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Study of Homogeneity and Water Absorption Behavior of Double Layer Silver Gray Tiles Using Film Neutron Radiography Technique

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Abstract

Neutron radiography (NR) technique has been adopted to study homogeneity and water absorption behavior of building materials, like double layer silver gray tiles obtained from Concord Real Estate & Building Products, Unit II, Salna, Gazipur, Dhaka, Bangladesh. Measurements of optical density differences between the film background and radiographic images of the dry/wet samples were used for investigation of the present work. The optical density was measured by using the digital optical densitometer (Model 07-424, S-23285, Victoreen Inc. USA). Large variation in optical density values of the radiographic image was observed. From this observation it shows that the rate of water absorption of the tiles increases with increase of immersion time. Through the investigation of radiographic image and subsequently analyzing the optical density we observed that distribution of the elements in the tiles are inhomogeneous.

Key words: Homogeneity, Water absorption, Silver gray, Neturon radiography.

Introduction

A very well known method of non-destructive examination for characterizing the internal structure of an object is the use of penetrating radiation, such as X-ray in X -ray radiography or neutron in neutron radiography. These two radiographic processes are often complementary. X-rays are stopped by dense materials and pass through light ones. In many instances neutrons have reverse properties. Neutrons can penetrate the body of a large sized metal to give a good image of its internal structure. But for X-ray, it would require long exposure which would obliterate most of the details available by the

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radiography. Neutron radiography is a powerful non-destructive testing technique for internal evaluation of materials, such as its voids/ cavity, cracks, homogeneity etc. (Alam *et al.* 2006). It involves attenuation of a neutron beam by an object to be radiographed and thus to make the registration of the attenuation process (as an image) on a film or video. This is the most important advantage of the neutron radiography method over other methods available for doing this experiment. Clay is a widely available raw material that survives very well in its fired form. Many ancient cultures have made useful decorative items such as pottery, figurines, building tiles, and burial containers that become important parts of the archaeological record. The material aspects of clay and ceramic technology, the physical properties of clay and various construction and firing methods can be investigated using archaeometric techniques (Renfrew and Bahn, 1996, Rice 1987). Ancient technologists and archaeological material researchers have employed standard techniques such as X-ray radiography, X-ray diffraction (XRD), scanning electron microscopy (SEM), and neutron activation analysis (NAA) to study structure and composition of ceramic materials (Renfrew and Bahn, 1996, Rice 1987). Neutron radiography has been used to detect internal defects in some materials such as ceramics (Alam 2005), single layer Italian tiles (Alam *et al.* 2007) and different building industries (Islam *et al.* 2000). The technique is also adopted for the study of water absorption behavior in biopol, jute-reinforced-biopol composite (Alam *et al.* 2006) and wood plastic composites (Islam *et al.* 2003) etc. In this paper, we study homogeneity and water absorption behavior in the double layer silver gray tiles using film neutron radiography technique. These results are obtained through the investigation of digital images obtained by the high-resolution digital camera, measurement of optical density using digital optical densitometer (Model 07-424, S-23285, Victoreen Inc. USA).

Neutron radiography facility

The neutron radiography facility installed at the Institute of Nuclear Science and Technology, Atomic Energy Research Establishment, Savar, Dhaka is equipped to facilitate inspection of radioactive samples (mainly, fuel rods) as well as non-radioactive materials. The tangential beamport of the reactor of the institute allows for selection of thermal neutron beam. The areas of application for this facility range from integrity analysis in nuclear and non-nuclear samples via testing of adhesion or bonding of coatings for detection of pores, cracks, voids and homogeneity of elements present in the sample. The tangential beam port has been used for taking the neutron radiographs of the sample because the neutron beam coming out of the port contains less amount of gamma - rays (Rahman *et al*. 1989, Islam *et al.* 1995) compared to other beam ports. Moreover, a 15cm long bismuth filter has been used inside the port to cut existing

gamma-ray because gamma-ray produces unwanted fogginess in the radiography film. To control the beam of neutrons, a 120cm long conically shaped cylindrical divergent collimator, having inner and outer diameters 5 cm and 10 cm, respectively, has been installed inside the reactor biological shielding assembly. A beam stopper has also been installed in front of the beam port. It is a wooden box, which contains neutron-shielding materials like paraffin wax and boric acid. The NR facility includes a beam catcher having a hole in the middle of the front face. A lead block has been placed at the back of the hole for gamma shielding and the remaining part of the beam catcher has been filled with neutron shielding materials. In between the beam stopper and beam catcher there is a sample and NR camera/cassette holder table. Finally, the NR facility has been housed to reduce neutron and gamma background by using special concrete containing cement, heavy sand (mixture of ilmenite, magnetite and ordinary sand) and stone chips. Details of the NR facility can be found elsewhere (Rahman *et al.* 1989, Islam *et al.* 1995). Neutron radiography parameters at the tangential beamport of the NR facility have been calculated previously by Islam *et al.*1995.

Materials and Methods

Collection and preparation of the sample

Double layer Silver Gray tiles have been collected from Concord Ready-Mix Company Bangladesh Ltd., located at Salna, Gazipur, Dhaka. This has been used as the sample to study homogeneity (elemental distribution) and water absorption behavior of the sample. It is manufactured with the help of very experienced management team consisting of skilled engineers and architects by using raw materials like cement, sand, stone chips etc. The company also has a fully automated Italian press machine, which also manufactures single layer (monolayer) tiles. For final preparation, the tiles are polished in a 10-head liner grinding machine and then the sample was dried at daylight until to get the constant weight.

Converter foil and film loading in the NR-cassette

A thin converter (gadolinium metal foil of 25 μm thickness) was placed at the back of the X-ray industrial film. The loading of the X-ray industrial film (Agfa structurix D4DW) into the NR cassette (18 cm x 24 cm) is a simple procedure, (Bouma 1969) which requires only a little practice to be carried out easily in the darkroom. Loading should be done near the cupboard where the film is stored. The "dry" table should only be used for this work to prevent any contact of developing liquids with the unexposed loading film. There are a number of steps to place the industrial X-ray film into the NR cassette to protect the film against daylight and lamplight. The best way to load the cassette is to open the NR cassette when it lies on the

"dry" table. While using proper illumination only, a film still in the interleaving paper is carefully drawn out from the box holding the film between thumb and finger and putting the film in the cassette. It is very convenient and easy to load in the dark room.

Placing the sample and the NR-cassette

The sample is placed in close contact with the NR cassette and directly on the sample holder table. The NR cassette is placed on the cassette holder table. Both are placed in front of the neutron beam (diameter 30cm).

Exposure

Exposure means passing of neutron beam through a sample and holding it onto a special film (X-ray industrial film) in order to create a latent image of an object in the emulsion layers of that film. Of course, this discussion is confined only to direct contact radiography (film in close contact with the Gd converter foil) of the film. The sample was then irradiated for the optimum time, i.e., the time required to obtain good neutron radiographs. Exposure time differs for different samples, depending on the intensity of the neutron beam, density and thickness of the sample and neutron cross-section. The optimum exposure time of the sample was determined by taking a series of experiments with different exposure time, while the reactor was operated at 250 KW. For the present experiment we found the optimum exposure time to be 1 hour.

Obtaining radiographic images

In the NR-Cassette, gadolinium foil of 25μm thickness is used as converter and Agfa structurix D4DW industrial X-ray film is used as detector. The samples were then irradiated at an optimum exposure time of 1 hour at dry condition and also for different immersion time e.g., 4, 6, and 24 hours. Then the film was chemically processed in a Kodak D-19 developer (5 minutes) solution for developing, in a flowing tap water for washing (1 minute), then fixing in a Kodak unifix powder solution (5 minutes) and then again washing in a flowing tap water for final washing. After completing these steps, the radiographic images of the sample were obtained. These images were visualized on a PC using a high-resolution digital camera.

Mathematical formulation

The neutron intensity before reaching the object is different from the intensity of the neutron after passing through it. This is expressed through the equation (Norris *et al.* 1996)

$$I = I_0 e^{-\mu x} \quad (1)$$

where, e = base of natural logarithms, x = thickness of the object, m = linear neutron attenuation coefficient, I and I0 are the neutron intensity after passing through the object and the neutron intensity incident on the object, respectively.

The mathematical expression for the optical density (Harms *et al*. 1986) at a point of the film/image, D is given by:

$$D = \ln(A_0/A) \quad (2)$$

Here, A_0= response of densitometer without the image and A= response of densitometer with the image. The change in image density, DD at each point of the sample image with the film background, may then be calculated using the formula:

$$\Delta D = D_{bk} - D_{image} \quad (3)$$

D_{bk} = optical density of the film (background), D_{image} = optical density at a point of the sample image. Adopting equation (3), $(\Delta D)_{av}$ can be written as

$$(\Delta D)_{av} = D_{bk} - D_{(image)av} \quad (4)$$

$D_{(image)av}$ = average optical density of each level of image points on the horizontal line of the sample image. $(\Delta D)_{av}$ = Average value of optical density difference between film background and radiographic image of the sample. Using equation (4) for the density distribution, the fluctuation with respect to the central position of each sample image can be defined as

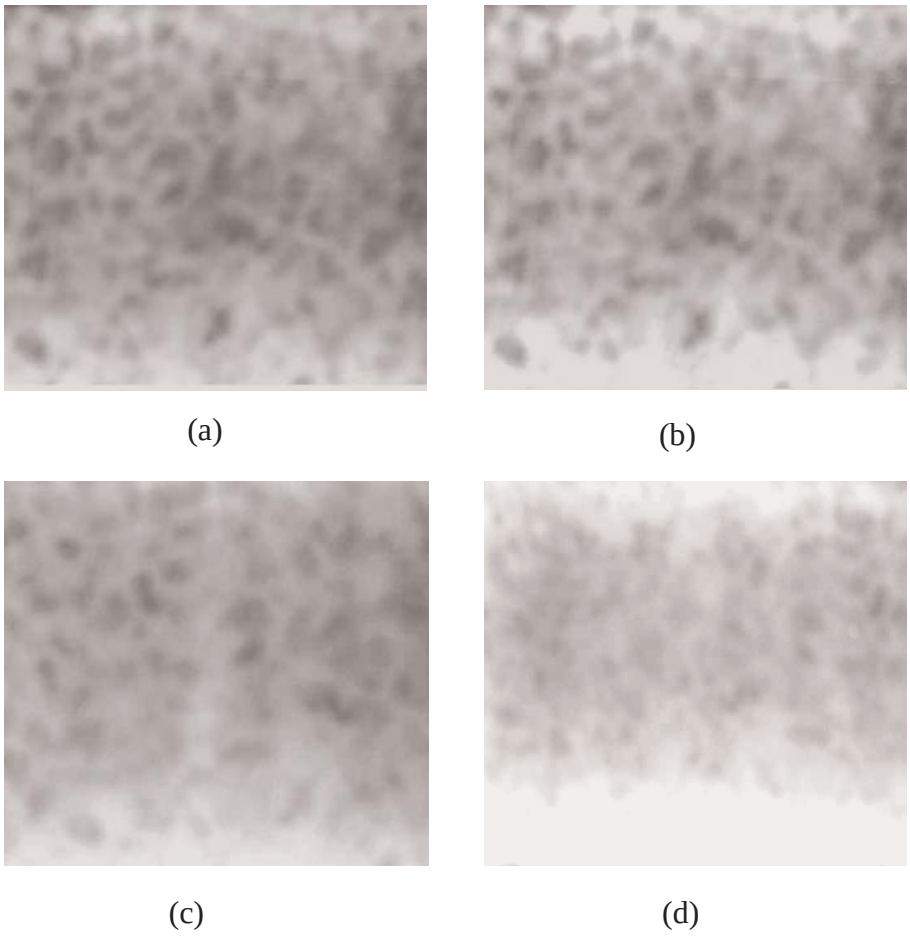
$$\% \text{ of } (\Delta D)_f = (D_{(image)av} - D_{cent})/ D_{cent} \text{ X } 100 \% \quad (5)$$

where D_{cent} is the optical density of the radiographic image at the center of the dry sample.

Results and Discussion

Homogeneity

In the work an effort has been made to investigate the homogeneity of the sample for which (ΔD)av measurements were carried out from the radiographic images by using the optical densitometer (Model 07-424, S-23285, Victoreen Inc. USA) (Islam *et al.* 2000). Fig. 1(a) shows the neutron radiographic image of the dry sample. Fig. 1(b) represents the radiographic image of the sample for 4 hours immersion time. Fig. 1(c) and 1(d) also shows the radiographic images of the sample for 6 hours and 24 hours immersion time respectively. A point related to this is worth mentioning here. The neutron beam falling on the sample is passed through a conically shaped divergent collimator before it appeared on the sample target. Hence we expect a larger neutron flux at the centre of the circular area of the target (Islam *et al.* 1995). The flux values should be decreasing continuously as we go off from the centre. Hence the levels 1 and 7, being at approximately equal distances from the centre, should have equal flux. The observed optical density $(\Delta D)_{av}$ values are respectively 1.40±0.17 and 1.45±0.12 for these two points and hence are in support of it. Levels 3 and 5 had the $(\Delta D)_{av}$ values 1.35±0.14, 1.36±0.14 respectively. Levels 2 and 6 form the similar pair for which we had the experimental results 1.36±0.14 and 1.44±0.06 respectively. Thus we can comment that



1. Neutron radiographic images of the sample at (a) dry condition (b) 4 hours immersion time (c) 6 hours immersion time and (d) 24 hours immersion time.

within the area of interest of the radiographic image (for dry sample) as the sample showed inhomogeneous elemental distribution in the sample. In this work, homogeneity means the uniformity in the distribution of the tiles materials. The best homogeneity would ensure the same optical density difference between the center and its surrounding positions on both sides of the radiographic image. Fig. 2 shows the optical density differences between film background and the image of the dry sample. Large variation of the optical density differences was observed on both sides from the center position of the sample image (Table I). From these investigations it can be concluded that raw materi

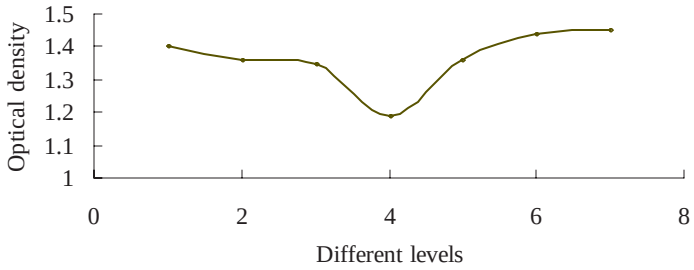


Fig. 2. Optical density differences at different levels of the film background and the radiographic image of the dry sample.

als of the sample are not in good mixing condition i.e., the sample is inhomogeneous mixing of tiles materials.

the 24-hour-duration data (0.29±0.11 and 0.29±0.08 respectively) are also changed except center position of the sample. The

Table I. Optical density difference (ΔD), and standard deviation (std.) between film background and the different levels of the sample (dry) image at an exposure time of 60 min.

Levels	ΔD							$(\Delta D)_{av}$	Std.
1	1.61	1.52	1.33	1.30	1.33	1.39	1.36	1.40	0.17
2	1.62	1.27	1.20	1.25	1.29	1.46	1.45	1.36	0.14
3	1.51	1.31	1.22	1.22	1.31	1.39	1.52	1.35	0.14
4 (center)	1.20	1.18	1.17	1.17	1.20	1.21	1.18	1.19	0.02
5	1.62	1.41	1.24	1.15	1.36	1.45	1.34	1.36	0.14
6	1.51	1.40	1.37	1.40	1.48	1.52	1.42	1.44	0.06
7	1.47	1.40	1.26	1.39	1.44	1.52	1.68	1.45	0.12

Water absorption characteristics

Table II shows the measured optical density values for 3 different immersion time, e.g., 4, 6 and 24 hours. Optical density values for 6-hour duration at location 1 is 0.23±0.05; and the corresponding result at the location 7 is 0.25±0.10. The 6-hour-duration data varies from the 4-hour-duration data (0.16±0.06 at location 1 and 0.02±0.01 at location 7), but later data are varies, indicating higher rate of water absorption at such a long duration after starting the experiment.

Obtained results for all the locations depict larger variation of optical density for the samples having 4, 6 and 24 hours of immersion time. Of the 24-hour water absorption time, at the center position of the sample the absorption of water is the same as the first 6

Table II. Average value of the optical density differences and standard deviation (std.) at different levels between the radiographic image of the dry and wet sample at different immersion time

Levels	Av. optical density differences and standard deviation (std.)					
	4h-dry		6h-dry		24h-dry	
	Density	Std.	Density	Std.	Density	Std.
1	0.16	0.06	0.23	0.05	0.29	0.11
2	0.09	0.08	0.21	0.09	0.30	0.08
3	0.04	0.03	0.17	0.04	0.27	0.05
4 (center)	0.12	0.02	0.33	0.33	0.33	0.02
5	0.04	0.04	0.19	0.11	0.22	0.10
6	0.01	0.01	0.12	0.09	0.25	0.14
7	0.02	0.01	0.25	0.10	0.29	0.08

hours. Towards the center position of the sample water absorption is different at all time duration but at 6 and 24 hours water absorption is same at the center position.

Fig. 3 shows the variation of the average values of the optical density differences for different levels when differences are measured between the dry and wet samples. These results show the larger optical density differences than that of the dry ones. On the basis of the observed average optical density values it is concluded that water absorption of this tiles is increased if it is exposed in water for a long time (24 hours). Tiles are becoming more and more important in industrial application and their quality control requires special testing methods. Very small cracks and voids are well inspected by X-ray microfocus computerized topography (CT) (Illerhaus 1988) whereas the homogeneity in larger objects is checked best with the neu

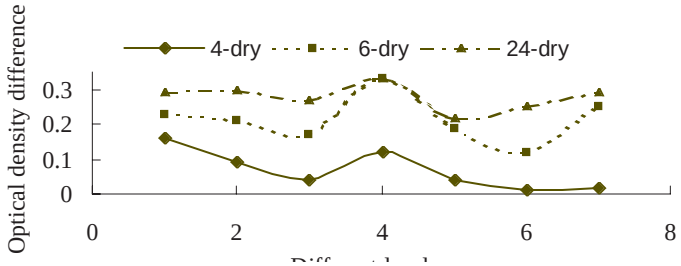


Fig.3. Average value of the optical density differences of different levels between the radiographic images of the dry and wet sample.

tron radiography technique. For a ceramic cylinder an inhomogeneous density distribution is found by some authors (Pfister *et al.* 1992) through their numerical analysis, the variation being 4 % only. But the present investigation shows average values of inhomogeneous density distribution, varying within 17.03%, with respect to the central position of the sample.

Conclusions

Neutron radiography technique can usefully be applied to study water absorption behavior and to detect the internal defects such as voids, cracks and inhomogeneity, etc. of the elemental distribution in the tiles. Additionally one may conclude that the elemental distribution is not homogeneous, and at every immersion time the rate of water absorption is increases with increasing time.

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