MASS TRANSFER IN WOOD EVALUATED WITH A COLORIMETRIC TECHNIQUE AND NUMERICAL ANALYSIS

Hwanmyeong Yeo†
Research Associate

William B. Smith†
Professor
Faculty of Wood Products Engineering

and

Robert B. Hanna
Director
Center for Ultrastructure Studies
College of Environmental Science and Forestry
State University of New York
Syracuse, NY 13210

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ABSTRACT

A colorimetric $CoCl_2$-treated wood technique was used for determining surface moisture content of wood nondestructively and continuously during unsteady state desorption conditions. Utilizing these surface moisture data, diffusion and surface emission coefficients have been determined simultaneously and continuously during drying. The conversion method, which has been developed to facilitate making comparison between mass transfer coefficients in this study, has proven that the boundary layer theory is useful for evaluating the external moisture resistance during wood drying. The moisture profiles simulated by the finite difference method were quite comparable to the actual moisture profiles in real dried wood, which substantiates the high credibility of using the colorimetric technique for determining surface moisture content and mathematical procedure for determining the diffusion and surface emission coefficient.

Keywords: Mass transfer, diffusion, surface emission, boundary layer theory, finite difference method, colorimetric technique.

INTRODUCTION

Moisture movement in wood during drying is constrained by two resistances. The internal resistance due to wood itself can be described by the diffusion coefficient. Second is an external resistance to moisture leaving the wood surface and being transferred to the ambient drying air. This external resistance can be described by the mass transfer coefficient. Solutions of Fick's law and boundary layer theory have been widely applied to determine the diffusion and mass transfer coefficients, respectively (Crank 1975; Geankoplis 1993). However, a fully reliable method to determine these coefficients for a hygroscopic material simultaneously and continuously over the entire moisture content range has not yet been developed. A primary reason is the difficulty of determining wood surface moisture con-

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† Member of SWST.

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centration nondestructively and continuously in unsteady state drying conditions. This difficulty results in most mathematical approaches for determining the diffusion coefficient using an improper assumption that the surface moisture concentration is at equilibrium with the surrounding air as soon as the drying process starts. Because of this difficulty, theoretically calculated mass transfer coefficients using boundary layer theory have not been well proved by experimental results (Siau 1995).

Accurate and continuous measurement of surface moisture concentration in wood in unsteady-state drying conditions can increase credibility of methods for determining the coefficients. In this study, color change on wood surfaces treated with $CoCl_2$ according to humidity has been used for determining surface moisture content (SMC) nondestructively and continuously during drying. Utilizing surface moisture data, diffusion and mass transfer coefficients have been determined. Also, using these coefficients and numerical analysis with the finite difference method, a computer simulation program was developed to predict two-dimensional moisture profiles in wood during drying.

MATERIALS AND METHODS

Determination of diffusion and mass transfer coefficients

Completely water-soaked cubes of hard maple (*Acer saccharum* Marsh), red oak (*Quercus rubra* L.), and southern pine (*Pinus* spp.), with 20-mm longitudinal, radial, and tangential dimensions were manufactured. Four side walls of each cubic specimen were wrapped with rubber tape (Magic Wrap, Glasgow Mfg. Ltd.), leaving open the two opposite surfaces of interest, to provide one dimensional moisture flow for drying. Six replicates, to determine the mass transfer coefficient based on moisture concentration at wood surface, also called the surface emission coefficient, and the diffusion coefficient in each orientation, longitudinal, radial, and tangential, were prepared.

Surface and average moisture content.—Using the recently developed colorimetric technique (Yeo et al. 2002) and oven-drying, surface moisture content (SMC) and average moisture content (AMC) changes in wood were determined, respectively, during drying at 25% RH and both 30 and 50°C in temperature- and humidity-controlled environmental chambers. Air velocity in these chambers was 1 m/s, measured with a hot wire anemometer, and airflow was parallel to exposed wood surfaces. Saturated $CoCl_2$ solution, approximately 5 μL, was dropped on the exposed surface area of three of those six replicates for each orientation, and spread with the tip of a dispensing micropipette. The surface spectral reflectance was measured with a portable spectrophotometer (Microflash 200d, manufactured by DataColor International) during drying at 25% RH and both 30 and 50°C, and then CIE L*a*b* color values were calculated on the basis of CIE (Commission Internationale de l'Eclairage) standards (CIE 1986). Using surface color differences (dE_{mc-eq}) between moist wood and wood equilibrated at 25% RH, surface moisture content of each species were determined.

$$dE_{mc-eq} = [(L_{mc}^* - L_{eq}^*)^2 + (a_{mc}^* - a_{eq}^*)^2 + (b_{mc}^* - b_{eq}^*)^2]^{1/2} \quad (1)$$

where

L^*_{mc}, a^*_{mc}, b^*_{mc}:

L*, a*, and b* values on the $CoCl_2$-treated surfaces equilibrated at any %RH

L^*_{eq}, a^*_{eq}, b^*_{eq}:

L*, a*, and b* values on the $CoCl_2$-treated surfaces equilibrated at 25% RH

Also, three untreated specimens were used as controls to prove the drying rate of treated wood to be similar to that of untreated wood.

Specific gravity.—Specific gravities of moist wood (G_{MC}) were not measured directly during drying because of the excessive time required to make many measurements. This likely would have decreased the efficiency of the method for determining the diffusion and surface emission coefficients. Instead of direct measurement, G_{MC} was approximated by linear interpolation between oven-dry specific gravity and green specific gravity.

Diffusion coefficient.—A new analytical solution was derived to determine the diffusion coefficient when surface moisture contents were varied. The experimentally measured SMC, AMC, the half thickness of specimen (L), drying time (t), initial moisture content in each drying time interval (IMC_i), and specific gravity of moist wood were used for determining the three directional diffusion coefficients. The analytical solution of the unsteady state diffusion equation was obtained by separation of variables for the constant diffusion coefficient case with the following initial and boundary conditions.

Initial condition:

$C = C(x, 0) \quad \text{at } x = x \quad \text{and} \quad t = 0$

1st boundary condition:

$C = C(0, t) = Cs(t) \quad \text{at } x = 0 \quad \text{and} \quad t = t$

2nd boundary condition:

$C = C(2L, t) = Cs(t) \quad \text{at } x = 2L \quad \text{and} \quad t = t$

where C = moisture concentration, kg/m^3, C(x, t) = concentration in any location and at any time, kg/m^3, Cs(t) = surface moisture concentration at any time, kg/m^3, L = half thickness of specimen, m, t = drying time, s, x = a space of the geometry, m.

Assumptions used in deriving the solution are that dimensional change in the moisture movement direction does not occur, and that the diffusion coefficient is constant in each drying time interval. By redefining the fractional change in moisture concentration (E) as $E = [C(x, t) - Cs(t)]/[C(x, 0) - Cs(t)]$, the unsteady-state diffusion equation $\partial C/\partial t = D \cdot \partial^2 C/\partial x^2$ can be expressed as $\partial E/\partial t = D \cdot \partial^2 E/\partial x^2$. The solution can be obtained by the separation of variables method.

$$E = \frac{C(x, t) - Cs(t)}{C(x, 0) - Cs(t)} = \sum_{n=0}^{\infty} \frac{4}{(2n+1)\cdot\pi} \cdot \sin\left(\frac{(2n+1)\pi}{2L} \cdot x\right) \times \exp\left(-\frac{(2n+1)^2\pi^2}{(2L)^2} Dt\right). \tag{2}$$

By integration of Eq. (2) over the half thickness from 0 to L, the fractional change in the average moisture concentration ($\bar{E}$) becomes

$$\bar{E} = \frac{C_{AVG}(t) - Cs(t)}{C_{IMC_i} - Cs(t)} = \frac{8}{\pi^2} \cdot \sum_{n=0}^{\infty} \frac{1}{(2n+1)^2} \cdot \exp\left(-\frac{(2n+1)^2\pi^2}{(2L)^2} Dt\right),$$

where $C_{AVG}(t)$ = average moisture concentration in wood, kg/m^3, and C_{IMCi} = initial moisture concentration in wood in each drying time interval, kg/m^3. Because water concentration in wood can be presented by $C(x, t) = G_{MC} \cdot \rho_w \cdot MC(x, t)/100$, where G_{MC} = specific gravity of moist wood, ρ_w = water density, kg/m^3, and MC = %moisture content, the diffusion coefficient (D) in each drying time interval can be calculated as follows:

$$D = -\frac{4 \cdot L^2}{\pi^2} \cdot \left(\frac{G_I IMC_i - G_S SMC(t)}{G_A AMC(t) - G_S SMC(t)}\right) \times \left(\frac{1}{G_I IMC_i - G_S SMC(t)} \cdot \frac{dG_A AMC(t)}{dt} + \frac{(-G_I IMC_i + G_A AMC(t))}{(G_I IMC_i - G_I SMC(t))^2} \times \frac{dG_S SMC(t)}{dt}\right) \tag{3}$$

where IMC_i = %initial moisture content in each drying time interval, SMC and AMC = %surface and %average moisture content, G_I, G_S, and G_A = specific gravity of wood at IMC_i, SMC, and AMC.

Mass transfer coefficients.—Mass transfer coefficient can be expressed in several ways, depending upon the potential of the gradient.

Surface emission coefficient.—The surface emission coefficients, mass transfer coefficient based on moisture concentration in wood, were experimentally determined by

$$S_{exp} = \frac{J}{(C_{s,wood} - C_{e,wood})} = \frac{\Delta W/(A\cdot\Delta t)}{G_S\cdot\rho_w\cdot SMC/100 - G_E\cdot\rho_w\cdot EMC/100} \qquad (4)$$

where S_{exp} = experimentally determined surface emission coefficient, m/s, J = moisture flux from surface to air, kg/s m^2, $C_{s,wood}$ = moisture concentration at wood surface, kg/m^3, $C_{e,wood}$ = moisture concentration in wood at equilibrium with surrounding condition, kg/m^3, W = mass of evaporated moisture, kg, A = surface area of specimen, m^2, EMC = %equilibrium moisture content, and G_E = specific gravity of wood at EMC. The S_{exp} were compared with mass transfer coefficients calculated by boundary layer theory (h_{air}).

Mass transfer coefficient calculated by boundary layer theory (h_{air}).—

$$h_{air} = \frac{0.66\cdot D_{H_2O,air}Re^{1/2}Sc^{1/3}}{L_s} \qquad (5)$$

(for laminar flow, Geankoplis 1993; Siau 1995) where h_{air} = mass transfer coefficient based on moisture concentration in air, m/s, $D_{H_2O,air}$ = diffusion coefficient of water vapor in air = $2.2\cdot 10^{-5}(1.013\cdot 10^5/P)\cdot(T/273)^{1.75}$, m^2/s, P = total pressure of air and water vapor, Pa, T = temperature, K, Re = Reynolds number = $(L_s\cdot v\cdot\rho_a)/\mu$, Sc = Schmidt numbers = $\mu/(\rho_a D_{H_2O,air})$, L_s = length of surface along which convection occurs, m, v = air velocity, m/s, ρ_a = density of air, kg/m^3, and μ = dynamic viscosity of air, Pa·s.

Conversion between mass transfer coefficients.—Because S_{exp} is based on the moisture concentration in wood, and h_{air} is based on the moisture concentration in air, a conversion method from h_{air} to surface emission coefficient is required to facilitate making comparisons to each other. Siau (1995) believed that water vapor pressure on the surface of wood (p_s) in the hygroscopic range could be determined by multiplying the saturated vapor pressure of water at dry bulb temperature ($p_{o,dry}$) by relative humidity at the surface (RH_s). This calculation, however, resulted in large deviations between experimental and theoretical surface emission coefficients. Siau's method overestimates the p_s because when very wet wood surfaces are being dried to the fiber saturation point (FSP), the p_s is close to the saturated vapor pressure at wet bulb temperature ($p_{o,wet}$) not $p_{o,dry}$. Correspondingly, when the surface is being dried from FSP to EMC in the hygroscopic range, the p_s might decrease from $p_{o,wet}$ to vapor pressure equilibrated to ambient air (p_e), which can be determined by multiplying $p_{o,dry}$ by environmental relative humidity (RH_e). Using this concept, a new conversion method between mass transfer coefficients has been developed (Yeo 2001). p_s can be determined, using the relationship of $(p_{o,wet} - p_s)/(p_{o,wet} - p_e) = (100 - RH_s)/(100 - RH_e)$, as

$$p_s = p_{o,wet} - \left(\frac{p_{o,wet} - p_e}{100 - RH_e}\right)\cdot(100 - RH_s) = p_{o,wet} - \left(\frac{p_{o,wet} - p_{o,dry}\cdot\left(\frac{RH_e}{100}\right)}{100 - RH_e}\right) \times (100 - RH_s) \qquad (6)$$

where p_s and p_e = water vapor pressure in air adjacent to surface of wood and in ambient air, Pa, $p_{o,wet}$ and $p_{o,dry}$ = saturated water vapor pressure at wet bulb and at dry bulb temperature, Pa, and RH_s and RH_e = %relative humidity of air adjacent to surface of wood and of ambient air.

Moisture concentration in air adjacent to a wood surface ($C_{s,air}$) and moisture concentration in ambient air ($C_{e,air}$) can be calculated by the gas law. $C_{s,air} = (M_{H_2O}\cdot p_s)/(R\cdot T_s)$, and $C_{e,air} = (M_{H_2O}\cdot p_e)/(R\cdot T_{dry})$, where M_{H_2O} = water molecular weight, 18 kg/kgmol, R = gas law constant, 8,314 m^{3}Pa/kgmol K, and T_s and T_{dry} =

temperature in air adjacent to surface and in ambient air, K. Finally, surface emission coefficient can be theoretically converted from h_{air} by,

$$S_{theo} = h_{air} \cdot \frac{(C_{s,air} - C_{e,air})}{(C_{s,wood} - C_{e,wood})} = h_{air} \cdot \frac{\dfrac{M_{H_2O} \cdot p_s}{R \cdot T_s} - \dfrac{M_{H_2O} \cdot p_e}{R \cdot T_{dry}}}{G_S \cdot \rho_W \cdot \dfrac{SMC}{100} - G_E \cdot \rho_W \cdot \dfrac{EMC}{100}}$$

$$= h_{air} \cdot \frac{\dfrac{M_{H_2O}}{R \cdot T_s}\left(p_{o,wet} - \left(\dfrac{p_{o,wet} - p_{o,dry} \cdot \left(\dfrac{RH_e}{100}\right)}{100 - RH_e}\right) \cdot (100 - RH_s)\right) - \dfrac{M_{H_2O}}{R \cdot T_{dry}}\left(p_{o,dry} \cdot \left(\dfrac{RH_e}{100}\right)\right)}{G_S \cdot \rho_W \cdot \dfrac{SMC}{100} - G_E \cdot \rho_W \cdot \dfrac{EMC}{100}} \quad (7)$$

Moisture content profiles in wood

Actual MC profiles.—Fully saturated maple, oak, and pine specimens with 200 (longitudinal) × 50 (radial) × 50 (tangential)-mm dimensions were wrapped in plastic at 30 and 50°C conditions prior to drying. Also, prior to each drying experiment, the specimens were end coated with a fibered asphalt compound (St. Louis Paint Mfg. Co.) to provide two-dimensional only, radial and tangential, moisture flow. MC profiles in each specimen were measured by the slicing method, with 7 sections each in the radial and tangential directions cut and MCs of those sections determined by oven-drying.

Simulated MC profiles.—Two-dimensional MC profiles in wood during drying were simulated by the finite difference method (FDM). The computer simulation program utilized DELPHI updated PASCAL program language. Moisture concentration equations in this program were solved by implicit formulation and Gauss-Seidel iteration.

RESULTS

Colorimetric technique

Surfaces of $CoCl_2$-treated wood exhibit a light yellow-red color at high MC, which changes to a dark green-blue color at low MC. Spectral reflectance from the surfaces was measured with the spectrophotometer. Surfaces of treated wood at higher MC reflected particularly well longer wavelength light. Using reflectance data and wood hygroscopicity, color value change on the $CoCl_2$-treated moist wood surface could be plotted in CIE L*a*b* color space. L*, a* and b* values continuously decrease over the high to low MC range.

Surface and average MC and specific gravity

Using the colorimetric technique and oven drying, SMC and AMC were determined, respectively, during drying at 25% RH and both 30 and 50°C (Fig. 1). Green specific gravity of hard maple, red oak, and southern pine were 0.47, 0.56, and 0.44, and their oven-dry specific gravities were 0.53, 0.64, and 0.50, respectively. Specific gravity of moist wood that was approximated by linear interpolation between green and oven-dry specific gravity has been used to determine diffusion and surface emission coefficients. From Fig. 1, it appears that faster surface drying takes place in the tangential and radial direction than longitudinal. This seems to be because of low permeability in the tangential orientation. The continuity of liquid water in the tangential direction was more likely to be broken at the very beginning of the drying process. This may make it look as though faster drying takes place in the tangential direction. But broken

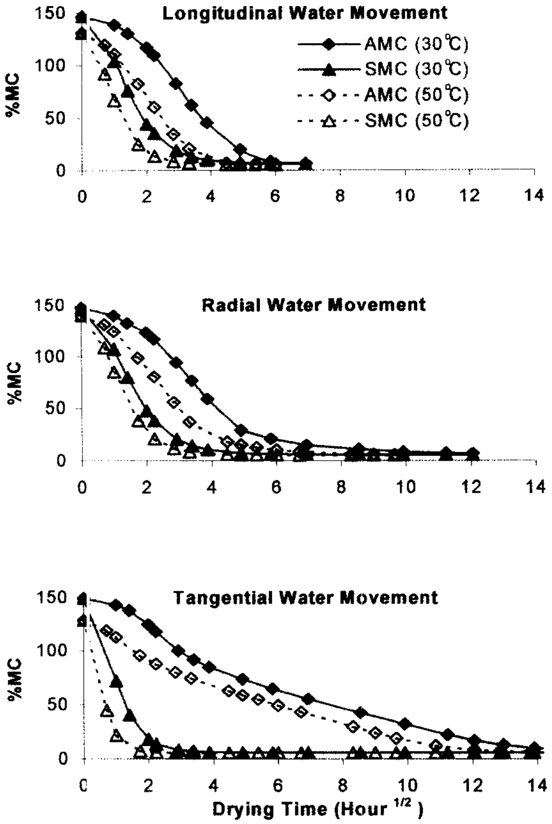

FIG. 1. Average and surface moisture contents of hard maple whose moisture movement is constrained to either longitudinal, radial, or tangential directions during drying at 25% RH and 30°C or 50°C.

liquid water continuity at the shell makes internal water movement from the core to surface slower. This can be seen by the slower drying AMC values.

Diffusion coefficient

During drying at 30°C and 25% RH, the diffusion coefficients in the longitudinal direction of each species remained constant in the range from maximum MC to 30% AMC, and then decreased to EMC (Fig. 2). These results might indicate that continuity of liquid water exists in the longitudinal direction from initial moisture content to 30% AMC, and that the longitudinal movement of moisture in that range is by capillary action. The diffusion coefficient in the transverse direction for each species, except the radial direction with hard maple, decreased from over 100% AMC to EMC. This could indicate that continuity of liquid water in the radial direction of red oak and southern pine and the tangential direction of each species was broken at the very beginning of the drying process. Major resistance to water movement in this case might be evaporation and condensation of moisture in wood and so governed by water vapor pressure and bound water gradients. The diffusion coefficient in the radial direction of hard maple remained constant to about 60% AMC, and then decreased to EMC. In this case continuity of liquid water exists in the radial direction to about 60% AMC. This result might be due to the anatomical structure of hard maple where ray cells may support continuous free water movement relatively well in the radial direction. Over the entire AMC range, diffusion coefficients decreased during drying, which means that internal resistance increased. Although the data are not presented, diffusion coefficients determined during drying at 50°C and 25% RH condition were twice as high as those determined during drying at 30°C and 25% RH.

Mass transfer coefficient

Experimentally determined surface emission coefficients (S_{exp}).—Relationships between S_{exp} and SMC of maple, oak, and pine were observed while drying at 30°C and 50°C with 25% RH (Fig. 3). As drying progressed, the coefficients slightly increased when above the FSP range and then rapidly increased from FSP to EMC. Also, a positive effect of temperature on the surface emission coefficient was found, as the coefficient values at 50°C were greater than those at 30°C. These results show that surface emission coefficients were strongly dependent upon the SMC and air temperature as well as air velocity, which is known as a critical factor.

Mass transfer coefficient calculated by boundary layer theory (h_{air}).—After the physical properties of air, such as velocity, density,

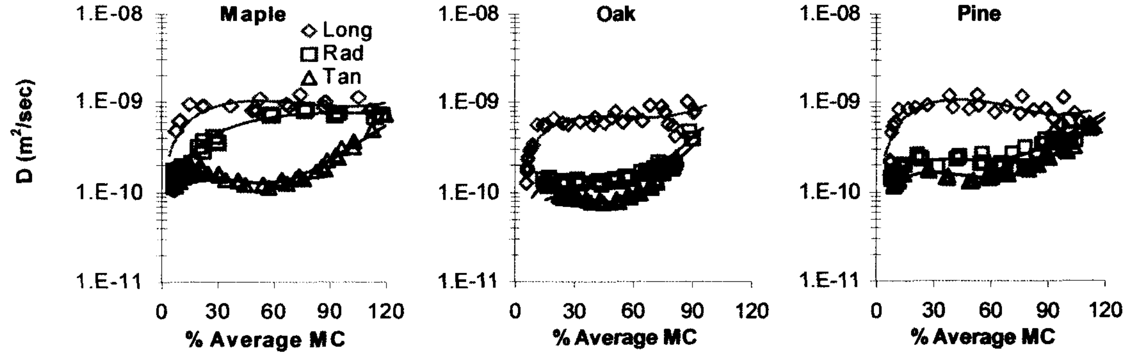

FIG. 2. Diffusion coefficients in three orientations for maple, oak, and southern pine at 30°C.

and viscosity, were measured and surveyed, mass transfer coefficients were calculated by boundary layer theory (h_{air}). Air velocities in the environmental chambers were 1 m/s. The surface lengths of specimens along which convection occurs were 0.02 m. Air densities at 30°C (303K) and 50°C (323K) were 1.167 and 1.096 kg/m^3, respectively. Air viscosities at 30°C and 50°C were 1.87×10^{-5} and 1.96×10^{-5} Pa·s, respectively. Based on those data, diffusion coefficients of water vapor in air ($D_{H_2O,air}$, 2.64×10^{-5} m^2/s at 30°C and 2.95×10^{-5} m^2/s at 50°C), Reynolds number (Re, 1,248 at 30°C and 1,118 at 50°C) and Schmidt number (Sc, 0.6069 at 30°C and 0.6062 at 50°C) were determined. Using those values, mass transfer coefficients (h_{air}) were determined to be 0.026 and 0.028 m/s at 30 and 50°C, respectively. Because the $D_{H_2O,air}$ used to calculate the h_{air} are constant coefficients for interdiffusion of water vapor in bulk air, and the Re and Sc numbers are also constant, the h_{air} values become constant. This helps explain the external moisture-evaporating rate from very wet wood surfaces and might lead one to conclude that the external resistance is constant at very high SMC condition. However, because there is no consideration of water vapor pressure change with surface moisture content in the hygroscopic range, the use of constant h_{air} is not good for explaining the resistance of moisture-evaporating rate from the surfaces when SMC is low, especially below FSP.

The previously developed mass transfer coefficient conversion method (Eq. 7) considers water vapor pressure change with surface moisture content in the hygroscopic range. To determine whether boundary layer theory is useful for evaluating the external moisture re-

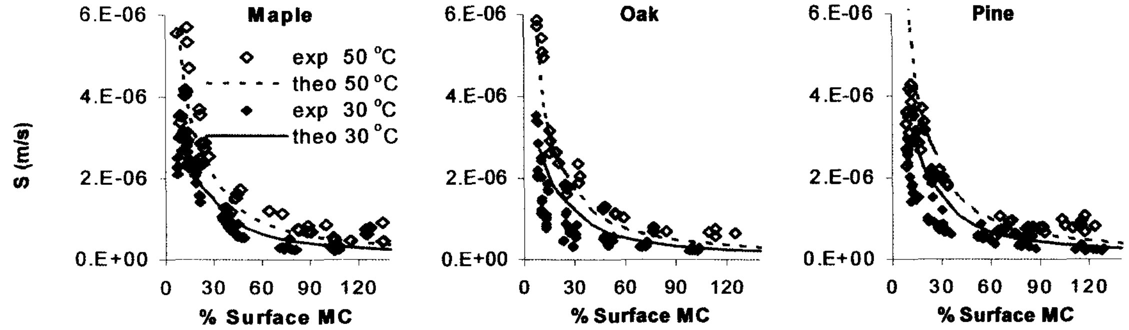

FIG. 3. Experimentally determined surface emission coefficient (S_{exp}) and surface emission coefficient theoretically converted from mass transfer coefficient calculated by boundary layer theory (S_{theo}).

sistance during wood drying or not, the S_{theo} converted from the h_{air} with the mass transfer coefficient conversion method has been compared with S_{exp}.

Surface emission coefficient theoretically converted from h_{air} (S_{theo}).—For example, with hard maple dried at 30°C and 25% RH, the wet bulb temperature was 16.8°C (290K); 140%, 29%, and 6.2% MC were used for initial surface moisture content, fiber saturation point, and equilibrium moisture content; 0.47 and 0.53 were used for green and oven-dry specific gravity;, and 1,923 Pa ($p_{o,wet}$) and 4,249 Pa ($p_{o,dry}$) were used for the saturated vapor pressure of water in air at wet bulb temperature 16.8°C and dry bulb temperature 30°C, respectively. Since RH_e was 25% RH, water vapor pressure in ambient air (p_e) was 1,062 Pa = 4,249·(25/100). Using those values, h_{air} were converted to surface emission coefficients. These theoretically converted surface emission coefficients (S_{theo}) were quite close to S_{exp} (Fig. 3). This proves that boundary layer theory with conversion method is useful for evaluating the external moisture resistance during wood drying.

Relationships between surface emission coefficient and SMC have been rarely observed in the past. While some papers have addressed the relationship between surface emission coefficient and AMC, it is doubtful whether AMC has a critical effect on external resistance, which includes surface resistance. Yeo has compared some experimentally determined constant surface emission coefficients in certain MC ranges, by Choong and Skaar (1969 and 1972), with theoretically converted surface emission coefficients (Yeo 2001). Results of this comparison have shown that surface emission coefficient can be determined theoretically with boundary layer theory and appropriate conversion method. More comparisons like this might be performed in further studies.

MC profiles in wood

Overall, the moisture profiles simulated by finite difference method were quite comparable to the actual moisture profiles in real dried wood. Results for maple at 30°C are illustrated in Fig. 4. This substantiates the high credibility of using the colorimetric technique for determining surface moisture content and mathematical procedure for determining the diffusion and surface emission coefficient.

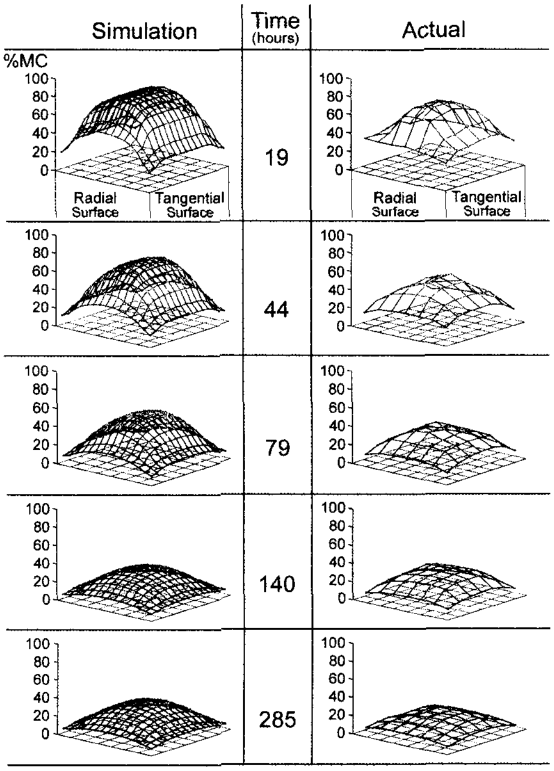

FIG. 4. Simulated and actual MC profiles in hard maple during drying at 30°C and 25% RH.

CONCLUSIONS

Use of the colorimetric $CoCl_2$-treated wood technique for determining surface moisture content of wood nondestructively and continuously in unsteady state drying conditions could increase the credibility of methods for calculating the internal and external resistances for moisture movement in, and surface evaporation from, wood.

Experimental results showed that diffusion coefficients were dependent upon the temperature and average moisture content in wood.

Also, the surface emission coefficients were shown to strongly depend on the surface moisture content of wood and air temperature as well as air velocity, which has been known as a critical factor.

The mass transfer coefficient conversion method developed using surface moisture data in this study has proven that the boundary layer theory is useful for evaluating the external resistance during wood drying. Because of this proof, the external resistance for wooden surfaces can be theoretically described in dynamic drying situations.

The moisture profiles simulated by finite difference method were quite comparable to the actual moisture profiles in real dried wood, which substantiates the credibility of using the colorimetric technique for determining surface moisture content and mathematical procedure for determining the diffusion and surface emission coefficient.

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