A NONLINEAR REGRESSION TECHNIQUE FOR CALCULATING THE AVERAGE DIFFUSION COEFFICIENT OF WOOD DURING DRYING

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ABSTRACT

A nonlinear regression technique for determining the optimum drying diffusion coefficient of wood is described. This technique uses the least squares principle to estimate the diffusion coefficient in the infinite Fourier series solution to the one-dimensional unsteady-state form of Fick's law. Five different nonlinear iteration methods for solving the normal equations were evaluated in terms of their ability to find a solution, the rate of convergence, and sensitivity to the parameter's starting value. Application of this technique to a series of experimental isothermal drying runs involving red oak (*Quercus rubra*) resulted in a better fit between the actual and predicted drying curves, as compared to the logarithmic, square-root, and half-$\bar{E}$ techniques. The technique yielded diffusion coefficients and residual sum of squares that were almost identical to those obtained from a Fortran-based optimization method reported by Chen et al. (1994). However, the technique described in this paper is more computationally efficient by converging to a solution in a fewer number of iterations than the optimization method.

Keywords: Diffusion, drying, diffusion coefficient, Fick's law, nonlinear regression, least-squares method.

INTRODUCTION

The moisture diffusion coefficient of wood is regarded as an important property because of its direct relation to the rate of moisture movement through wood. Various methods for estimating the diffusion coefficient of wood have been reported in the literature (Stamm 1964; Siau 1995). The earliest experimental measurements of the diffusion coefficient involved data obtained under steady-state condition. However, the time it takes for diffusion to attain steady state is sometimes considerable and thus necessitates the use of unsteady-state methods. The main disadvantage of the unsteady-state method is the mathematical complexity of the governing equation, which requires making simplifying assumptions for its solution. Under the assumptions that the surface moisture content of wood immediately decreases to the equilibrium moisture content of the drying air and the diffusion coefficient not being dependent on moisture content, the unsteady-state form of the diffusion equation has been solved using three approaches: logarithmic (LN), the square-root (SQRT), and the half-E (HALF-E) approach. Chen et al. (1994) described these three approaches and pointed out their limitations. They then presented a new approach for calculating the average moisture diffusion coefficient based on the entire wood drying curve. Their optimization method employs a Fortran program that performs a least squares analysis of the experimental and theoretical values of the fraction of evaporable moisture $\bar{E}$ still held by the wood at time t, and locates the unique value

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of the drying diffusion $\underline{c}$oefficient D when the sum of squares of the $\bar{E}$ differences is at the minimum. This paper presents an approach similar to that proposed by Chen et al. (1994), but uses nonlinear regression instead of a Fortran-based optimization method.

Equations that are nonlinear in the parameters may be represented by the general model

$$y_i = f(x_i, \theta) + \varepsilon_i \tag{1}$$

where y_i is the dependent variable, $f(x_i, \theta)$ is the nonlinear function relating the mean of the dependent variable to the independent variable(s), x_i is the vector of observations on k independent variables for the ith observational unit, θ is the p-dimensional vector of unknown parameters, and ε_i represent the unobservable experimental errors. Just as in linear regression, the least squares principle is used to estimate the parameters in nonlinear models. That is, the least squares estimate of θ is the choice of parameters that minimizes the residual sum of squares, SSR:

$$\mathrm{SSR} = \sum_{i=1}^{p} [Y_i - f(x_i, \hat{\theta})]^2 \tag{2}$$

where $\hat{\theta}$ is the least squares estimate of θ. The partial derivatives of SSR are set equal to zero to obtain the p normal equations, whose solution gives the least squares estimate of θ. The difficulty with nonlinear least squares is that the normal equations cannot be solved explicitly and therefore iterative numerical methods must be used. Starting values or initial guesses for the parameters must first be provided to calculate the initial residual sum of squares. Adjustments are then made to these starting parameter values so that the residual sum of squares is reduced. The process is repeated until a very small adjustment is being made at each step, at which point the residual sum of squares is considered minimized and the process is said to have converged to a solution.

This paper describes the method of determining the average diffusion coefficient of wood using nonlinear regression. Five different iteration methods for solving the normal equations were evaluated in terms of their ability to find a solution, the rate of convergence, and sensitivity to the parameter's starting value. The results of the nonlinear regression analysis (herein referred to as NLIN) were then compared with those obtained using the optimization method (Chen et al. 1994), logarithmic approa$\underline{c}$h, the square-root approach, and the half-$\bar{E}$ approach.

MATERIALS AND METHODS

Partially air-dried boards of red oak (*Quercus rubra*) with nominal thickness of 2.7 cm and nominal width of 6.5 cm were cut to lengths of 14.0 cm for the longitudinal diffusion study and 20.3 cm for the radial diffusion study. Actual dimensions of each sample were measured during the experiments using a caliper with 0.001-cm readability. For the longitudinal diffusion experiments, the samples were coated with a commercial kiln-sample sealant on all surfaces except the two ends; for the radial experiments, the samples were coated on all surfaces except the wide faces. The samples were subjected to water soaking under vacuum to raise their moisture contents above the fiber saturation point. The samples were then dried to equilibrium at a temperature of 36°C and relative humidity of 86% (equilibrium moisture content, EMC = 16.7%) to obtain the diffusion coefficient at high moisture content. The same samples that were equilibrated at the initial condition were further dried to equilibrium at a temperature of 36°C and relative humidity of 65% (EMC = 11.5%), and then at a temperature of 36°C and relative humidity of 45% (EMC = 8%) to obtain the diffusion coefficients at lower moisture content levels. The weights at different times during equilibration, together with the sample dimensions, were used to calculate the diffusion coefficient. The radial diffusion experiments were performed with four replications, while the longitudinal experiments consisted of six replicates. The drying experiments were performed in a wind tunnel placed inside a walk-in conditioning chamber. The wind tunnel consisted of a drive section powered by a 35-cm-diameter fan/245-watt motor, a 43-cm-long diffuser section, a 25-cm $\times$ 18-cm $\times$ 30-cm test section, an entrance cone, and a settling chamber. Air velocity in the test section was maintained at 760 cm/s. Temperature- and humidity-controlled air was supplied to the conditioning chamber by a PGC generator.

For all drying data, the sample half-thickness a and the values of $\bar{E}$ at different times t during the drying process were used in a nonlinear regression analysis to obtain an optimized value of the diffusion coefficient D based on the following equation (Crank 1975):

$$\bar{E} = \frac{8}{\pi^2} \sum_{n=1}^{\infty} \left(\frac{1}{2n - 1}\right)\left[\exp\left(\frac{-(2n - 1)^2\pi^2 tD}{4a^2}\right)\right] \tag{3}$$

All nonlinear regression analyses were performed using the latest version of the Statistical Analysis System (SAS 1999) software. To enable the reader to validate or examine the NLIN technique, the SAS program is included at the end of this paper. Only the first four terms of the infinite Fourier series solution of the one-dimensional unsteady-state form of Fick's law are shown in the program, although more terms may be added to make the calculation more accurate especially at high values of $\bar{E}$. The generated values for the diffusion coefficient were compared with those obtained from other schemes described by Chen et al. (1994).

RESULTS AND DISCUSSION

Several iterative methods are available for computing the least squares estimate of the parameters in nonlinear models. Five of these are available in the NLIN procedure of the SAS software: the modified Gauss-Newton method (referred to in this paper as GAUSS), the steepest-descent or gradient method (GRADIENT), the Marquardt method (MARQUARDT), the Newton method (NEWTON), and the multivariate secant or false position method (DUD). In the SAS program, the iteration method is specified as an option in the PROC NLIN statement. Thus in the sample program in the Appendix, the option ''Method=Marquardt'' indicates that the program must use the MARQUARDT iteration method. If the method option is omitted, the GAUSS method is used by default. The different iteration methods use derivatives or approximations to derivatives of the residual sum of squares with respect to the parameters to guide the search for the parameters producing the smallest residual sum of squares. The details of the numerical procedure for finding the least squares solutions are found in Gallant (1987) and SAS (1999). The choice of a method is important since they differ in terms of their ability to converge and in their rates of convergence to a solution. The sensitivity to the value of the initial estimate is also an important consideration in the choice of a method of iteration. If local minima exist in the residual sum of squares surface, a poor starting value increases the chance that the method will converge to a local minimum instead of the global minimum. Table 1 summarizes the results of nonlinear regression analyses using the five different NLIN iteration methods. The data used for the analyses were those for the drying from 16.7% EMC to 11.5% EMC in the radial direction. Similar results were obtained at the other radial drying conditions and for drying in the longitudinal direction. The table shows that the GAUSS and MARQUARDT methods are more robust than the NEWTON, GRADIENT, and DUD methods. When the starting values of D were chosen to be within the expected range of 1×10^{-7} to 1×10^{-4} cm^2/s for the transverse diffusion coefficient and 1×10^{-6} to 1×10^{-3} cm^2/s for the longitudinal diffusion coefficient (Siau 1995), both GAUSS and MARQUARDT methods converged to a solution at the least number of iterations. The other three methods either did not converge to a solution, were slow to converge to a solution, or converged to a local minimum. These methods are therefore not recommended for analyzing wood-

TABLE 1. *Summary of results of the evaluation of five different nonlinear regression iteration methods for calculating the radial diffusion coefficient of red oak dried from 16.7% EMC to 11.5% EMC.*

Evaluation criteria	Evaluation results for the different iteration methods				
	Gauss	Marquardt	Newton	Gradient	DUD
Initial estimate (cm^2/s)	1.00E-10	1.00E-10	1.00E-10	1.00E-10	1.00E-10
Converge?	Yes	Yes	Yes	No	Yes
No. of iterations	9	9	12	0	2
D value (cm^2/s)	8.73E-07	8.73E-07	8.73E-07	1.00E-10	1.10E-10
Residual SS	0.00935	0.00935	0.00935	7.7158	7.7126
Initial estimate (cm^2/s)	1.00E-08	1.00E-08	1.00E-08	1.00E-08	1.00E-08
Converge?	Yes	Yes	Yes	No	Yes
No. of iterations	8	8	10	0	2
D value (cm^2/s)	8.73E-07	8.73E-07	8.73E-07	1.00E-08	1.10E-08
Residual SS	0.00935	0.00935	0.00935	6.2764	6.1909
Initial estimate (cm^2/s)	1.00E-06	1.00E-06	1.00E-06	1.00E-06	1.00E-06
Converge?	Yes	Yes	Yes	No	Yes
No. of interations	4	4	4	0	1
D value (cm^2/s)	8.73E-07	8.73E-07	8.73E-07	1.00E-06	1.00E-06
Residual SS	0.00935	0.00935	0.00935	0.0227	0.0227
Initial estimate (cm^2/s)	1.00E-04	1.00E-04	1.00E-04	1.00E-04	1.00E-04
Converge?	Yes	Yes	No	No	Yes
No. of iterations	10	8	0	22	10
D value (cm^2/s)	8.73E-07	8.73E-07	1.00E-04	8.73E-07	8.01E-07
Residual SS	0.00935	0.00935	5.8206	0.00935	0.015
Initial estimate (cm^2/s)	1.00E-02	1.00E-02	1.00E-02	1.00E-02	1.00E-02
Converge?	Yes	Yes	No	No	Yes
No. of iterations	11	16	0	21	2
D value (cm^2/s)	8.73E-07	8.73E-07	1.00E-02	8.73E-07	−1.58E-01
Residual SS	0.00935	0.00935	9.8080	0.00935	0.00041

drying data. The GAUSS and MARQUARDT methods were robust enough that even when very low (1×10^{-10} cm^2/s) or very high (1×10^{-2} cm^2/s) starting values were used, the two methods still converged to a solution. The two methods differed in terms of their rate of convergence as shown by the total number of iterations required to reach a solution. However, the difference is so slight that either one may be used for determining the diffusion coefficient of wood.

Table 2 shows part of the output of the NLIN procedure using the Marquardt iteration method with a starting value of 1×10^{-10} cm^2/s. The drying data were the same as those used in Table 1. The table shows that it took the program only nine iterations to converge to a solution. The letter R in the output should not be confused with the sample correlation coefficient often encountered in linear regression. Instead, R (together with PPC, RPC, and Object) is a measure of convergence: the relative offset convergence measure of Bates and Watts (1981). It measures the degree to which the residuals are orthogonal to the Jacobian columns, and approaches zero as the gradient of the objective function becomes small. When this measure is less than 1×10^{-5}, convergence is declared. PPC and RPC are, respectively, the prospective and retrospective parameter change measure. A PPC value of 9.30×10^{-8} cm^2/s indicates that the diffusion coefficient D would change by that relative amount if NLIN were to take an additional iteration step, while an RPC value of 1.35×10^{-6} cm^2/s indicates that D changed by that amount relative to its value in the previous iteration. Object measures the relative change in the objective function value between iterations. Thus, in this particular example, E

TABLE 2. *Part of the output of the Statistical Analysis System nonlinear regression program using the Marquardt iteration method for calculating the radial diffusion coefficient of red oak dried from 16.7% EMC to 11.5%. The starting value used for the iteration was* 1×10^{-10}*cm*^{2}/s.

Method	Marquardt
Iterations	9
R	1.84E-06
PPC(D)	9.30E-08
RPC(D)	1.35E-06
Object	7.65E-10
Objective	0.009351

Source	DF	Sum of squares	Mean square	F value	Pr > F
Regression	1	10.8339	10.8339	28964.8	<.0001
Residual	25	0.00935	0.000374		
Uncorrected total	26	10.8432			
Corrected total	25	2.9602			

Parameter	Estimate	Approx std error	Approximate 95% confidence limits		Skewness
D	8.73E-07	1.89E-08	8.34E-07	9.12E-07	0.0763

changed by 7.65×10^{-10} in relative value from the last iteration.

The next part of the output shows the least squares summary statistics for the model. For the least squares estimate of the diffusion coefficient ($D = 8.73 \times 10^{-7}$ cm^2/s), which is given in the next part of the output, the residual sum of squares is 0.00935 cm^4/s^2, resulting in a variance estimate of 0.000374 cm^4/s^2. The measure of skewness shown on the next part of the output is added to the parameter estimation table if the Hougaard output option is included in the PROC NLIN statement (see sample program in the Appendix). The Hougaard measure of skewness determines whether a parameter is close to linear or whether it contains considerable nonlinearity. This is important since the least squares estimates of the parameters of a nonlinear model are close to being unbiased, normally distributed, and minimum variance estimators only if the nonlinear model is close to linear. Bias in the parameters can render inferences using the reported standard errors and confidence limits invalid. If the skewness is less than 0.1, the estimator of parameter is very close to linear in behavior; while if it is greater than 1, the nonlinear behavior is considerable. The skewness measure of 0.0763 shown on Table 2 indicates that the diffusion coefficient D is nearly linear and that its standard error and 95% confidence interval can be safely used for inferences.

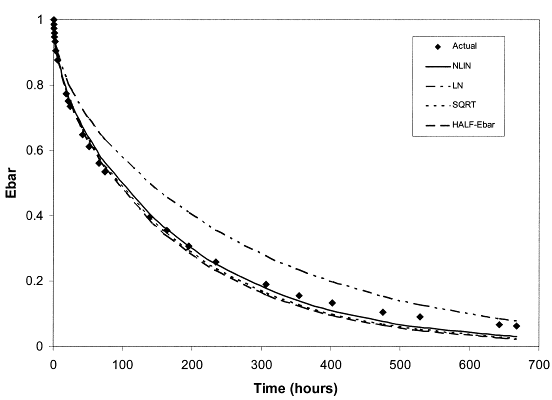

FIG. 1. Graph showing the fraction of evaporable moisture $\bar{E}$ plotted as a function of time for the actual experimental data and those predicted by the nonlinear regression (NLIN), logarithmic (LN), square-root (SQRT), and half-$\bar{E}$ (HALF-$\bar{E}$) approaches to calculating the diffusion coefficient.

A representative graph showing the fraction of evaporable moisture $\bar{E}$ plotted as a function of time for the actual experimental data and those predicted by the NLIN, LN, SQRT, and HALF-$\bar{E}$ methods is presented in Fig. 1. It is apparent from the graph that the nonlinear regression approach came closest to the actual data. The curve for the optimization approach proposed by Chen et al. (1994) is not included in the figure since it was practically identical to that of the nonlinear regression approach. The data used in Fig. 1 were those for the drying from 16.7% EMC to 11.5% EMC in the radial direction. Table 3 summarizes the results for radial drying at other conditions and for the drying in the longitudinal direction. In the NLIN and CHEN approaches, the diffusion coefficient for the radial flow direction increased with increasing moisture content, while the reverse is true for moisture movement in the longitudinal direction. These results are consistent with the theoretical discussion of the drying diffusion coefficient presented by Siau (1995). A comparison of the five approaches to calculating the diffusion coefficient of wood shows that the NLIN and CHEN approaches yielded values that were consistently lower than those obtained using the SQRT and HALF-E approaches, but generally higher than those obtained using the LN approach. More important, however, is the lower residual sum of squares obtained using the NLIN and CHEN approaches compared to the other three. This means that the NLIN and CHEN approaches gave better estimates of the diffusion coefficient, thereby resulting in better fit between the predicted and the actual drying curves. In the radial direction, the diffusion coefficients that were calculated by the HALF-E, SQRT, and LN approaches deviated, on average, from those obtained using the NLIN approach by 5%, 3%, and 19%, respectively. In the longitudinal direction, the respective average percent deviations are 21%, 30%, and 34%. In evaluating the diffusion coefficients of six different species, Chen et al. (1994) reported that in the longitudinal direction, the HALF-E, SQRT, and LN approaches yielded values that had maximum percent deviations of 12.8%, 170.6%, and 62.5%, respectively, from their optimization approach. In the transverse direction, the respective maximum percent deviations were 6.2%, 45.3%, and 27.5%.

The NLIN and CHEN approaches yielded

TABLE 3. *A comparison of the average diffusion coefficient (D) and residual sum of squares (Residual SS) obtained using five different calculation methods: nonlinear regression (NLIN), optimization by Chen et al. (CHEN), logarithmic (LN), square-root (SQRT), and half-E (HALF-E). The data are for red oak subjected to three drying conditions in the radial and longitudinal directions.*

Direction and condition[1]	NLIN D (cm^2/s)	NLIN Residual SS	CHEN D (cm^2/s)	CHEN Residual SS	LN D (cm^2/s)	LN Residual SS	SQRT D (cm^2/s)	SQRT Residual SS	HALF-E D (cm^2/s)	HALF-E Residual SS
Radial										
I	1.02E-06	0.00246	1.02E-06	0.00246	9.05E-07	0.02576	1.03E-06	0.00277	1.04E-06	0.00329
II	8.73E-07	0.00935	8.73E-07	0.00935	6.16E-07	0.10451	9.16E-07	0.01106	9.38E-07	0.01318
III	7.51E-07	0.00340	7.51E-07	0.00340	6.25E-07	0.02393	7.75E-07	0.00402	7.98E-07	0.00576
Longitudinal										
I	1.80E-05	0.04122	1.75E-05	0.04053	1.18E-05	0.21044	2.34E-05	0.11922	2.17E-05	0.08366
II	2.10E-05	0.00419	2.12E-05	0.00414	2.31E-05	0.00942	2.27E-05	0.00750	2.17E-05	0.00458
III	2.20E-05	0.00408	2.22E-05	0.00404	1.86E-05	0.01494	2.41E-05	0.00669	2.27E-05	0.00426

[1] Condition I, II and III refer to the drying from green to 16.7% EMC, from 16.7% to 11.5% EMC, and from 11.5% to 8% EMC, respectively.

TABLE 4. *Number of iterations needed to converge to a solution for the Marquardt iteration method of the nonlinear regression approach (NLIN) and for the golden section search routine of the optimization approach (CHEN) by Chen et al. (1994). The two starting values (SV) for the nonlinear regression approach correspond to the upper and lower bounds of the optimization approach.*

	Number of iterations		
	NLIN		
Direction and condition[1]	SV = 1×10^{-4} cm^2/s	SV = 1×10^{-10} cm^2/s	CHEN
Radial:			
I	15	8	29
II	8	9	29
III	8	7	29
Longitudinal			
I	8	9	29
II	5	6	29
III	5	6	29

[1] Condition I, II and III refer to the drying from green to 16.7% EMC, from 16.7% to 11.5% EMC, and from 11.5% to 8% EMC, respectively.

almost identical values for the diffusion coefficient and the residual sum of squares. But a closer look at the algorithms for performing the calculations shows that NLIN is more efficient than CHEN. Table 4 shows the number of iterations it took for the NLIN and CHEN approaches to converge to a solution. Two starting values were used in the NLIN approach: 1×10^{-4} and 1×10^{-10} cm^2/s. These starting values correspond to the upper and lower bounds of the golden section search routine of the optimization method by Chen et al. (1994). As shown in Table 4, the Marquardt iteration method of the NLIN approach converged to a solution in fewer iterations than the golden section search routine of the CHEN approach. When run on a Pentium III desktop computer with 256 MB memory and operating at 733 MHz, the processing time for the CHEN approach was at least an order of magnitude longer than that for the NLIN approach. Using the data for the drying from green to 16.7% EMC, it took about 1.3 s for the CHEN approach to converge to a solution while it only took 0.05 s for the NLIN approach to converge to a solution. For drying from 16.7% to 11.5%, and from 11.5% to 8% EMC, the CHEN approach took about 0.6 s while the NLIN approach took about 0.05 s to converge to a solution. The low efficiency of a line-search approach such as the golden section search routine is well documented in the literature (Fletcher 1980).

CONCLUSIONS

The nonlinear regression approach offers a dependable alternative for calculating the diffusion coefficient of wood from drying data. Just like the optimization approach by Chen et al. (1994), this approach is objective and yields a diffusion coefficient value that gives better fit between the predicted and the actual drying curves than previously available methods. By converging to a solution in fewer numbers of iterations, the nonlinear regression approach is more computationally efficient than the optimization approach.

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APPENDIX

Nonlinear regression program for the Statistical Analysis System to calculate the diffusion coefficient of wood from drying data

TITLE '⟨Descriptive title⟩';
DATA TimeEbar;

```
   INFILE '⟨path and filename of input data⟩';
   INPUT Time Ebar;
RUN;
PROC NLIN METHOD=MARQUARDT Hougaard;
   PARMS D=⟨starting value⟩;
   pi=arcos(−1);
   a=⟨half-thickness of the sample⟩;
   b= ((pi**2)*time)/(4*(a**2));
   term1=exp(−(b*D));
   term2=(1/9)*(exp(−(9*b*D)));
   term3=(1/25)*(exp(−(25*b*D)));
   term4=(1/49)*(exp(−(49*b*D)));
   MODEL Ebar=(8/(pi**2))*(term1+term2+term3+
         term4);
   OUTPUT OUT=B PREDICTED=EBARHAT
         RESIDUAL=RES;
RUN;
PROC PRINT;
   VAR Time Ebar EBARHAT RES;
RUN;
PROC PLOT;
   PLOT Ebar*Time='*' EBARHAT*Time='P' /
         OVERLAY;
RUN;
```