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# Evaluation of water transfer from saturated lightweight aggregate to cement paste matrix by neutron radiography

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ABSTRACT

In high-strength concrete with low water–cement ratio, self-desiccation occurs due to cement hydration and causes shrinkage and an increased risk of cracking. While high-strength concrete has a denser matrix than normal-strength concrete, resulting in lower permeability, early-age cracks would cancel out this advantage. For the mitigation of this self-desiccation and resultant shrinkage, water-saturated porous aggregate, such as artificial lightweight aggregate, may be used in high-strength concrete. In this contribution, for the purpose of clarification of the volume change of high-strength concrete containing water-saturated lightweight aggregate, water transfer from the lightweight aggregate to cement paste matrix is visualized by neutron radiography. As a result, it is clear that water was supplied to the cement paste matrix in the range 3–8 mm from the surface of the aggregate, and the osmotic forces may yield water transfer around lightweight aggregate in a few hours after mixing.



## 1. Introduction

Cracking of reinforced concrete member may jeopardize its durability performance, and this has recently become a well-known social problem for public infrastructures or private buildings. Responding to the social atmosphere and looking for additional benefits, using high-strength concrete for reinforced concrete structures has nowadays become popular. Basically, high-strength concrete is attained by mixing less water and more cement in the concrete mixture, and in consequence, high-strength concrete is denser than normal-strength concrete. This implies that high-strength concrete has a high potential of durability because of its lower permeability to substances that induce rebar corrosion, when it is used for reinforced concrete members.

An increased risk of cracking of high-strength concrete, however, is induced by self-desiccation due to cement hydration itself. As cement hydration progresses, the available water decreases, and inner concrete environment yields lower chemical potential of water, especially in the case of low water to cement ratio. As a result, driving forces for shrinkage of concrete are produced during the period in which concrete develops its strength.

Using water-saturated lightweight aggregate (LWA) in high-strength concrete is one of the countermeasures against early-age cracking [1–7]. The water trapped in porous aggregates behaves as a reservoir in the concrete, and the LWAs supply water to the cement paste matrix when the self-desiccation occurs. Additionally, this supplied water not only compensates the shrinkage of concrete, but also causes expansion of the concrete in the first days after casting. This expansion mechanism is not cleared and this deformation of concrete cannot be estimated quantitatively (see Appendix and Fig. A1).

For this purpose, several attempts for measuring water transfer from LWA to cement paste have been made. A method involving saturating a LWA with an ink solution was used in Ref. [8]. With this indirect method, it was concluded that the water penetration depth from LWA surface was at least 1 mm, while a previous assumption is a few hundreds of micron in a hardened cement paste [18,19]. Recently for more precise data, one-dimensional X-ray absorption [9], three-dimensional X-ray micro-tomography [10] and magnetic resonance imaging [11] were applied. The detected distance of water supplied from the aggregate surface ranged in these studies from 4 to 10 mm.

Neutron radiography seems to have the potential to clarify the mechanism of water supply from LWA during cement hydration, because neutron is particularly sensitive to $H_2O$, which has large neutron cross-section and this makes it possible to illustrate the water distribution in the specimen.

Application of neutron radiography to cement-based material goes back to the paper of 1972 by Rijonen and Pihlajavaara [12], whose study aimed at non-destructive monitoring of carbonation in concrete. Recently, as digitalization of graphics is promoted due to the development of charge coupled device (CCD) cameras, several reports have been published [13–15].

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In this contribution, neutron radiography was applied to visualize the water supply from LWA to cement paste matrix. A precise quantification of water transfer from LWA is a prerequisite for developing quantitative control methods for avoiding cracking of high-strength concrete.

## 2. Methods and materials

The facility used for this experiment was the Thermal Neutron Radiography Facility (TNRF) installed at the research reactor, JRR-3M, of the Japan Atomic Energy Agency (JAEA). The neutron flux was $1.2\times10^{8}\,n/cm^{2}\,s$. The TNRF consists of a fluorescent converter ($^{6}LiF/ZnS$: Ag), two quartz mirrors, one lens (Nikon Micro-Nikkor 105 mm) and one cooled charge coupled device (C-CCD) camera (C4880: Hamamatsu photonics), as shown in Fig. 1. A full transfer type C-CCD camera with an effective array of 1008 pixels $\times$ 1024 pixels of 100 mm $\times$ 100 mm each was used. The spatial resolution is approximately 100 μm/pixel. Neutrons falling on the converter are transformed into visible light in proportion to the flux and guided to the C-CCD camera using two quartz glass mirrors. The brightness of the image is digitized by the image processor.

It takes approximately 8.0 s to get one image, including data transfer time, and the specimen is exposed to the neutron flux for 1.2 s. Regarding the image taken by this TNRF, white spots that are pixels hit by neutrons and gamma-rays directly in a captured image are produced inevitably due to highly sensitivity of the CCD camera. Therefore, for the correction of these white spots, a n intelligent filter was applied. Additionally, each figure was corrected with reference to two background images, i.e. a background image when neutrons were not irradiated and an image without the sample. For each image, a dark current image subtraction and a shading correction were performed. More details of this set-up and image-related procedures are described in Refs. [16,17].

From the statistical analysis of the detected intensity of the neutron flux, the relationship between the intensity of the neutron beam and the water-related (free water and chemically bound water) characteristics of the sample can be described as follows:

$$I_{t,0} = I_0 e^{-(\Sigma_c\delta_c+\Sigma_{w0}\delta_{w0})} \quad (1)$$

where $I_{t,0}$, $I_0$, $\Sigma_c$, $\delta_c$, $\Sigma_{w0}$ and $\delta_{w0}$ denote the intensity of the neutron flux passing through the sample, the incident intensity of the neutron flux, the macroscopic cross-section of water in the sample, the thickness of the sample, the macroscopic cross-section of water and the thickness of the sample, respectively. In addition during this experiment, the thickness of the sample may vary a little during the test due to expansion or shrinkage of concrete. This effect is not considered.

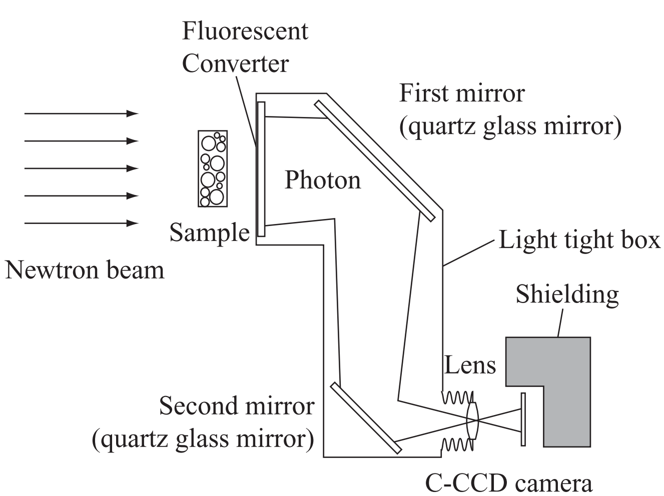


**Fig. 1.** Schematic of TNRF.

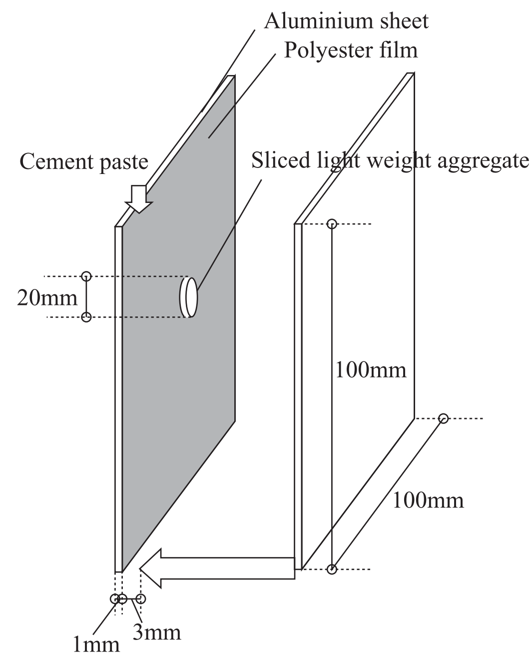


**Fig. 2.** Detail of specimen aluminum sheet is better.

The size of the concrete specimen composed of cement paste and sliced saturated LWA is 100 mm $\times$ 100 mm $\times$ 3 mm, as shown in Fig. 2. The used cement is ordinary Portland cement, and a polycarboxylic-acid-based water-reducing agent was added for attaining water to cement ratio of 0.23. The mold was made of an aluminum sheet, which has low neutron absorption, covered with a polyester film on which cement paste was contacted. After casting, all the surfaces of specimen were wrapped with aluminum adhesive tape to prevent evaporation. The artificial LWA used in this experiment is a kind of expanded shale, with surface-dry density of 1.65 g/cm$^3$ and adsorption ratio of 32% by mass. This adsorption ratio can be considered as the porosity of this aggregate.

## 3. Results and discussion

Neutron radiography images of the specimen were taken at 1, 4 and 21 h after mixing. Fig. 3 shows the experimental results of neutron radiography images. It was observed that the intensity distribution of the neutron flux changes as time progressed.

During this test, the specimen was completely sealed and the total amount of water in the system was constant, while the averaged intensity is slightly changed. This phenomenon could be explained by the reaction of cement and production of hydrates, which may change the space density of water in it. The difference of average intensity is summarized in Table 1.

In Fig. 4, the intensity distribution along line A shown in Fig. 3 is plotted. The intensity in the LWA is remarkably reduced as time progressed; from this evidence it can be concluded that the water in LWA was supplied to the surrounding cement paste.

To visualize the areas to which water was supplied at 4 and at 21 h, the points that have a positive intensity difference greater than 0.02 compared to the image at 1 h are plotted in Fig. 5. This mapping shows that the area to which water is supplied from 1 to