Anomalous Water Diffusion in Concrete Based on Neutron Backscattering Measurements

A. M. Abdel-Monem\textsuperscript{a}, A. El Abd\textsuperscript{a}, R.M. Megahid\textsuperscript{a} and I. I. Bashter\textsuperscript{b}

\textsuperscript{a}Reactor Physics Department, Nuclear Research Centre, Atomic Energy Authority, Cairo, Egypt.
\textsuperscript{b}Physics Department, Zagazig University, Zagazig, Egypt.

Received: 13/12/2009  Accepted: 18/1/2010

ABSTRACT

This work presents a new method based on neutron backscatter measurements to study isothermal water flow and distribution in concretes. Ordinary concrete samples with different percentages of silica fume are used to study the water profiles ($\theta - x$) which describe the water infiltration in the samples along the flow directions, $x$, for different absorbing times. The macroscopic theory of water infiltration in porous media was used to model the infiltration process.

The obtained results show that the diffusion process is anomalous; i.e., the water front position, $x_p$, does not follow the square root behavior of the absorption time, $\sqrt{t}$, and the water profiles ($\theta - x/\sqrt{t}$) do not collapse to the universal one master curves for all samples. It is shown that the diffusion process is a super, the power law, $x_p = \gamma_p t^{-0.85}$, successfully describes the advance of water front with time and the water profiles collapse to universal one master curves ($\theta - \gamma$). The water diffusivities were derived in an analytical form for the investigated samples. They have the hyper-diffusion behavior; i.e. diffusivities decrease as the water content increase. It is shown that the diffusivity for all water contents decreases with increasing the silica fume percentage up to 15%. However, for concrete with silica fume $\geq 20\%$ the observed phenomenon is reversed due to the deterioration of concrete physical and mechanical properties.

Key Words: Neutron backscattering / Water profiles / Silica fume.

INTRODUCTION

Studies on the transport and distribution of fluids in porous media (especially concrete which used for under water constructions) are an important issue. Since the deterioration of building materials are largely mediated by water intrusion, the durability of building structures is critically determined by the rate at which water and other contained deleterious chemical agents infiltrate through the porous structure. Accordingly, the problem of water movements in porous building materials has received great attention in the past decades and there is still a strong interest in finding methods and techniques that can be used to determine the macroscopic hydraulic properties of partially saturated porous media to estimate and eventually improve their durability\textsuperscript{(1, 2)}.

Non-destructive, methods based on nuclear techniques with neutron and gamma radiography\textsuperscript{(1, 3, 4, 5, 6, 7)} X-ray radiography\textsuperscript{(8)} nuclear magnetic resonance\textsuperscript{(9, 10, 11, 12)}; and neutron, gamma and X-rays attenuations\textsuperscript{(13, 14)} however, destructive ones such as gravimetric method\textsuperscript{(15)}; are still the most used to study water infiltration in porous media\textsuperscript{(16)}. The best candidate method for studying liquid flow and
distribution in porous media is neutron radiography, because neutrons have different interaction processes with high scattering cross sections for light nuclei such as hydrogen (17).

While neutron scattering from hydrogen in the neutron radiography studies of water infiltration in porous media, is a removal process and attenuating neutron passing through the object, it is unwanted neutron component that obscure obtained information (1). A lot of methods are used to avoid its effect on the obtained results (1, 18, 19). However, it is advantage and useful in the so-called neutron backscattering methods.

The neutron backscattering (NBS) technique is a well established method to find hydrogen in objects. It is used in monitoring water levels in large containers and detection of land mines (20, 21).

The NBS method for land mine detection is based on the fact that, explosive materials contain much more hydrogen atoms than the surrounding soil. Thus the problem to find the explosives is shifted to detect the hydrogen atoms. This is done easily, by irradiating the suspected area, with fast neutrons from any suitable neutron source. The propping neutrons are slowed down by multiple scattering (i.e. moderated). A neutron detector detects slow neutrons as they scattered from the irradiated area. An increase in the recorded counts by the detector is an evidence of the presence explosives in the irradiated area, since explosives contain hydrogen that increases thermal flux of the scattered neutrons (20, 21, 22).

In this work the idea of NBS method in land mine detection, is extended to study the water transportation and diffusion in concrete samples under investigation. The following topics were studied: (i) the isothermal water infiltration in ordinary concrete, (ii) the effect of silica fume addition on the infiltration process, and (iii) determining the water diffusivities by applying the macroscopic theory of water infiltration in porous media.

THEORETICAL TREATEMENT:

Moisture transport in unsaturated building materials is commonly described at the macroscopic scale by Fick’s law (Draey law) of diffusion. It can be written in one dimension, $x$ (23, 24) as

$$ q(\theta) = -D(\theta) \frac{\partial \theta}{\partial x}, $$

where;

$q(\theta)$ is the water flux which is a function of the water content $\theta$, and $D(\theta)$ is the water content dependent diffusivity - it is considered as one material property.

By combining equation (1) with the continuity equation, $\frac{\partial \theta}{\partial t} = -\frac{\partial q}{\partial x}$, ($t$ is the time), the moisture transport in unsaturated porous media (building materials) can be described by the non-linear partial differential diffusion (23, 24) given by:

$$ \frac{\partial \theta}{\partial t} = \frac{\partial}{\partial x} \left( D(\theta) \frac{\partial \theta}{\partial x} \right). $$
Equation (2) can be transformed to an ordinary differential equation using the Boltzmann variable, 
\[ \varphi = \frac{x}{\sqrt{t}} \] and the water diffusivity can be derived from this equation using the well-known boundary conditions and it is given by 
\[ D(\vartheta) = -\frac{1}{2} \frac{d \varphi}{d \vartheta} \int_{0}^{\vartheta} \varphi \, d \vartheta. \tag{3} \]

According to Fick’s hypothesis as well as the Boltzmann transformation, all points along the flow direction \( x \) at the same water content scale with \( \sqrt{t} \). This means that the \(( \vartheta - \varphi \) profiles should be a single master curve.

If \(( \vartheta - \varphi \) profiles do not collapse to one master curve, namely all points along the flow direction \( x \) at the same water content do not scale with \( \sqrt{t} \), equation (1) can not be used to describe moisture transport in unsaturated building materials and this is so-called anomalous diffusion, and an alternative approaches are needed. A number of approaches were used in literature. Kuntz et al. have used this equation \((1, 25)\).

\[ \frac{\partial \vartheta}{\partial t} = \frac{\partial}{\partial x} \left( D(\vartheta) \left( \frac{\partial \vartheta}{\partial x} \right)^{\alpha} \right), \tag{4} \]

Where; \( \alpha \) is a real number, \( \alpha = 1 \) corresponds to the Darcy or Fick’s law

Assuming that \( \vartheta \) depends on the single variable \( \gamma \) – the so-called generalized Boltzmann variable 
\[ \gamma = \frac{x}{t^{n}}, \tag{6} \]

(this assumption is reasonable at least when the water front position \( x_m \) is proportional to the imbibition time to the power \( n \), with the index \( n \) being a real number). From their approach the following form of the diffusivity is derived

\[ D_a(\vartheta, t) = -n t^{(1+\alpha)-1} \left| \frac{d \gamma}{d \vartheta} \right|^{\alpha} \int_{0}^{\vartheta} \gamma \, d \vartheta, \tag{7} \]

with \( \alpha = \frac{1}{n} - 1 \), \( D_a(\vartheta, t) \) is only a function of the water content \( \vartheta \) and is given by

\[ D_a(\vartheta) = -\frac{1}{1+\alpha} \left| \frac{d \gamma}{d \vartheta} \right|^{\alpha} \int_{0}^{\vartheta} \gamma \, d \vartheta, \tag{8} \]

The case \( \alpha = 1 \) corresponds to the Fick’s law \((1, 26)\).

\[ D(\vartheta, t) = -n t^{2n-1} \left| \frac{d \gamma}{d \vartheta} \right|^{\alpha} \int_{0}^{\vartheta} \gamma \, d \vartheta, \tag{9} \]

The classic time-independent form of the diffusivity corresponds to \( n = \frac{1}{2} \) in Eq. (9) and \( \alpha = 1 \).
EXPERIMENTAL SET UP AND MEASUREMENTS

Cubical concrete samples of 10 cm length and water to cement ratio = 0.4, were made according to the standard code for concrete mix and design\(^{(27)}\). Silica fume is added to the samples with percentages 0 \%, 5 \%, 10 \%, 15 \% and 20 \%. The samples were left to dry in normal atmospheric conditions for 28 days. The samples were then immersed in water bath with all sides except one of its sides to allow water to move in one dimension. The immersing times were 7 days, 14 days, 28 days and 35 days. The concrete samples were submerged in water with all sides except one of its side which was facing the incident neutron beam during the measurement of backscattered thermal neutron.

A neutron backscattered measuring system was designed and constructed to measure thermal neutrons backscattered from different positions along water follow. The measuring system consists of sample manipulating, neutron source, neutron detector and measuring electronics. The neutron source is Californium – 252 (It emits \(\sim 5 \times 10^6\) neutrons / second) and accommodated in specially design source shield made of high density polyethylene and boric acid. The incident neutrons on the sample under investigation, is a fine beam that leaves the shielded source through a slit collimator of the dimensions 2 mm width and 20 mm height. The beam is allowed to scan the middle line of the sample side which water did not infiltrate through it and along the flow direction. The manipulating table allows to move the investigated concrete sample across the incident fast neutron beam by certain distances. The sample was shifted across the beam by 2.5 mm steps. The back-emitted thermal neutrons result from moderation of fast neutron incident on the sample were recorded by a BF\(_3\) detector fixed on the shield housing the \(^{252}\)Cf source and beside the beam-exit. The BF\(_3\) detector was covered from all sides by Cd sleeve except a slit window, which has the same dimension as the neutron beam exit slit and fixed towards the same irradiation position to measure any thermal neutrons coming from the scanned position on the concrete sample. Further, Cd screen window (2.5 mm x 20 mm) was fixed between the sample and detector to prevent the contribution of side scattered thermal neutrons.

The signal obtained from the recorded scattered neutrons is amplified and then fed to a counter–timer for counting registration. Figure 1 shows a schematic diagram of the experimental set up. Once the scanning process has been finished for an infiltration time, the sample is returned back to the water bath to continue the infiltration process. Later, and after a longer time of infiltration, the process of neutron scanning is repeated and so on. The above experiments were performed for the samples with 0 \%, 5 \%, 10 \%, 15 \% and 20 \% additions of silica fume.

Fig. 1: Experimental arrangement.
RESULTS AND DISCUSSION

The relative count rates measured for different scanned positions on the investigated sample were used to plot relations between flux of thermal neutrons and scanned position for samples immersed in water for different periods of time. The results of water profiles measured with cadmium for the 0 %, 5 %, 10 %, 15 % and 20 % SF addition are shown in figs 2, 3, 4, 5 and 6 respectively. The relations presented in these figures show that they nearly have the same profile shape given by thermal neutrons measured from both sides of the sample for samples with same percentage of silica fume and immersed in water for the same time. They also show that the count rate decreases with increasing water penetration position and increasing silica fume percentages up to 15 %, but reincreases again for sample with 20 % this due to the increase of porosity of concrete sample. This be attributed to the silica fume has finer particles than other cementitious materials (average diameter of 0.1 μm, i.e., approximately 100 times smaller than Portland cement particles) and makes pore structure of concrete denser[28]. The rate of count depression depends on water penetration position; immersion time and silica fume percentage.

The obtained results were used to test the macroscopic theory of water infiltration in porous media. The \((\theta - \varphi)\) profiles for \(\varphi = \frac{x}{t^{0.5}}\) do not collapse to the universal one master curve for all the profiles. Figs. 7 & 8 show examples for this test. As a result, the macroscopic theory of water infiltration in porous media can not be applied to model the water infiltration in the concrete samples.

A literature approach to model the infiltration process is used in this work. The anomalous diffusion and the water front position is assumed to flow the \(t^n\) behavior with the exponent \(n\) more or less than the classical exponent 1/2. The water profiles were re-plotted against the generalized Boltzmann variable (GBV), \(\gamma = \frac{x}{t^n}\) with arbitrary values for the exponent \(n\). These \((\theta - \gamma)\) profiles for all \(n\) values less than the classical exponent 1/2, do not collapse to one master curve. Namely the data do not belong to the sub-diffusion behavior. Figures. 9 & 10 show examples for the \((\theta - \gamma)\) profile for 0 % SF addition; and assuming \(n = 0.4\) and 0.3. However, it was observed that when \((\theta - \gamma)\) profiles tend to collapse to one master curves, as the value of the exponent \(n\) increases above 1/2. The best value for the exponent \(n\), at which the profiles converge to one master curves for all SF additions, was found for \(n = 0.85\). Figs 11 & 12 shows examples for the collapse of the profiles \((\theta - \gamma)\) with \(n = 0.85\) for 0 % and 15 % SF additions. The master curves are much better than for the \(t^{1/2}\) - scaling, shown in Figs 8 & 9. As a result, the infiltration process in the studied samples belongs to the super diffusion behavior. Namely, the average distance that the advance of water particles moves in the samples is faster than the classical behavior predicted by Fick's law.

The diffusivity can be intuitively described as the ease of the spread of water particles[29]. The shape of the \(D(\theta)\)-function depends strongly on the experimental function \(\theta(\gamma)\). And vice versa, the shape of the diffusivity as a function of the water content determines the shape of water profiles[30, 31]. Let us consider the limits: (i) if \(D(\theta) \rightarrow 0\) for \(\theta \rightarrow 0\), the profiles are sharper than in the classical case (constant diffusivity) and it is called the hypodiffusive flow[30]. It occurs if a dry porous medium is allowed to imbibe fluids, for example water at low saturations, as shown from measured data. (ii) if \(D(\theta) \rightarrow \infty\) for \(\theta \rightarrow 0\), the water profiles have no sharp fronts, but show diffusive tails, which are longer than in the classical case. This happens when a pre-wet porous medium is allowed to imbibe wetting fluid and this hyperdiffusive behavior has been experimentally observed[31, 32].
Fig. 2: Water profiles under Cd for 0% SF for left (A) and right (B) sides.

Fig. 3: Water profiles under Cd for 5% SF.

Fig. 4: Water profiles under Cd for 10% SF.

Fig. 5: Water profiles under Cd for 15% SF.

Fig. 6: Water profiles under Cd for 20% SF.

Fig. 7: The $\Theta - \varphi$ master curve profiles with $\varphi = \frac{X}{x^{0.5}}$ for 0% SF for left side.
Based on the experimental knowledge of the well-converged master curves the profiles i.e., the \((\theta - \varphi)\) curves, the water diffusivities can be derived in analytical way. The following analytical formula was used to fit \(\theta\) vs \(\gamma\):

\[
\frac{\theta}{\varphi} = \frac{x}{t^{0.5}}
\]
\[ \theta = \theta_s \cdot \text{Exp} \left( m \gamma \right), \]  
(10)

where \( \theta_s \) and \( m \) are free fit parameters, with \( \theta_s \) corresponds to the highest water content. An example of the fitting using this formula is shown in Figs 13 & 14. From the fitting, for all the studied samples, the parameters \( m \) and \( \theta_s \) were determined. \( m \) was roughly found constant within 5 % for \( m = -8.87 \). Using this formula, the water diffusion for the case \( \alpha = 1 \) and corresponds to the Fick's law \( (1, 26) \), is derived in the form given by:

\[ D(\theta, t) = t^{2n-1} \cdot \frac{n}{m^2} \cdot \left[ 1 - \ln \left( \frac{\theta}{\theta_s} \right) \right], \]  
(11)

The water diffusivities were determined using formula 11 for all silica fume additions, and the obtained results are shown in Fig.16. This figure shows that, the diffusivity decreases as the water content increases. Namely, the concrete samples show a hyperdiffusive character. This hyperdiffusive behavior for the investigated samples was not been observed and reported in literature before. However, it was mentioned that hyperdiffusive behavior is observed if a pre-wet porous medium, is allowed to imbibe wetting fluid \( (31, 32) \). As a result, this is a new flow regime: the concrete samples are dry before starting the infiltration process and the water diffusivities belong to the hyper diffusive behavior.

Accordingly, it can be expected that, the effect of silica fume additions to concrete mix on the infiltration process and hence on water diffusivities is positive. And the infiltration process as well as the water diffusivities, decreases as the silica fume addition increases. This behavior is valid till 15 % SF addition. At the 20 % SF addition, the diffusivity is higher than for the 15 % SF (see Fig. 15). This means that there is a maximum value for the silica fume addition \( \approx 15 \% \) beyond which the diffusivities starts to increase. i.e., for the silica fume percentage \( \geq 20 \% \), the available pore space in the sample becomes higher than the pore space at 15 %. This could be attributed to deterioration in concrete physical and mechanical properties as shown Fig 16. This means that the method NBS for water infiltration can be used for rough estimation the changes and damage in mechanical properties by increasing the percentages of SF.
[Image of Fig. 15 Water diffusivities for all silica fume additions.

Fig. 15 Water diffusivities for all silica fume additions.]

[Image of Fig. 16 The effect of silica fume additions on compressive strength of concrete.

Fig. 16. The effect of silica fume additions on compressive strength of concrete.]

The anomalous diffusion behavior which is observed in this work via the time-dependence of diffusivity, Eq. 10, is considered within the Darcy (Fick) approach ($\alpha = 1, \, n \neq 1/2$). The time-dependence of the diffusivity was before observed and discussed in soil measurements. According to Guerrini and Swartzendruber (33), the wetting (imbibition) process produces some microlevel rearrangement of the soil particles - a change in the microstructure, but without any net movements of the particles in the direction of water flow. Hence there is no change in the bulk-density of the soil. Nonetheless, this bulk-density invariant change in the microstructure can still alter the geometry of the pore space, with a resultant (probably reduction) in mean pore size. Recently, for building materials the situation was summarized as follows (26): the interactions of the of water with the porous materials “the so-called chemo-mechanical changes” tend to restrict the pore throats and affect connectivity without significantly changing the material’s total effective porosity. Such understanding of the mechanism was a motivation for introducing the time-dependence of the diffusivity and for interpreting of the water front movement deviations from the $t^{1/2}$-scaling. The anomalous diffusion ($\alpha \neq 1, \, n \neq 1/2$), with time-independent diffusivity was not used in this work. From a physical point of view, it seems to support the opinion of Lockington and Parlange (26), that water in porous materials, may have some non-Newtonian features.

Kuntz and Lavallee (34, 35) have made some numerical simulations for the diffusion in a microscopically random heterogeneous structure using, the so-called lattice gas automaton model. The purpose of their study was to search for the possible reasons for the deviation of the water front from the $\sqrt{t}$-scaling (34). They have found that the deviation from the $\sqrt{t}$ behavior of liquid flow may be due to the diffusivity gradient with respect to the water content. Moreover, if the diffusivity gradient is positive, i.e., the diffusivity increases with the water content; the water front position can be scaled with the power law in time with indices higher than $1/2$. They claimed that if the diffusivity gradient is negative, i.e. diffusivity decreases with the water content; the water front can be scaled with power law in time with indices smaller than $1/2$. Such findings are in contradiction with the present results. Thus, diffusivity gradient either negative or positive is not responsible for anomalous diffusion; either it is super or sub-diffusion.
CONCLUSIONS

Addition of silica fume to concrete mixes with percentage up to 15 % improves the physical and mechanical properties of concrete. These improvements tend to decrease the water infiltration in concrete due to the decrease of porosity. However, for concrete with silica fume more than 15 % the concrete properties deterioration and with an increase in water intrusion is observed.

Water infiltration in concrete with all silica fume additions, follows the super diffusion behavior, i.e., the average distance that water particles moves in concrete is more faster than the classical behavior predicted by Fick’s low.

A hyper diffusion behavior is observed due to the decrease of diffusivity by increasing the water content in concrete.

Water infiltration and diffusivities are less in concrete mixes with silica fume less than 20 %. (as the SF addition increases, the diffusivity decreases till a threshold value of 15 % of SF).

The obtained results indicate that a measurement of water infiltration by neutron backscatter is a reliable method to study water intrusion in porous media.

REFERENCES

(2) C. Hall, Build. Environ.; 12, 117 (1977).