. Conclusions

The X-ray microtomography-based analysis showed that the alternating dark and bright concentric rings both on the surface and in the bulk of the specimens were formed at a distance of about 0.006 m between them.

Based on calculations, it was assumed that one of the possible mechanisms of the observed phenomenon could be the generation of internal electromagnetic waves having the acoustic and ultrasonic frequency bands. The obtained experimental data and theoretical calculations indicated to a possible wave mechanism of the structure formation in cement compositions.

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