A NUMERICAL ANALYSIS TECHNIQUE TO EVALUATE THE MOISTURE-DEPENDENT DIFFUSION COEFFICIENT ON MOISTURE MOVEMENT DURING DRYING

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ABSTRACT

The conventional method of determining the diffusion coefficient of wood as a function of moisture content is based largely on the moisture content profile data using a slicing technique. This method is destructive and subject to inherent error because it uses average moisture content values and different samples at various drying times. This paper describes an alternative nondestructive approach using a numerical technique to evaluate the moisture-dependent diffusion coefficient using drying curves. It gives an accurate prediction of moisture movement through the entire drying process.

Keywords: Numerical analysis, diffusion coefficient, drying, fiber saturation point.

INTRODUCTION

The diffusion model based on Fick's second law has been used to predict the removal of moisture from wood since diffusion controls the moisture movement during drying (Stamm 1964). When a constant diffusion coefficient is assumed, theoretical solutions to the diffusion model are readily available under simple boundary conditions, e.g., letting the surface moisture content (MC) drop immediately to equilibrium with the surroundings or assuming a constant surface emission coefficient (Skaar 1954). However, as summarized by Moschler and Martin (1968), the simple assumptions usually do not hold. The diffusion coefficient is generally not a constant, and the surface MC changes gradually toward equilibrium with the surroundings. An accurate prediction of drying curves and the distribution of moisture within wood cannot be achieved without considering the effect of wood MC on the solution to the diffusion equation and the change of surface MC (Choong 1965; Bui et al. 1980).

When the diffusion coefficient is a function of MC, there is no closed form of theoretical

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solutions to the governing partial differential equation based on Fick's second law. If a constant diffusion coefficient and functional boundary conditions are assumed, the theoretical solutions expressed as infinite Fourier series may exist, but are usually quite complicated (Crank 1975; Carslaw and Jaeger 1986). Therefore, in both cases, numerical analysis techniques are the most practical means to evaluate the diffusion coefficient.

In the past, the moisture dependence of the diffusion coefficient has been evaluated based on the MC profile data obtained by a slicing technique. Moschler and Martin (1968) have given several polynomial relationships between the diffusion coefficient and MC at 40°C for yellow-poplar. These relationships were derived from MC profile data at specific times during the drying process. When substituting the derived expression into the diffusion equation, they could obtain a better prediction of the MC profiles than by using other available diffusion coefficients from the literature. The best prediction could be achieved only when the samples were examined for a relatively long drying time. They attributed the discrepancy to several factors: the violation of initial and boundary conditions, experimental error caused by using the average MC distribution, lack of homogeneity on the macro or micro scale in wood, and the time-dependent stressrelated factor.

Stevens et al. (1985) presented a similar study on the relationship of the diffusion coefficient to MC at 32°C for yellow-poplar. Their empirical equation is in an exponential form with three adjustable parameters. The initial condition was obtained from the MC gradient after the samples had been dried for a period of time to allow the surface MC to drop close to equilibrium with the surroundings. They reported that the experimentally determined relationship between the diffusion coefficient and MC could predict moisture gradients fairly accurately during isothermal nonsteady-state moisture movement, and the relationship between the diffusion coefficient and MC did not change over the course of desorption.

In the above studies, obtaining accurate data of MC profiles is essential for the calculation and requires specific tools and laboratory techniques. Since only the average values of MC profiles are measured, an inherent error is introduced into the calculation. Inhomogeneity among samples may lead to inaccuracy since variation in moisture profiles among samples is confounded with the calculated parameters used to relate the diffusion coefficient to MC.

In order to reduce the experimental effort and eliminate the inherent error accompanied by the slicing technique, an alternate approach to evaluating the moisture-dependent diffusion coefficient using drying curves is suggested. This approach is based on the idea that the drying process is diffusion-controlled and the diffusion coefficient is moisture-dependent. Therefore, the diffusion model should satisfactorily predict the experimental drying curve if a functional relationship between the diffusion coefficient and the MC as well as the boundary condition of the model is appropriately defined. In this study, a linear relationship between the diffusion coefficient and MC is assumed because of its simplicity and informativeness. An exponential boundary condition is used to account for the fact that the surface equilibrium condition is not established instantaneously. This choice of the boundary condition is justified by Moschler and Martin's (1968) experimental data and Liu's (1989) theoretical analysis.

The objective of this study was to determine if and to what extent the moisture-dependent diffusion coefficient and the change of surface of moisture content affect the predictability of moisture removal during drying. The hypothesis is that using a numerical analysis technique, a close prediction of the experimental drying curves could be achieved if the effect of the variation of diffusion coefficient and surface moisture content was considered when solving the diffusion equation.

NUMERICAL ANALYSIS TECHNIQUE

The diffusion equation used in the present study was the common form of Fick's second

law, which has been applied by many researchers (Moschler and Martin 1968; Rosen 1976; Stevens et al. 1985)

$$\frac{\partial \mathbf{m}}{\partial t} = \frac{\partial}{\partial \mathbf{x}} \left(\mathbf{D} \frac{\partial \mathbf{m}}{\partial \mathbf{x}} \right) \tag{1}$$

where m is MC/100, t the drying time, x the distance coordinate, and D the diffusion coefficient which is generally a function of m.

Several finite difference schemes can be used to solve Eq. (1) under given initial and boundary conditions. The classic explicit approximation that has been used by previous researchers (Moschler and Martin 1968; Stevens et al. 1985) is simple and easily programmable, but the stability requirement must be strictly satisfied in order to obtain a convergent solution. In addition, it contains a relatively large truncation error of the time grid of the order of Δt , or O(Δt). To avoid the limitations of the classic explicit approximation, a linear threelevel finite difference scheme was applied. This scheme is an implicit approximation, which is unconditionally stable, convergent, and possessing a smaller truncation error in the time grid of $O(\Delta t)^2$.

The three-level finite difference scheme can be stated as follows (Lapidus and Pinder 1982):

$$(\mathbf{m}_{i+1,j} - \mathbf{m}_{i-1,j}) = \frac{2}{3} \rho \{ \mathbf{D}_1 [(\mathbf{m}_{i+1,j+1} - \mathbf{m}_{i+1,j}) + (\mathbf{m}_{i,j+1} - \mathbf{m}_{i,j}) + (\mathbf{m}_{i-1,j+1} - \mathbf{m}_{i-1,j})] - \mathbf{D}_2 [(\mathbf{m}_{i+1,j} - \mathbf{m}_{i-1,j})] + (\mathbf{m}_{i,j} - \mathbf{m}_{i,j-1}) + (\mathbf{m}_{i,j} - \mathbf{m}_{i,j-1})] \}$$
(2)

where $\rho = \Delta t/(\Delta x)^2$ and i and j refer to the location of m along the t and x coordinates. In this study, the Δt and Δx were 0.01 hour and one-tenth of the sample half-thickness.

With the assumption of a linear relationship between D and m, D_1 and D_2 can be calculated according to the following equations:

$$D_1 = a_0 + a_1[(m_{i,j+1} + m_{i,j})/2]$$
(3)

$$D_2 = a_0 + a_1[(m_{i,j} + m_{i,j-1})/2]$$
 (4)

where a_0 and a_1 are constant.

For the surface MC, m_s , an exponential decay from the initial MC to equilibrium with the surroundings was assumed, i.e.:

$$m_s = m_e + (m_i - m_e)exp(-\beta t)$$
 (5)

where m_i and m_e are the initial and equilibrium MC (on a fractional basis), and β is a surface parameter to account for the changing rate of surface MC.

The only restriction in the above equations is that the diffusion coefficient D must be greater than zero.

At the beginning of the calculation, the MC values on the first level are determined according to the initial condition. The values on the second level can be calculated according to the following equation:

$$(\mathbf{m}_{i+1,j} - \mathbf{m}_{i,j}) = \rho[\mathbf{D}_1(\mathbf{m}_{i+1,j+1} - \mathbf{m}_{i+1,j}) - \mathbf{D}_2(\mathbf{m}_{i+1,j} - \mathbf{m}_{i+1,j-1})]$$
(6)

When a set of parameter values in Eqs. (3), (4), and (5) is chosen, a theoretical drying curve can be generated. At each time the average MC of the sample is measured experimentally, the theoretical MC is calculated by applying Simpson's rule. The criterion for the optimum parameter values is that they yield the minimum absolute value of Negative Mean Sum of Squares (NMSS), defined as $-\Sigma (\Delta m)^2/N$. Here Δm and N represent the difference in m between the theoretical drying curve and the experimental data, and the number of data points, respectively. The above definition of the NMSS is applicable to multidrying curves as well. FORTRAN codes have been developed to facilitate the calculation of the NMSS for either a single drying curve or multidrying curves. Several multivariable search methods (Pike 1986) may be applied to help find the optimum values of a_0 , a_1 , and β .

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NUMERICAL EXAMPLES AND DISCUSSION

Four cases were examined based on the following assumptions:

- Case 1. The diffusion coefficient is constant, and the surface MC drops to equilibrium with the surroundings at the beginning of drying.
- Case 2. The diffusion coefficient is a linear function of moisture content, and the surface MC drops to equilibrium with the surroundings at the beginning of drying.
- Case 3. The diffusion coefficient is constant, but the surface MC decays exponentially to equilibrium with the surroundings.
- Case 4. The diffusion coefficient is a linear function of moisture content, and the surface MC decays exponentially to equilibrium with the surroundings.

In order to avoid frequent statements of the properties, the cases will be referred to by numbers unless mentioned otherwise. For convenience, the term "equilibrium boundary condition" refers to the condition when the surface MC drops to equilibrium with the surroundings, and the term "exponential boundary condition" is defined by Eq. 5 where the surface MC decays exponentially to equilibrium with the surroundings.

Example 1. Drying below the fiber saturation point (FSP)

Choong and Skaar (1969) evaluated the diffusion coefficient and the surface emission coefficient using their approximate method for yellow-poplar sapwood and heartwood specimens, dried in either the tangential or the radial direction. Two samples of different thicknesses were dried for each combination in an environmental chamber at a temperature of $32 \pm 0.2^{\circ}$ C and 40% relative humidity (RH) with an air speed of 3.3 m/sec. Using these data, Chen et al. (1995b) showed that if the diffusion coefficient was treated as a constant, a close prediction to the drying data for yellowpoplar sapwood in the tangential direction over the entire drying period could not be achieved. For the convenience of comparison, the same data were also used to analyze the performance of the current numerical analysis technique. In Choong and Skaar's experiment, the initial MC of the samples was 25%. The equilibrium moisture content (EMC) was not given. Only the environmental conditions of the chamber were reported. From the relationship of EMCtemperature-relative humidity given in the Wood Handbook (U.S. Department of Agriculture 1987), the EMC was predicted to be 7.4%. The above initial MC and final EMC were assumed to be the same for both the thick sample, with a half-thickness a = 14.31 mm, and the thin sample, with a = 4.79 mm. Uniform initial MC and equilibrium surface MC with the surroundings were assumed in order to initiate the calculation.

In this example, only the results of Cases 1 and 2 are reported. This is because no optimum β values, as defined in Eq. (5), were found in Cases 3 and 4. With each increase in β , the absolute NMSS always decreased, although only by a small amount. The assumption that the surface MC dropped immediately to equilibrium with the surroundings is valid because the final β values are large ($\beta > 1.5 \times 10^{-2} 1/$ sec). Since β represents the external resistance to the moisture movement, an increase in air speed results in an increase in β value. In the experiment, the air speed was relatively large (3.3 m/sec) and greater than the threshold value 3.0 m/sec (Rosen 1978) above which the external resistance is negligible.

The values of D and NMSS of Cases 1 and 2 are summarized in Table 1. To calculate the constant diffusion coefficient, a_1 in Eqs. 3 and 4 was set to zero with the boundary condition in Eq. (5) reduced to $m_s = m_e$ by setting β equal to ∞ . The optimum average diffusion coefficient with a minimum absolute value of the NMSS was searched. Its value for either the thick or the thin sample is practically the same as that obtained from the theoretical solution (Chen et al. 1995b), indicating that this numerical technique is accurate and reliable. To

| a the second of | TABLE 1. | Summary | of the | valuesa o | of D | and | NMSS | for ye | ellow-pop | lar sapwood.b |
|---|----------|---------|--------|-----------|--------|-----|------|--------|-----------|---------------|
|---|----------|---------|--------|-----------|--------|-----|------|--------|-----------|---------------|

| Sample type | Case | D (×10 ¹⁰ m ² /sec) | NMSS |
|----------------------------------|------|---|---------|
| Thick ($a = 14.31 \text{ mm}$) | 1 | 5.24 (5.25) ^c | -2.20N5 |
| | 2 | -2.476 + 0.6246(MC) | -2.45N6 |
| Thin $(a = 4.79 \text{ mm})$ | 1 | 4.26 (4.28) ^c | -3.54N5 |
| | 2 | -4.610 + 0.7082(MC) | -5.90N6 |

^a The notation xNy means $x \times 10^{-y}$.

^b Data used for the calculation are from Choong and Skaar (1969).
 ^c The value in the bracket is obtained from a theoretical approach (Chen et al. 1995b).

find the linear relationship between D and MC, the optimum values of both a_0 and a_1 in Eqs. (3) and (4) were searched.

As illustrated in Fig. 1, the application of the linear model enhances the predictability of drying curves significantly. Compared to that of the constant diffusion coefficient, the absolute value of the NMSS for either the thick or the thin sample decreases by one order of magnitude when the linear model is applied. This demonstrates that the diffusion coefficient is a function of the MC.

Sample thickness may affect the diffusion coefficient. The optimum constant diffusion coefficient for the thin sample is 18% smaller than that of the thick sample. As illustrated in Fig. 2, the linear functions have different intercepts and slopes for the thick and thin samples. The intercept of the thin sample (Table 1) is about twice that of the thick sample. Choong and Skaar (1969, 1972) reported a similar result, explaining that the discrepancy in the "apparent" diffusion coefficients was due to differences in the external resistances. The discrepancy in the present study may not be explained by the external resistance alone since the external resistance was found negligible. Other factors such as a time-dependent deformation of the cellulose-lignin matrix (Hart 1964) and drying stress could also be significant.

Example 2. Drying from above the fiber saturation point (FSP) to below the FSP

Data from Chen (1994) for redwood (Sequoia sempervirens) and red oak (Quercus sp.) heartwood were used to study the moisturedependent diffusion coefficient together with an exponential boundary condition. In these experiments, described in Chen et al. (1995a),



FIG. 1. Predicted drying curves derived from constant or linear diffusion coefficients as a function of moisture content.



FIG. 2. Comparison of constant diffusion coefficient and linear diffusion coefficient as a function of moisture content.



FIG. 3. Drying curves comparison of the four cases in redwood heartwood.

TABLE 2. Summary of the values^a of β , D, and NMSS for redwood heartwood and red oak heartwood^b (Half-thickness = 7.5 mm).

| Species | Case | β (×10 ⁴ 1/sec) | $\overset{\mathrm{D}}{(\times 10^{10}\mathrm{m}^2/\mathrm{sec})}$ | NMSS |
|---------|------|--|---|---------|
| Redwood | 1 | œ | 11.78 | -6.03N2 |
| | 2 | ∞ | 21.07 - 0.078(MC) | -5.69N2 |
| | 3 | 1.28 | 35.46 | -5.09N3 |
| | 4 | 1.13 | 68.18 - 0.178(MC) | -4.41N3 |
| Red oak | 1 | ∞ | 6.93 | -1.16N3 |
| | 2 | ∞ | 9.41 - 0.070(MC) | -1.05N3 |
| | 3 | 3.28 | 8.21 | -7.64N4 |
| | 4 | 2.56 | 6.82 + 0.051(MC) | -7.45N4 |

^a The notation xNy means $x \times 10^{-y}$.

^b Data used for the calculation are from Chen (1994).

the samples were dried in an AMINCO environmental chamber from near saturation to $6 \pm 1\%$ MC (45 ± 0.5 °C, 30% RH) with an air speed of 2.0 m/sec. The nominal half-thickness of the samples in the radial direction was a = 7.5 mm. Experimental data from three samples were used for each species. The initial and equilibrium MCs were determined from the average value of the three samples.

The predicted drying curves resulting from the optimization of the four cases are shown in Fig. 3 for redwood and in Fig. 4 for red oak. The results of the optimization search are summarized in Table 2. As expected, the best prediction with the smallest absolute values of the NMSS was achieved by the conditions as-



FIG. 4. Drying curves comparison of the four cases in red oak heartwood.

sumed in Case 4. However, almost the same accuracy could be achieved in Case 3. The relative magnitude of the NMSS values indicates that the enhancement of the prediction on drying curves is mainly due to using the exponential boundary condition instead of the equilibrium boundary condition. For instance, while the value of NMSS of Case 3 for redwood heartwood is 1.2 times larger than that of Case 4, the values of Cases 1 and 2 are 13.7 and 12.9 times larger, respectively. For red oak heartwood, the enhancement of the prediction is relatively small.

In this example, the diffusion coefficient can be considered a constant. Even though the calculated diffusion coefficient of Case 4 changes with MC, its use in the diffusion model does not result in significant improvement in the predictability of drying curves. The removal of moisture above the FSP is controlled by moisture diffusion at the wood surface. Based on the kinetic theory, Stamm (1964) explained that moisture diffuses through the cell wall by molecular jumps from one sorption site to another. When wood shrinks, the total number of sorption sites decreases; therefore, the diffusion coefficient decreases. However, a constant diffusion coefficient can prevail when the average moisture content is above the FSP. Figures 5 and 6 show the moisture content profiles at various times during the drying process for redwood and red oak, respectively. As



FIG. 5. Moisture content profile during drying in redwood heartwood.

drying progresses, the surface layers of the wood are the first to dry below the FSP, but are restrained from shrinking fully because the inner part of wood, which is still above the FSP, lags in shrinking. Consequently, the wood in the surface layers is stretched. Barkas (1949) reported an increase in EMC when wood is subjected to tension stress, which increases the sorption sites for bound water. The net effect of tension stress and shrinkage results in the number of sorption sites remaining about the same with the diffusion coefficient staying constant.



FIG. 6. Moisture content profile during drying in red oak heartwood.

CONCLUSIONS

The parameters in the linear function for D in an exponential boundary condition can be determined from drying curves using the optimization technique. Depending on MC range and drying conditions, the predictability of moisture removal during drying can be improved by using the moisture-dependent diffusion coefficient or the exponential boundary condition. Their relative importance depends on the initial MC of the wood and the drying condition.

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