

DYNAMIC SORPTION AND HYGROEXPANSION OF WOOD SUBJECTED TO CYCLIC RELATIVE HUMIDITY CHANGES

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(Received November 2009)

Abstract. To investigate the behavior of sorption and hygroexpansion of wood at nonequilibrium, Sitka spruce (*Picea sitchensis* Carr.), 4-mm along the grain and 20-mm in radial and tangential directions, was exposed to sinusoidally RH between 45-75% at 20°C for 1, 6, and 24 h. Moisture changes and radial and tangential dimensional changes measured during the cycling gave the following results: moisture and dimensional changes of the specimens were generally sinusoidal but lagged behind the imposed RH. The phase lag decreased and amplitude increased with increasing cyclic periods. Furthermore, a mathematical model proposed in a previous study was modified to describe the dynamic sorption of wood exposed to cyclical RH. The model not only produced kinetics in good agreement with experiment results, but also can provide moisture gradient distributions developed throughout the wood during cyclical processes.

Keywords: Cyclical relative humidity, dynamic sorption, hygroexpansion, moisture gradient distribution, nonequilibrium state.

INTRODUCTION

Wood is continually changing in moisture content (MC) both during the initial drying from the green condition and subsequent use. The latter changes are because of changes in atmospheric RH and temperature, which may be cyclic or

sporadic. These moisture variations inevitably produce dimensional changes in wood. The initial shrinkage is certainly an important consideration in manufacturing. Even more important, however, are the changes in dimensions that accompany the usual fluctuations of RH and temperature after wood has been placed in service (Ma and Zhao 2006). This behavior has been termed “movement” by Stevens (1963) to distinguish it from

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the initial shrinkage of wood. Movement is considerably smaller for a given range of RH change and cannot be predicted from the initial shrinkage (Skaar 1988). It is a useful index to the use of wood under the range of atmospheric conditions that affect it in service.

However, studies of sorption and hygroexpansion of wood under cyclic environmental conditions have not received enough attention. Some typical studies have implications for the present study and therefore are briefly mentioned here. Harris (1961) exposed hardwood and softwood specimens to equilibrium with three consecutive cycles between 65-95% RH at 26°C. At the beginning and end of each 24-h cycle, moisture contents and tangential dimensions were recorded. Wu and Ren (2000) investigated sorption behavior of oriented strandboard under long-term cyclic RH conditions. The EMC-RH data were fit to Nelson's sorption model through a nonlinear regression technique, and the model parameters were determined from the first sorption cycle. The study showed that isotherms calculated by the model and parameters from the initial exposure cycle are reproducible under cyclic moisture conditions. García Esteban et al (2005) explored the effect of cyclic moisture treatments on wood. They subjected wood specimens to five wet-dry cycles and measured MC and dimensional changes in radial and tangential directions at three increasing levels of RH before and after the treatment. It was found that repeated RH cycles will cause a reduction in wood hygroscopicity and associated dimensional response. Furthermore, the linear relationship usually observed in normal wood between MC and dimensional change is no longer present with the cycled wood. These studies have limitations because their measurements were carried out either with a moisture equilibrium state or before and after the cycling rather than a full cyclical test.

Chomcharn and Skaar (1983) subjected roundwood wafers of green basswood, yellow birch, and black cherry to sinusoidally varying RH between 77-47% at 25°C for many cycles at each of four different cyclic periods. Moisture and radial and tangential dimensional changes

were measured during the cyclic processes. Schmidt numerical solution for moisture diffusion with a constant diffusion coefficient was used to simulate the average normalized MC. The resulting curves can be considered at most as qualitatively similar to those obtained experimentally, especially for the data before steady state at the shortest cyclic period. Droin-Josserand et al (1988) proposed a mathematical model based on numerical methods with a concentration-dependent diffusivity. The model can not only describe the sorption process of wood when RH varies at a constant rate, but also attains the profiles of concentration of moisture within the thickness of wood. However, the study tested only one cycle of moisture changes and poor agreement was found between the experiment and the model for desorption. Lu and Leicester (1997) combined Fick's law for predicting the MC with a simple mechanosorptive model to analyze creep resulting from cyclic moisture variations represented by daily and annual sine waves. Nevertheless, the boundary condition assumed was the EMC for the temperature and RH of the air around the wood. This is not practical because of the time delay in response of the surface of wood to varying environmental conditions. Time (2002) made a basic exploration on hygroscopic moisture sorption by exposing wood to cyclical RH. Jönsson (2004) conducted an experimental investigation on moisture and strain distributions in the cross-grain direction in glulam induced by cyclic climate change, which provides experimental data for the comparison with theoretical analysis on moisture distributions under the same condition. Ma et al (2005) made a test similar to Chomcharn and Skaar (1983) with the RH varied triangularly. The same general behavior in moisture and dimensional responses of wood was found.

Because there has not been adequate experimental data and theoretical models from the previous research, the work reported here was conducted to 1) investigate moisture and transverse dimensional responses of wood to cyclically varying RH; and 2) develop a model to reproduce moisture sorption in wood exposed to cyclic RH changes, from which moisture gradient distributions can be

determined as well during cycling. The results should be helpful in providing a fundamental understanding on the sorptive and hygroexpansive behavior of wood at nonequilibrium, minimizing the problems associated with shrinking and swelling of wood products in service and improving environmental variation properties of wood.

MATERIALS AND METHODS

Experimental

Sitka spruce (*Picea sitchensis* Carr.) was chosen as the species. The specimens, 4 mm along the grain and 20 mm in both radial and tangential directions, were initially oven-dried at 105°C. After their oven-dry weights were measured, they were conditioned at 45% RH controlled by saturated salt solution of potassium carbonate (Macromolecule Academy) at $20 \pm 0.2^\circ\text{C}$. They were then moved into an RH conditioning chamber where temperature was kept at 20°C throughout the experiment.

Before the cyclic tests, the specimens were initially re-exposed to $45 \pm 1.0\%$ RH for 24 h to further equilibrate at this condition, after which they were subjected to sinusoidally varying conditions of $45\text{--}75 \pm 1.0\%$ RH for periods of 1, 6, and 24 h. The RH in the chamber was programmed to vary in discrete steps according to a predetermined schedule, and a temperature

and RH transmitter was placed at the test specimens to ensure the desired conditions. Weight changes as well as radial and tangential dimensional changes in response to the imposed RH were recorded by an electronic balance and three CCD laser displacement sensors (Fig 1). The precision of measurement was 0.1 mg and 1 μm for weight and dimension, respectively.

In addition, every group had three end-matched samples for each cycle: one for dimension measurement and the other two for weight measurement. The dimension and weight measurements were repeated three times. The dimension measurement samples were rotated so that each of the three samples was measured for dimension once and weight twice. In this way, the effect of variability among specimens could be reduced considerably. The average values of the three tests for weights and dimensions of the specimens were taken as the final result.

Fourier Analysis

Experimental results were analyzed by the Fourier approach that was introduced by Chomcham and Skaar (1983) on their dynamic sorption and shrinkage data. It is an effective method in which important parameters can be determined. These parameters are also helpful in investigating the properties of the observed results, for

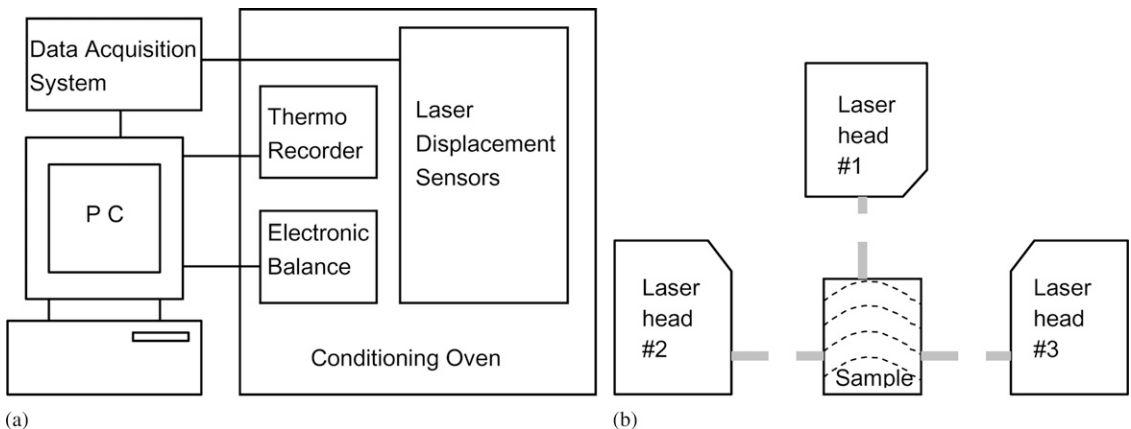


Figure 1. Diagram showing the instrumentation for (a) the entire assembly and (b) detection of dimensional changes in the sample, in which laser head 1 is for radial measurement and laser heads 2 and 3 are for tangential.

example, the phase lag and amplitude of moisture and dimensional changes.

First, the following equation was used to fit all observed curves.

$$G(t) = \frac{G(t)_{\max} + G(t)_{\min}}{2} + \frac{(G(t)_{\max} - G(t)_{\min})}{2} \times [A_1 \sin \omega t + B_1 \cos \omega t] \quad (1)$$

where $G(t)$ is the observed value of RH, MC, or radial or tangential dimensional changes. A_1 and B_1 are the Fourier coefficients for the fundamental angular frequency ω . $G(t)_{\max}$ and $G(t)_{\min}$ are the peak values of $G(t)$ at time t obtained by fitting a six-power polynomial to the observed maximum and minimum peaks.

Rearranging Eq 1:

$$A_1 \sin \omega t + B_1 \cos \omega t = \frac{2G(t) - G(t)_{\max} - G(t)_{\min}}{G(t)_{\max} - G(t)_{\min}} = Q(t) \quad (2)$$

where $Q(t)$ is used to represent the complex expression involving the $G(t)$ terms. By Fourier analysis, A_1 and B_1 can be evaluated as

$$A_1 = (2/T_0) \int_0^{T_0} Q(t) \sin(\omega t) dt \approx (2/T_0) \sum_{i=1}^N Q_i(t) \sin(\omega t) \Delta t \quad (3)$$

$$B_1 = (2/T_0) \int_0^{T_0} Q(t) \cos(\omega t) dt \approx (2/T_0) \sum_{i=1}^N Q_i(t) \cos(\omega t) \Delta t \quad (4)$$

where T_0 is the cyclic period and N is the number of discrete, equally spaced time steps at which the observed data were recorded for each complete cycle.

The phase angle ϕ (radians) was then calculated for RH, MC, and radial and tangential dimensional changes for each cycle by:

$$\phi = \arctan(B_1/A_1) \quad (5)$$

Then, the phase lag φ (radians) of each response curve can be obtained from the difference in the phase angle ϕ between each response cycle and the corresponding RH cycle.

$$\varphi_{MC} = \phi_{MC} - \phi_{RH}, \quad \varphi_R = \phi_R - \phi_{RH}, \quad \varphi_T = \phi_T - \phi_{RH} \quad (6)$$

where the subscripts RH , MC , R , and T correspond to RH, MC, radial, and tangential dimensional changes, respectively.

In addition, the amplitude A of each response curves is defined in this study as

$$A = \frac{1}{n} \sum_{i=1}^n (G_i(t)_{\max} - G_i(t)_{\min}) \quad (7)$$

where n is the number of discrete, equally spaced time steps at which the observed data were recorded for each whole test.

Theoretical Model for Dynamic Sorption

In a previous study (Ma et al 2009), a mathematical model was developed for moisture sorption by wood in an atmosphere of constant RH (static sorption). In this work, the model was modified and tested for dynamic sorption in cyclically varying RH conditions.

Both surface moisture exchange through the air-wood interface and internal diffusion within wood were considered in the model. The moisture exchange on the wood surface is given by

$$m_i = m_{i-1} + a(h - \exp(K_2 K_1^{m_{i-1}} + K_3)) \Delta t$$

$$K_1 = 1.0327 - 0.000674T$$

$$K_2 = 17.884 - 0.1432T + 0.0002363T^2$$

$$K_3 = 0.16 \quad (8)$$

where m is MC (%), a is sorption rate constant (1/h), h is RH for the surrounding atmosphere (%), and T is temperature (K). K_1 , K_2 , and K_3 are given by the Bradley equation (Bramhall 1979),

and their values were validated by experimental isotherm curves.

For the transfer of moisture inside the wood, a numerical solution by the finite difference method (Nakao 2002) for the Fick's second law was used.

$$m_{ji} = \frac{D(m_{j-1} - 2m_j + m_{j+1})_{i-1}}{\Delta l^2} + (m_j)_{i-1} \quad (9)$$

where D is the moisture diffusion coefficient (m^2/s), the subscripts $i-1$, i are ordinal time points, $j-1$, j , $j+1$ are ordinal elements in thickness direction, and Δl is the thickness of each element (m). The value of D was taken as a constant in the previous study. However, in the present case, it is considered to be MC-dependent and expressed by an exponential function for Sitka spruce in longitudinal direction (Skaar 1988):

$$D = 1.44 \times 10^{-8} \exp(0.11m) \quad (10)$$

RESULTS AND DISCUSSION

General Moisture and Dimensional Responses

Moisture and dimensional responses of the specimens to sinusoidally varying RH cycled at 24 h, as an example, is shown in Fig 2 in which dimensional changes are given in terms of swelling based on oven-dry dimensions. It is apparent that the moisture and dimensional changes are generally sinusoidal but lag behind the imposed RH. It also seems as if a repetitive “steady state” in the moisture and dimensional responses is reached from the third cycle.

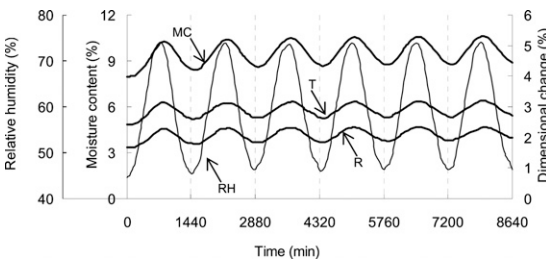


Figure 2. Plots of RH, moisture content (MC), and radial (R) and tangential (T) dimensional changes against cyclic time for Sitka spruce cycled 24 h.

Phase Lag and Amplitude

Figure 3 gives the observed and fitted RH, MC, and radial and tangential dimensional curves calculated by the Fourier analysis for the specimens cycled at 24 h. As shown in this figure, the agreements between observed and fitted curves in all cases are generally quite good. This indicates that the Fourier coefficients A_1 and B_1 and the terms $G(t)_{\max}$ and $G(t)_{\min}$ can be used to investigate the phase lag and amplitude of the experimental data by using Eqs 5-7.

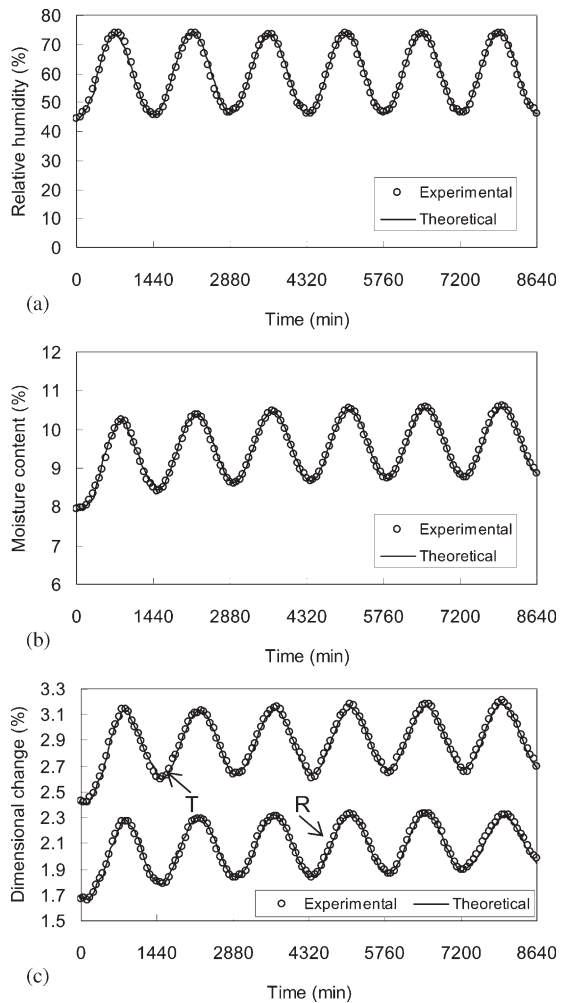


Figure 3. Plots of observed and fitted (a) RH, (b) moisture content (MC), and (c) radial and tangential dimensional changes against cyclic time for Sitka spruce cycled 24 h.

Figure 4 shows the average phase lag of moisture, radial, and tangential responses against numbers of cycles for all three different cyclic periods. It is clear that as the cyclic period increases, the phase lag decreases. This is as expected because with longer cyclic periods, the specimen response could more nearly follow the RH changes. Moreover, the phase lag decreases with increasing number of cycles.

Table 1 lists the phase lags of MC and radial and tangential dimensional responses for each of the three cyclic periods. It is apparent that the phase lag for the moisture change is greater than that for the dimensional changes at a given cyclic period, which was also the case for Chomcham and Skaar (1983). This suggests that dimensional changes are faster than moisture changes at the nonequilibrium state. No satisfactory explanation could be given in the present work.

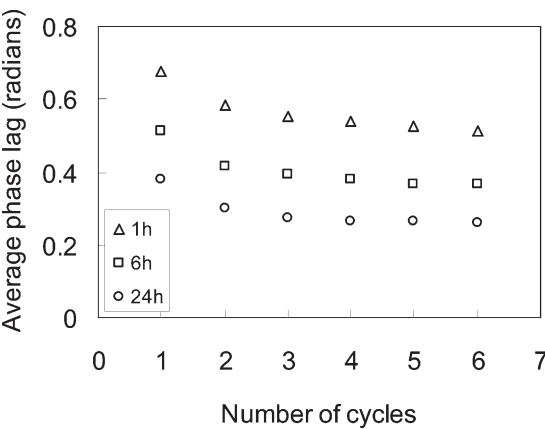


Figure 4. Plots of average phase lag against number of cycles for Sitka spruce at three cyclic periods.

Table 1. Phase lag ϕ (radians) and amplitude A (%) of moisture and radial and tangential dimensional responses.

Cyclic period	ϕ_{MC}^a	ϕ_R^a	ϕ_T^a	A_{MC}	A_R	A_T
1 h	0.60	0.53	0.46	0.79	0.17	0.36
6 h	0.44	0.41	0.36	1.75	0.39	0.53
24 h	0.31	0.30	0.27	1.93	0.52	0.58
5.33 h ^b	0.60	0.58	0.54	/	/	/
26.67 h ^b	0.38	/	0.38	/	/	/

^a Average values of all cycles.
^b From Chomcham and Skaar (1983).

Chomcham and Skaar related this interesting phenomenon to the fact that stresses associated with moisture gradients generally have a greater effect on dimensional than on moisture changes. If this is the reason, the phase lag of moisture should have almost the same value as that of dimensional response at the first cycle during cycling. However, it is not true because a large difference was found between the two at the initial cycle for each period, especially for the shortest one. Furthermore, there are differences in the radial and tangential phase lags as well. Greater lags in the radial swelling indicate that radial responses act slower than tangential responses. It also appears in this table that the corresponding phase lag values from Chomcham and Skaar’s research are greater than those from this work for a similar cyclic period. This is probably because of the differences in the species and measuring system between the two studies.

Figure 5 and Table 1 present the moisture and dimensional response amplitude with cyclic period. It is evident that the amplitude is less for the specimens subjected to the shortest cycle. This was anticipated because there was insufficient time for the specimen, especially the interior, to respond to the RH changes at short cyclic periods. As a result, the specimen in a short period never experiences the large changes in a long period.

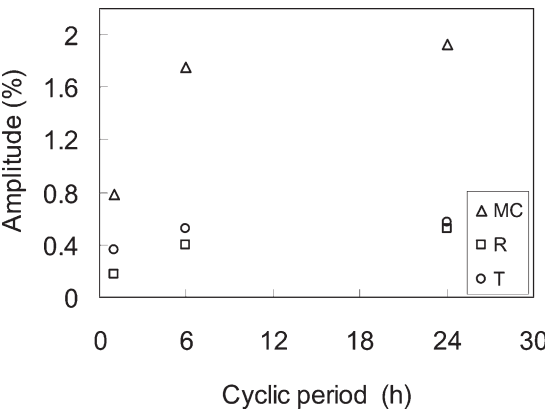


Figure 5. Plots of moisture content (MC) and radial (R) and tangential (T) dimensional response amplitude against cyclic periods for Sitka spruce.

Dynamic Sorption Modeling

Figure 6 shows a comparison of theoretical curves with experimental results of dynamic sorption for the three periods. The plots suggest that the theoretical curves calculated by the mathematical model agree satisfactorily with the experimental data. The calculated D ranges $0.96\text{--}1.29 \times 10^{-11} \text{ m}^2/\text{s}$. The value of a , determined when it can give the best fit to the observed data, is 40/h, 30/h, and 15/h for cyclic periods of 1, 6, and 24 h, respectively. It is

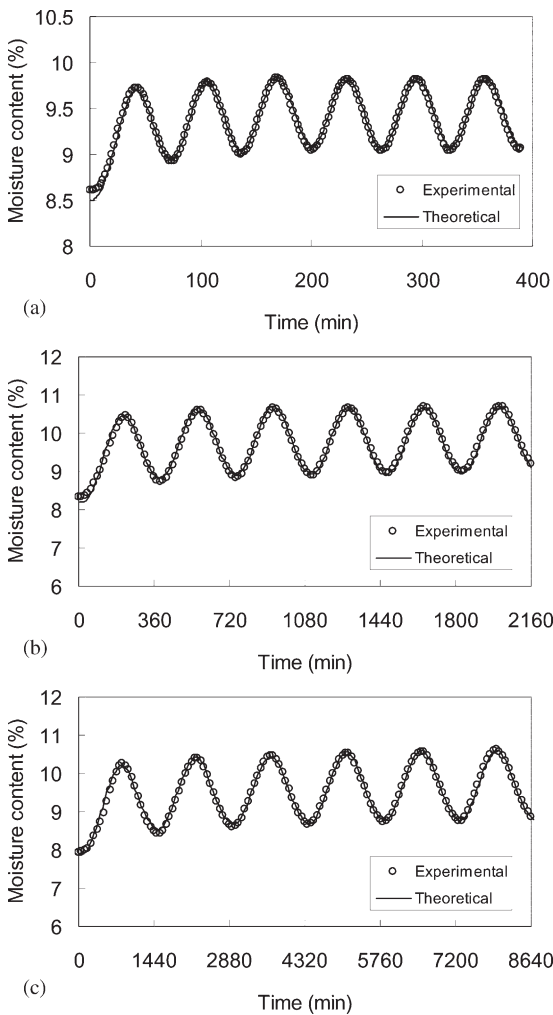


Figure 6. Comparison of theoretical curves with experimental results of dynamic sorption for Sitka spruce cycled at (a) 1 h, (b) 6 h, and (c) 24 h.

obvious that the sorption rate constant decreases with increasing cyclic period. This may be related to the fact that the boundary layer developed is very thin and therefore the actual thickness for moisture diffusion is greater for a shorter cyclic period.

Figure 7 gives the calculated moisture gradient distributions at some typical time steps for the adsorption and desorption of the second cycle at a period of 24 h. In the case of adsorption, 1500 min is when the specimens just started to pick up moisture. The RH at this time had already increased for about 1/24 cycle. However, as shown in this figure, moisture still takes on a regular shape of desorption distribution (Jönsson 2004). When adsorption reaches 1620 min, which is the time at about 1/6 of the adsorption process, the most uniform distribution is obtained by the adsorption of the surface and desorption of the central part along the thickness direction of the specimens. At 1860 min, the midpoint of adsorption, the entire sample began to sorb water

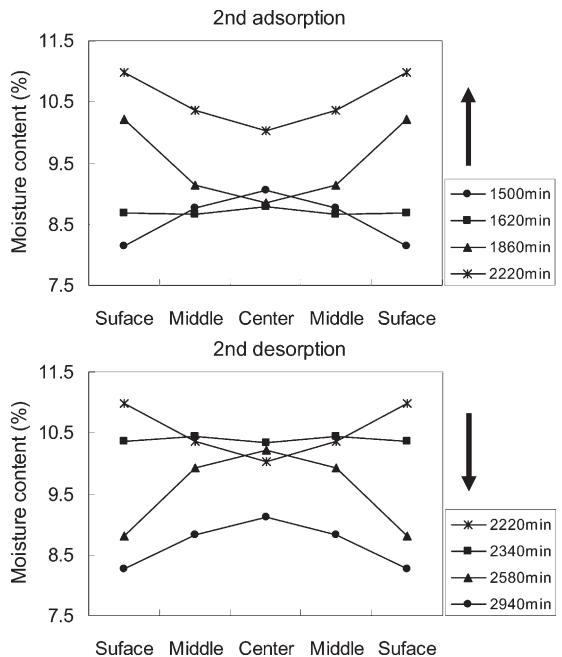


Figure 7. Moisture distributions of Sitka spruce for adsorption and desorption in the second cycle at the 24-h cyclical period.

and a typical adsorption moisture distribution was found. Therefore, initially the central part of the specimens does not respond to the moistening. It takes about 6 h until the center part is affected. Finally, 2220 min is the time at which MC reached the peak value, although RH had already decreased. The moisture gradient at this time is lower than that at 1860 min. Nevertheless, the moisture distribution is still far from equilibrium. We also assume that moisture distributions for desorption have the same tendencies as those for adsorption.

CONCLUSIONS

Dynamic sorptive and hygroexpansive behavior of wood under cyclic RH conditions was investigated in this study. The moisture and dimensional changes of the specimens were generally sinusoidal but lagged behind the imposed RH. The phase lag decreased and the amplitude increased with increasing cyclic periods. The mathematical model developed was verified not only for static sorption with constant RH (Ma et al 2009), but also for dynamic sorption with cyclical RH. The model takes into account the diffusion of moisture with a MC-dependent coefficient, and it is also able to provide moisture distributions developed throughout the wood. It is hoped that the results of this work could enable one to gain fuller and deeper insight into moisture sorption and hygroexpansion at nonequilibrium.

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