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An Approach to Determine Diffusivity in Hardening Concrete Based on Measured Humidity Profiles

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A novel method to numerically determine the diffusion coefficient of hardening concrete from measured pore relative humidity profiles is presented, which is based upon an inverse nonlinear diffusion analysis. A system for nondestructively measuring the development of pore relative humidity required for this analysis in a uniaxially drying concrete specimen is described. The diffusion equation is solved by the method of lines, with collocation in the space variable to reduce the problem to a system of ordinary differential equations. Based on the solution of the diffusion equation, an auxiliary formulation is derived by searching the calibration factor in diffusivity that minimizes the residual between the calculated relative humidity and the measured data at each measured point in each small time interval. A working algorithm and numerical results are presented in this paper. ADVANCED CEMENT BASED MATERIALS 1995, 2, 138–144

KEY WORDS: Calibration, Desorption isotherm, Diffusion coefficient, Hardening concrete, Inverse problem, Pore relative humidity

During the last few decades, a number of efficient numerical methods have been developed for the numerical solution of initial and boundary value problems in physics and applied science. The main emphasis in the research on numerical analysis has been on developing accurate methods. In both theoretical and practical applications of numerical methods, it is usually assumed that the physical parameters appearing in the methods are accurately known. In practice, however, this may rarely be the case. Also, in many cases, the exact determination of the model representing a physical phenomenon under consideration may be more difficult to achieve than a determination of the actual solution of the model. From a practical perspective, measurements made of moisture content, or pore relative humidity, and temperature field in a concrete material may provide an input for estimating the physical quantity that allows the model to accurately fit the measured data.

The diffusivity or diffusion coefficient of drying concrete is an important physical parameter in the mathematical simulation of moisture transport process in concrete and the numerical analysis of the macroscopic behavior of concrete [1,2]. It is impossible, or at least considerably difficult, to obtain this material parameter directly by experiment. Traditionally, this parameter is evaluated by fitting the measured moisture content data obtained from drying tests by nonlinear regression analysis. Sakata [3] proposed an analytical solution for the diffusion coefficient as function of moisture content based on the Boltzmann transformation theory. The solution required the knowledge of the relationship between the measured moisture content and the Boltzmann variable, which was formulated approximately by fitting the experimental data from drying tests. Penev and Kawamura [4] adopted a similar approach to get diffusivities for soil-cement mixtures and lean concrete. However, the requirement that the initial and boundary conditions should be expressible in terms of Boltzmann variable is hard to satisfy when drying occurs in a concrete structure with finite thickness such as concrete pavement. Moreover, little effort has been documented verifying the results by substitution of the diffusion coefficient into a drying model to obtain a solution to match the measured data at different times and locations. Recently, the solution method for the diffusion coefficient has been improved by Wittmann [5,6]. The "best" diffusion coefficient is determined based on the finite difference method and combined with nonlinear least square fit method by comparing the numerical solution with the experimental results. The purpose of this paper is to present a method of determining the calibrated diffusion coefficient from inverse diffusion analysis for a given mixture of concrete while hardening.

Recent research on the experimental investigation of pore humidity profiles in drying concrete has sug-

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Received April 12, 1993; Accepted November 2, 1994

ISSN 1065-7355/95/$9.50
SSDI 1065-7355(94)00034-B

gested the use of small probe-type sensors to make continuous measurements of bulk relative humidities at different distances from an exposed surface. This method gives an opportunity to obtain a clear indication of spatial variations of pore humidity in concrete without excessive labor or destructive testing of the specimen and gives an improved accuracy over conventional gravimetric methods for the measurement of moisture content [7,8]. These experimental instruments demonstrate a high potential for practical implementation in the quality control of construction [9] and can provide a measure of model validation with respect to the drying process in concrete. This type of information can also provide insight into the properties and behavioral characteristics of concrete structures such as pavements due to different exposure conditions. Therefore, it is expected that the pore humidity profile near the concrete surface can be determined with sufficient accuracy, particularly at early ages, to provide significant insight into the analysis noted previously. A numerical procedure is given in detail in this paper to recover the diffusivity based on the measured pore humidity profiles.

Physical Basis and Assumptions

Drying of concrete is a complicated moisture transport process. It includes primarily the effects of capillary pressure, disjoining pressure, and liquid and vapor diffusion. It is generally accepted (or perhaps preferred) that the drying process in porous concrete can be purely described by diffusion theory based on the Fick's law [3–6,10,11]. It is assumed phenomenologically that the rate of transfer of moisture content through a unit area of a section is proportional to moisture content gradient as the solely driving force measured normal to the section. Mathematically, this moisture transport process can be formulated as:

$$J = -D \operatorname{grad} C \tag{1}$$

where J = moisture flux ($ML^{-2}\,T^{-1}$); C = moisture content (ML^{-3}); and D = diffusion coefficient ($L^2\,T^{-1}$). The mass conservation equation is expressed as:

$$\frac{\partial C}{\partial t} = -\operatorname{div} J \tag{2}$$

Substituting eq 1 into eq 2, one obtains

$$\frac{\partial C}{\partial t} = \operatorname{div}\,[D \operatorname{grad} C] \tag{3}$$

For one-dimensional case, the eq 3 can be simplified to:

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial x}\left[D\frac{\partial C}{\partial x}\right] \tag{4}$$

By introducing the desorption isotherm that defines the relation between the moisture content C of the concrete and the relative humidity h of the surrounding air, the eq 4 can be rewritten by using the chain rule as:

$$\frac{\partial C}{\partial h}\cdot\frac{\partial h}{\partial t} = \frac{\partial}{\partial x}\left[D\frac{\partial C}{\partial h}\cdot\frac{\partial h}{\partial x}\right] \tag{5}$$

If the slope of the desorption isotherm is constant over a wide range of humidity [12], or if the moisture content is assumed to be a piecewise linear function of relative humidity in a small range of humidity, the eq 5 can be simplified to:

$$\frac{\partial h}{\partial t} = \frac{\partial}{\partial x}\left[D\frac{\partial h}{\partial x}\right] \tag{6}$$

where h = relative humidity (%) and D = diffusion coefficient ($L^2\,T^{-1}$).

It is worth noting that the diffusion coefficient D is still a function of the moisture content C, which is in turn a function of relative humidity h manifest in the desorption isotherm relationship, although the diffusion equation is expressible in terms of relative humidity. Experiments show that the desorption isotherm for a given mixture is not only a function of temperature but is also the degree of hydration [13,14] (Figures 1 and 2). For the sake of simplicity, it is assumed that drying occurs under quasi-isothermal conditions with constant ambient temperature, and present studies focus on the initial desorption with constant ambient humidity. Although adjustment of cement content in a concrete mix and the ambient temperature of specimen in a laboratory may control the hydration process to reach the quasi-isothermal condition, the temperature development due to heat of hydration cannot be avoided. It is found that the effect of temperature on pore humidity variation is too small and may be neglected when relative humidity in concrete is higher than 90% (Figure 2).

Mathematical Model

It is well known that if the diffusion coefficient, boundary conditions, and geometric information are known, pore relative humidity distribution in concrete structure could be obtained by solving the diffusion equation when it can be written in terms of relative humidity for the aforementioned restrictions as:

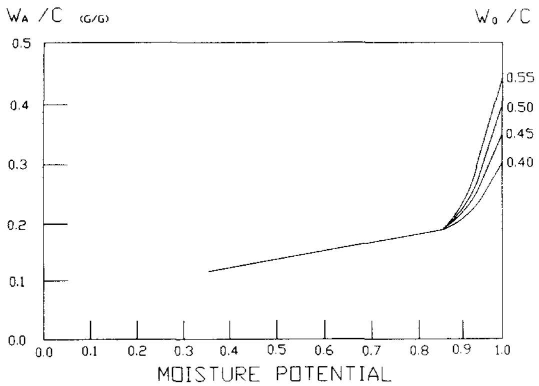

FIGURE 1. Typical desorption isotherms for a given degree of hydration and temperature varying water: cement ratio [14].

$$\frac{\partial h}{\partial t} = \frac{\partial}{\partial x}\left(D\frac{\partial h}{\partial x}\right), \; 0 < x < d, \quad 0 < t \leq T \tag{7}$$

where the relative humidity $h = h(x,t)$ in concrete depends only on one space variable x in diffusion direction and the time variable t. It is subject to the initial condition:

$$h(x,0) = 1.0 \quad 0 < x < d \tag{8}$$

and boundary conditions:

$$\begin{aligned} h(0,t) &= f(t), \quad 0 \leq t \leq T \\ h(d,t) &= g(t), \quad 0 \leq t \leq T \end{aligned} \tag{9}$$

where D is the diffusion coefficient, which is a given material parameter in the aforementioned diffusion equation.

Conversely, the diffusion coefficient could be recovered from the solution of the inverse problem or inverse diffusion analysis if the part of the relative humidity distribution, boundary conditions, and geometric information are known. Assuming that the measured relative humidity $h^*(x_i,t_j)$ at the points x_i, $i = 1, \ldots, M$ (total number of measured points) at time levels t_j, $j = 0, \ldots,$ NK ($= T/\Delta t$) is known from drying test, in addition to the initial and boundary conditions stated earlier, the diffusion coefficient D must be determined such that the solution $h = h(x,t)$ of eq 7 matches the measured values at each measured point and at each time level.

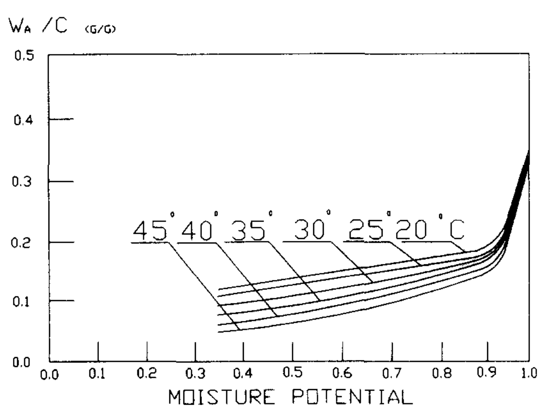

FIGURE 2. Typical desorption isotherms for a given degree of hydration and water: cement ratio varying temperature [14].

Formulation of Diffusivity

Experimental studies [15] show that diffusion coefficient is not a constant but depends on number of factors, mainly the moisture state, the concrete composition, the concrete age, and pore structures when pore humidity is the physical quantity to be measured. Concrete properties may change with time particularly during the early ages, that is, the transport parameters are dependent on degree of hydration. The composition of a concrete mix is typically not uniform in the direction of diffusion due to the inhomogeneity inherent in concrete construction practice and the effect of thermal gradients. The gradient of microstructure below the exposed surface has been observed by many researchers [16–18]. The spatial variation of pore structures quantitatively described by porosity $\eta(x)$ and its effect on vapor diffusion cannot be ignored when pore humidity is used as measured physical quantity to back-calculate the moisture transport coefficient for concrete. For illustrative purposes, a general form of diffusivity (D) is assumed in this paper as the following:

$$D = D\,(\eta(x),\, t_e,\, h) \tag{10}$$

where $\eta(x)$ defines the spatial variation of porous structures and t_e is the equivalent hydration period. Due to the effects of porosity and the degree of hydration on vapor movement in concrete, these parameters are included directly in the general formulation. However, they are only indirectly considered here (in terms of the coordinate parameter, x, and real time t) to demonstrate a process to back-calculate (D). Realizing this limitation, efforts are currently underway at Texas A&M University by the authors to formulate a porosity relationship as a part of the back-calculation process to improve upon the utility of this method.

Nevertheless, the simplification of the mathematical form in eq 10 is necessary to numerically evaluate diffusivity in terms of measurable data taken from laboratory specimens. A commonly used method assumes an explicit function for (D) with a finite set of undetermined coefficients. Those unknown coefficients are identified by the inverse moisture diffusion analysis, or an auxiliary optimization problem, that minimizes the residual between calculated solution and measured data. The challenge is to determine the mathematical form for (D) when there is more than one dependence. Pilant and Rundell [19] recommended an approximation to (D) with the following form:

$$D(x, t, h) \approx D_1(x) + D_2(h) + D_3(t) \tag{11}$$

where (D_i) ($i = 1,2,3$) corresponds to the first few terms in an expansion for (D). If one dependence [say $D(h)$], dominates the others [$D(t)$ and $D(x)$], the primary dependence can be determined by the commonly used method when the other dependencies are fixed, and possibly the other dependencies can be determined after fixing the form of the main dependence. However, the shortcoming of this method is the validation of the assumed mathematical form of (D).

In this paper, the following simplified form is used for determining the diffusivity D by the strategy of data-coefficient mapping:

$$D(x, t, h) = \alpha \cdot h\,(x_i, t_j) \tag{12}$$

It is assumed that the diffusivity (D) is a linear function of h, that is, the (D) is proportional to h over a small time interval. The assumption of linearizing the nonlinear diffusion coefficient is acceptable, because the drying diffusion rate of concrete is low except at the surface immediately after placement. When this form is substituted into the partial differential equation, the diffusion problem is still nonlinear. The term α in eq 17 is called a calibration factor that may be determined by matching the calculated relative humidity to the measured data at each measured point in each small time interval. Because the diffusivity (D) is recovered point by point in the x direction and a linear function is used in a piecewise approximation in the t dimension, the diffusivity (D) is still a function of x, t, and h. Because of this, interior measurements and boundary measurements are generally needed. It is preferable to use the piecewise approximation, because the readings in time dimension are much larger than the number of measured points. After the D field is recovered by the iterative scheme discussed in the following section, a spline interpolating function can be used to express this (D) field, or a simplified explicit function for (D) may be found by visualizing the recovered D field.

Solution of Inverse Problem

In the process of the inverse diffusion analysis, the drying diffusion equation is solved repeatedly. Therefore, the accuracy and efficiency of the inverse diffusion analysis depends on the solution method for the diffusion equation. An approach to the solution of the nonlinear drying diffusion equation has been achieved by the use of a step-by-step integration process in time using the forward differences approach and the finite difference method in the space coordinate. To solve this nonlinear equation, the nonlinear diffusion coefficients are linearized by evaluating them at the previously calculated solution for this time step. However, this may lead to serious stability and accuracy problems when the diffusion coefficient (D) becomes so small as relative humidity decreases. Further improvements may be made by decreasing the step Δt. But for convergence, Δt may be required to be so small that it is impractical to find a solution. Not only nonlinearity, accuracy, and stability difficulties exist in using the finite difference method; there are also difficulties with flexibility, complexity, and the required effort in the implementation of this method [20].

In this paper, the so-called numerical method of lines, one of the more widely used methods, is chosen for the solution of the time-dependent partial differential equation (PDE). The method can be described as follows. The spatial variable x is discretized by the use of a finite element collocation procedure with piecewise polynomials. The collocation procedure reduces the PDE system to a semidiscrete system [actually an initial value ordinary differential equation (ODE) system], which then depends only on the time variable t. The time integration is then accomplished by the use of the robust ODE solver [21,22]. With the software available in IMSL MATH/Library or NAG Fortran Library for such analysis, the user can specify an error tolerance and also monitor the stability of the calculations.

Based on the solution of the moisture diffusion equation, an auxiliary problem to the inverse problem is formulated. A class of factor, α, is sought to ensure best agreement between the calculated moisture and measured moisture at each measured point for certain time interval. Now the minimization problem is written when $x = x_i$, $i = 1, \ldots, M$:

$$\min_{\alpha_i} \left\{ \sum_{k=1}^{NK} \left(h(x_i,t_k,\alpha_i) - h^*(x_i,t_k)\right)^2 \right\} \quad t_k \in (t_j,t_{j+1}) \tag{13}$$

First the finite set of parameters $\{\alpha_1, \alpha_2, \ldots, \alpha_{NK}\}$ must be determined based on the previous results [12] to define the possible range of the calibration factors. Then the optimum factor α in diffusivity is found at each measured point in each small time interval by the golden section search technique [23]. The iterative scheme is shown in Figure 3.

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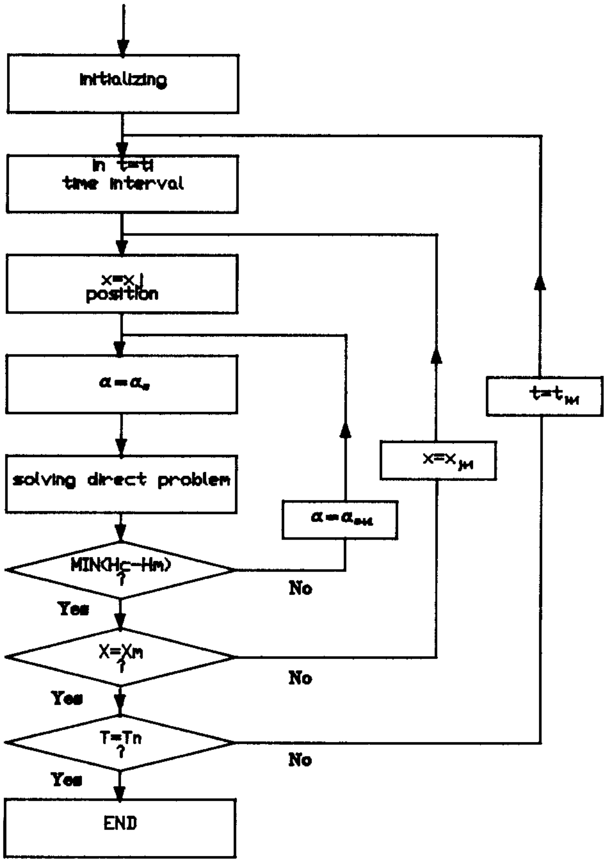

FIGURE 3. Iterative scheme.

Experimental Method

A drying experiment was conducted with a small probe-type relative humidity sensor in a environmental chamber to estimate the diffusion coefficient. The most common sensor in practice is the thin-film capacitance-type humidity sensor [7,15]. These sensors comprise parallel electrodes on a glass substrate covered by a thin film of a hygroscopic polymer. The sensor monitors the change in capacitance of the thin polymer film as it absorbs water vapor. The full response of the probe can be achieved within a few seconds at relative humidities below 80%, but at higher humidities the response becomes inconveniently slow. Contrary to the capacitive humidity sensor, the bulk polymer resistive humidity sensor, in which the electrical resistance of the humidity sensitive film responds to changes in humidity absorption and desorption, responds more quickly at very high humidities with negligible hysteresis. This feature is essential when the moisture movement and the effect of the heat of hydration in concrete at early ages is a factor. In addition, this type of sensor apparently provides more accurate results than the capacitive-type sensor does at high humidities.

To ensure the accuracy of the relative humidity measurements in concrete materials with these sensors, especially at high humidity range, the chilled mirror optical dew point meter was used to calibrate the low level capacitive type or resistive type RH sensors. A typical calibration curve is shown in Figure 4 after re-adjustment for obtaining linear performance at high humidities. Any nonlinear behavior of these sensors at low humidities can be accounted for by utilizing the calibration curve.

A 2.5-ft^3 specimen was cast in a plywood mode. The cast specimen is covered on all sides by insulation board covered with aluminum foil except the top surface, which allows one-dimensional moisture movement vertically through the specimen as shown in Figure 5. Relative humidity measurements are made through holes, with their center-line axis perpendicular to the moisture flow, that extend from the side wall into the interior portion of the specimen. The depth of the holes from the top surface range from 1.5 inches to 11.5 inches with 1-inch intervals. The holes are formed with PVC pipe to protect the surface of the hole as the sensors are moved in and out of position. One end of each pipe is sealed by a rubber stopper when the sensor is not in place. The opposite end of the pipe can be covered by fine screen to block the cement paste and fine aggregate from making contact with the sensor but to allow the moisture vapor to freely move into the PVC pipe. With these instruments, the temperature and relative humidity can be measured simultaneously at any concrete age. Initially, an experiment was conducted under constant ambient temperature and relative humidity even though it may be considered that moisture conditions under varying thermal condition is more representative of conditions in the field.

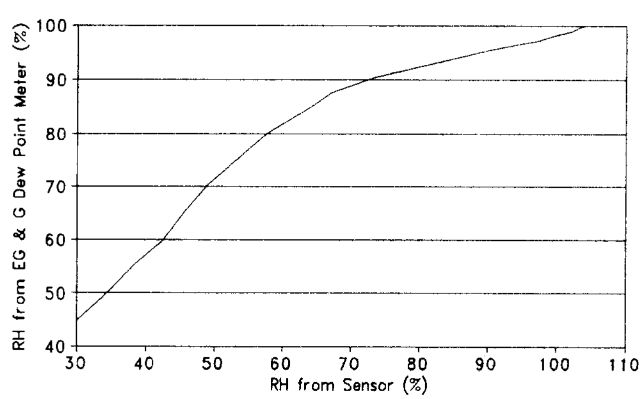

FIGURE 4. Typical calibration curve for RH sensor after re-adjustment.

FIGURE 5. Specimen mould sealed on five faces and with 11 cylindrical holes for measuring relative humidity.

Application and Discussion

A practical application of the mathematical method described previously is in the determination of the diffusion coefficient of the concrete specimen shown in Figure 5. The concrete that was used in the specimen consisted of 292.2 lb of river gravel as coarse aggregate, 231 lb of sand, 65.8 lb of type I cement, 23.1 lb of fly ash, and 35 lb of water. After casting, the concrete specimen was cured in the laboratory at 70% relative humidity and 76°F. The measured moisture profiles are shown in Figure 6. The diffusion coefficient for the concrete specimen is shown in Figure 7, which is determined by the aforementioned algorithm. It is noted that the diffusivity of the outer skin or top layer of the concrete specimen declines sharply in the first few days after casting. At later concrete ages, the diffusivity is so small that little movement of interior moisture occurs as evidenced by the high level of the interior relative humidity in the concrete specimen.

The maximum error between two values at each point is less than 0.2%, which is much less than the accuracy of the sensors used in this research. It is found that the recovered moisture diffusivity depends heavily on the length of time interval, that is, the number of measuring point in each interval. The length of time interval is chosen in such a way that the maximum difference between calculated relative humidity and measured relative humidity is less than the possible system error of relative humidity measurement.

The diffusivity field recovered by the proposed method is discrete, because the measured relative humidity field is discrete. For practical application in predicting relative humidity distribution and history in concrete structure, the diffusivity field can be expressed by a two-dimensional tensor-product spline function as:

$$D = \alpha \times h(x_i, t_j) =$$

$$\sum_{i=1}^{M} \sum_{j=1}^{N} c_{ij} B_{i,k_x,t_x}(x) B_{j,k_t,t_t}(t) \times h(x_i, t_j) \qquad (14)$$

where c_{ij} is the coefficient and $B_{i,k_x,t_x}(x)$ and $B_{j,k_t,t_t}(t)$ are the B-spline.

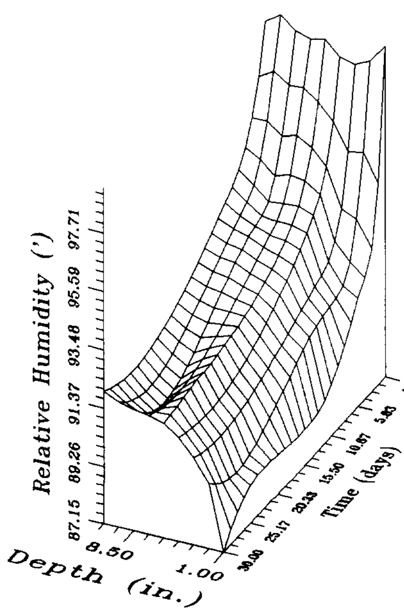

FIGURE 6. Three-dimensional view of measured moisture profile in the concrete specimen.

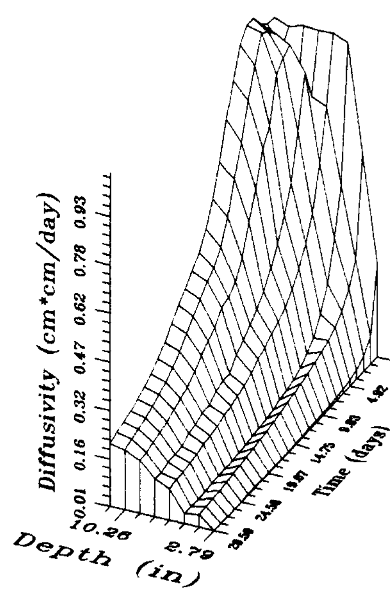

FIGURE 7. Three-dimensional view of diffusion coefficient of the concrete block.

The spline function can be evaluated using the software available in IMSL MATH/Library or NAG Fortran Library.

Validation of the correlation between diffusivity and microstructural properties can be made with the proposed numerical method. However, further research is needed to quantitatively describe porous structures such as porosity, tortuosity, and constrictivity.

Conclusions

Complete numerical method has been presented in this paper to determine the diffusion coefficient required for the drying shrinkage analysis based on the measured relative humidity profiles. The method uses a least square minimization for ensuring best agreement between the measured and calculated relative humidities. With the software available in IMSL MATH/Library of NAG Fortran Library for the solution of time-dependent partial differential equations, the proposed algorithm can be easily implemented.

Acknowledgments

The authors are pleased to acknowledge the support of the Texas Department of Transportation (TxDOT), in cooperation with the U.S. Department of Transportation, Federal Highway Administration (FHWA). Special acknowledgment is made to A-TEK for the conscientious assistance with the experimental instrumentation.

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