

# An irreversible thermodynamics model for unsteady-state nonisothermal moisture diffusion in wood

S. Avramidis, S. G. Hatzikiriakos, J. F. Siau

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**Summary** A model that predicts heat and moisture transfer through wood in the hygroscopic range and which is based on the principles of irreversible thermodynamics, was evaluated with unsteady-state nonisothermal moisture desorption experimental data. The model predicted the phenomenon of thermal diffusion during the initial stages of desorption and results in a very good simulation of the desorption curve and the center's temperature change with time.

## Symbols

$c_p$	specific heat of air (= 0.24 cal/g K @ 70 °C)
$c_T$	specific heat of wood, cal/g K
$D$	transverse diffusion coefficient, cm <sup>2</sup> /s
$E_b$	activation energy, cal/mol
$E_o$	heat of vaporization, cal/mol
$E_L$	differential heat of sorption, cal/mol
$G$	specific gravity of wood
$H$	relative humidity, %
$h_T$	convective heat transfer coefficient, cal/cm <sup>2</sup> s K

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*Received 16 December 1992*

Dr. Stavros Avramidis  
Associate Professor  
Department of Wood Science

Dr. Savvas G. Hatzikiriakos  
Assistant Professor  
Department of Chemical Engineering

University of British Columbia  
Vancouver, B.C. V6T 1Z4  
Canada

Dr. John F. Siau  
Professor Emeritus  
P.O. Box 41  
Keene, NY 12942  
USA

## *current address*

Department of Wood Science and Forest Products  
Virginia Polytechnic Institute and State University  
Blacksburg, Virginia 24061-0503

$h_c$	convective mass transfer coefficient based on the concentration of moisture in wood, cm/s
$h_v$	convective mass transfer coefficient based on the concentration of moisture in the air in equilibrium with the wood surface, cm/s
$K_M$	coefficient for diffusion due to moisture gradient, g/cm s %
$K_T$	transverse thermal conductivity coefficient, cal/cm K s
$M$	moisture content, %
$p_o$	saturated vapor pressure, atm
$R$	universal gas constant, cal/mol K (= 82.056 cm <sup>3</sup> atm/mol K)
$t$	time, s
$T$	temperature, K
$x$	distance, cm

### Greek letters

$\varepsilon$	evaporation or condensation criterion
$\rho$	wood density, g/cm <sup>3</sup>
$\rho_w$	water density (=1), g/cm <sup>3</sup>
$\rho_a$	air density, g/cm <sup>3</sup>

### Introduction

In a hygroscopic porous material like wood, a model which adequately describes moisture and heat transfer can be used to study complex industrial processing schemes, to facilitate experimental testing and to explain the physical mechanisms underlying those particular transfer processes. Areas of application include drying, moisture sorption under adverse environmental conditions and interpretation of mechano-sorptive phenomena.

A considerable volume of research has been carried-out regarding modeling heat and moisture transfer in materials like, polymers, wood, soils, coatings and agricultural products (Parrauffe, Mujumdar 1988; Perre et al. 1986; Parry 1985; Strongin, Border 1986; Oliveira, Fernandes 1986; Croll 1987; Blondin et al. 1987). In wood, model developments have been based on either mechanistic approaches in which, macroscopic descriptions of transfer phenomena have been derived from Fourier's and Fick's laws, or the principles of irreversible thermodynamics and the entropy production in the system which can identify the driving forces and relate them to the heat and moisture fluxes through a number of phenomenological coefficients (Stanish et al. 1984; Plumb et al. 1984; Beard et al. 1983; Thomas et al. 1980; Adesanya et al. 1988; Bramhall 1979 a; 1979 b; Liu 1990; Kayihan 1986; Avramidis, Siau 1987; Avramidis et al. 1992; Barbour, Johnson 1989).

Modeling of moisture fluxes and profiles in wood under unsteady-state nonisothermal conditions, has been noticeably absent from the literature. Heat and moisture transfer should be considered as coupled processes and the thermally induced mass transfer (Soret effect), which could considerably contribute to the total moisture flux, should always be taken into account (DeGroot, Mazur 1962; Prigogine 1961; Fortes, Okos 1978). Traditionally, moisture content gradients have been employed as the driving forces for diffusion (Droin et al. 1988; Droin-Josserand et al. 1988, 1989 a, 1989 b; Vergnaud 1991). In addition, the gradients of chemical potential, water potential, partial and spreading pressures have also been used (Nelson 1986 a, 1986 b, 1986 c; Siau 1983 b, 1984 a, 1984 b; Cloutier et al. 1992)

The phenomenological coefficients which describe the direction and magnitude of the moisture flux through wood, were derived based on either a gradient of activated water molecules or gradients of water or chemical potential as diffusion driving forces and the developed models were evaluated under steady-state nonisothermal conditions (Avramidis et al. 1987; Avramidis, Siau 1987; Siau et al. 1986; Siau, Avramidis 1992; Siau, Avramidis 1993). The purpose of this study was to employ a recently derived phenomenological coefficient of the temperature

gradient in the mass balance equation, in the formulation of a set of coupled partial differential equations to describe moisture and heat transfer in wood undergoing unsteady-state non-isothermal desorption.

### Model development

The general equations that describe one dimensional coupled heat and moisture transfer in the hygroscopic range (no free water) of a porous material, may be written in the following form (Prigogine 1961; Luikov 1966),

$$J_m = -K_{mq} \frac{dT}{dx} - K_{mm} \frac{dM}{dx} \quad (1 a)$$

$$J_q = -K_{qq} \frac{dT}{dx} - K_{qm} \frac{dM}{dx} \quad (1 b)$$

where  $J_m$  and  $J_q$  are the moisture and heat fluxes, respectively,  $dT/dx$  and  $dM/dx$  are the gradients of temperature and moisture content, and the quantities  $K_{mq}$ ,  $K_{mm}$ ,  $K_{qm}$ , and  $K_{qq}$ , are the phenomenological coefficients. In a previous series of studies (Avramidis, Siau 1987; Avramidis et al. 1987), two expressions for the phenomenological coefficients of Eq. (1 a), were proposed and the predictions were in good agreement with the experimental moisture fluxes obtained in steady-state nonisothermal experiments. Furthermore, the above expressions were successfully evaluated by Avramidis et al. (1992) under dynamic nonisothermal conditions.

Equations analogous to (1 a, b) can be derived from the principles of irreversible thermodynamics. One form of these equations may be written as:

$$J_m = -(L_{mq}/T)(dT/dx) - L_{mm}(\partial M/\partial x)_T \quad (2 a)$$

$$J_q = -(L_{qq}/T)(dT/dx) - L_{qm}(\partial M/\partial x)_T \quad (2 b)$$

where the fluxes  $J_m$  and  $J_q$  are the same as those in equations (1 a) and (1 b), but the phenomenological coefficients  $L_{mq}$ ,  $L_{mm}$ ,  $L_{qq}$  and  $L_{qm}$ , as well as the assumed gradients  $(1/T)(dT/dx)$  and  $(\partial \mu/\partial x)_T$ , where  $\mu$  is the chemical potential of the sorbed water, are all different from those in equations (1 a) and (1 b). They also differ in that the Onsager relations are satisfied, that is,  $L_{mq} = L_{qm}$ , whereas  $K_{mq} \neq K_{qm}$ .

It can be shown from the equations (1 a, b) and (2 a, b) that

$$K_{mq} = K_{mm} \left( \frac{H}{RT} \right) \left( \frac{\partial M}{\partial H} \right)_T \left( \frac{E_b}{T} \right) \quad (3)$$

provided the activation energy  $E_b$  for diffusion is equated to  $Q^*$ , the heat of transfer of bound water (Siau 1992), as suggested by Briggs (1967). The transport coefficient  $K_{mm}$  based on a moisture content gradient is related to  $D$  as follows,

$$K_{mm} = K_M = \frac{G\rho_w D}{100} \quad (4)$$

The single component moisture conservation equation without chemical reaction is given by (Siau 1983 a),

$$\begin{aligned} \frac{\partial M}{\partial t} = & D \left( \frac{\partial^2 M}{\partial x^2} \right) + \left( \frac{\partial D}{\partial M} \right) \left( \frac{\partial M}{\partial x} \right)^2 + \left( \frac{\partial \phi}{\partial M} + \frac{\partial D}{\partial T} \right) \left( \frac{\partial M}{\partial x} \right) \left( \frac{\partial T}{\partial x} \right) \\ & + \left( \frac{\partial \phi}{\partial T} \right) \left( \frac{\partial T}{\partial x} \right)^2 + \phi \left( \frac{\partial^2 T}{\partial x^2} \right) \end{aligned} \quad (5)$$

where

$$\phi = \left(\frac{E_b}{T}\right) \left(\frac{DH}{RT}\right) \left(\frac{\partial M}{\partial H}\right) \quad (6)$$

When  $\phi$  is multiplied by  $G\rho_w/100$ , it yields the phenomenological coefficient  $K_{mq}$  of Eq. (1 a). Since  $\phi$  is a strong function of both moisture and temperature, then

$$\frac{\partial \phi}{\partial M} = \left(\frac{\partial M}{\partial H}\right) \left(\frac{D}{RT^2}\right) \left[E_b \left(\frac{70H}{RT} + \frac{\partial M}{\partial H}\right) - 70H\right] \quad (7)$$

where  $\partial M/\partial H$  is assumed constant and  $dE_b/dM = 70$  cal/mol %, and

$$\frac{\partial \phi}{\partial T} = \left(\frac{\partial M}{\partial H}\right) \left(\frac{DHE_b}{RT^2}\right) \left(\frac{E_b}{RT} - 2\right) \quad (8)$$

where  $\partial M/\partial H$ ,  $E_b$  and  $H$  are assumed constant.

The energy balance equation introduced by Luikov (1966), which describes heat transfer through a capillary porous medium like wood, is required in addition to Eq. (5). This equation is given as:

$$\frac{\partial T}{\partial t} = \frac{K_T}{\rho c_T} \left(\frac{\partial^2 T}{\partial x^2}\right) + \frac{Q_b \varepsilon}{100 c_T} \left(\frac{\partial M}{\partial t}\right) \quad (9)$$

where  $Q_b = E_b/18$  (cal/g), and  $\varepsilon$  is the ratio of the vapor diffusion coefficient in wood to the coefficient of total diffusion of moisture inside wood. Hence,  $\varepsilon$  defines the amount of transferable vapor in wood in relation to the total flow of moisture in wet wood. In this study, since moisture transfer occurred only by bound-water diffusion without any liquid phase present,  $\varepsilon$  was set equal to one. The equations of the parameters, most of which are strong functions of moisture content and temperature, and of the sorption isotherms required in the solution of Eqs. (5) and (9), are given in the appendix. They were derived based on experimental data obtained in previous studies (Choong 1962; Stamm 1964).

The boundary conditions utilized to solve Eqs. (5) and (9) are those of no heat and mass flux at the axis of symmetry and convective heat and mass transfer at the exposed surface. The consideration of the convective heat and mass transfer boundary conditions was necessary, particularly for the heat transfer coefficient in order to take into account the energy required for a bound water molecule to vaporize when it diffuses to the interface. Thus, the boundary conditions can be summarized as follows:

$$\frac{\partial T}{\partial x} = 0 \wedge \frac{\partial M}{\partial x} = 0 \quad (10)$$

at the axis of symmetry ( $x = 0$ ), and

$$-D \frac{\partial M}{\partial x} - \phi \frac{\partial T}{\partial x} = h_c (M - M_\infty) \quad (11)$$

$$-K_T \frac{\partial T}{\partial x} + \frac{(E_0 + E_L)\rho D}{1800} \frac{\partial M}{\partial x} = h_T (T - T_\infty) \quad (12)$$

at the wood/air interface ( $x = x_0$ ), where  $h_c$  and  $h_T$  are the convective mass and heat transfer coefficients, respectively, and  $h_c = h_v (18p_0/RTG\rho_w)(\partial H/\partial M)$ . For  $h_T$ , a value of  $0.0004$  cal  $\text{cm}^{-2}$   $\text{K}^{-1}$   $\text{s}^{-1}$  was considered. This value compares very well with other values reported in literature by other investigators in the field (Thomas et al. 1980, Liu 1990). To calculate the convective mass transfer coefficient,  $h_v$ , a very simple heat and mass transfer

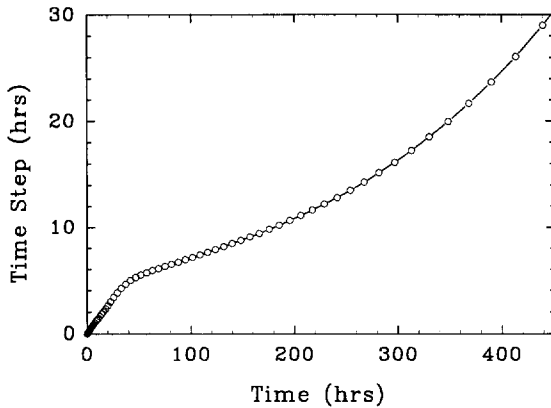


Fig. 1 The time step illustrating the accumulation of the calculations

correlation was used. The Lewis number (ratio of thermal diffusivity,  $\alpha$ , to binary diffusion coefficient of water vapour to air,  $D_v$ ) at the boundary layer is about 1 and thus, the thermal and concentration boundary layers thicknesses are of comparable size. It is therefore, reasonable to assume that the Nusselt number,  $Nu$ , is about equal to the Sherwood number,  $Sh$ . This leads to a relationship between the mass and heat convective transfer coefficients, that is  $h_v = h_T / \rho_a c_p$ , where  $\rho_a = (0.0013)(273/T)$ . Consequently, this results in an average value for  $h_v$  of 1.66 cm/s. The properties of the vapor-air system change only at the very early stages of the desorption process, since the temperature and moisture content at the exposed specimen surface attain their steady-state value ( $T_\infty$  and  $M_\infty$ ) within a very short period of time. Thus, the consideration of average convective heat and mass transfer coefficients is deemed as a fairly good assumption for this particular study.

**Method of solution**

The Galerkin finite element method was used to solve Eqs. (5) and (9). This method has been used in the past to successfully solve coupled heat and mass transfer problems (Chen, Pei 1989). A graded mesh was generated with a denser element concentration at the wood/air interface and fifty elements were used in all simulations. Their relative size is shown in Figs. 5 and 7.

A forward-backward Euler scheme (corrector-predictor), was used for the time integration. In addition, a control time step algorithm was also incorporated into the computer program in order to expedite the calculations. Figure 1 shows the time step as a function of time. As seen, initially the time step was very small due to the steep temperature and moisture gradients. However, as the temperature and moisture gradients decreased, the time step increased dramatically, thus accelerating the calculations. It should also be noted that the time step at the end of the simulations could be as large as 30 hours.

**Experimental**

A rectangular 100 mm by 200 mm by 40 mm in the radial direction all-heartwood specimen of Douglas-fir (*Pseudotsuga menziessi* (Poir.) Britton), was cut from a piece of lumber never dried before. The specimen was initially oven-dried at  $103 \pm 2$  °C to constant weight and then placed in a conditioning chamber at ambient temperature of 20 °C until it reached an equilibrium moisture content of 17.3%, approximately. The conditioning process lasted until there was no detectable weight change in 48 hours.

After reaching equilibrium, the specimen was immediately placed in a plastic bag and stored in a controlled environment room at 20 °C. Concurrently, the conditions in the conditioning chamber were changed to 70 °C and 35% relative humidity resulting in an equilibrium moisture content of 5%, approximately. As soon as the new conditions were reached, the specimen was immediately transferred back to the chamber and the change of temperature at its

geometric center was monitored with time by a copper-constantan type T, teflon insulated thermocouple that was connected to a data logger. At the same time, the change of its weight with time was monitored with a digital balance. Since the weight monitoring required removal of the specimen from the chamber, efforts were made for this to be done as fast as possible (10–15 seconds), thus, minimizing any sorption interruptions.

Heat and moisture transfer took place along the specimen's thickness through the top and bottom surfaces. To accomplish this, the four sides of the specimen were sealed with epoxy resin and rubber sheets and covered flush by thick styrofoam as shown in Fig. 2. A state of equilibrium was assumed to be reached and the experimental run completed when there was no detectable weight change of the styrofoam-specimen assembly for 48 hours.

### Results and discussion

Figure 3, shows the experimentally obtained average moisture contents of the specimen plotted against time. The total time required to reach the targeted equilibrium moisture content of 5% was 418 hours, approximately. The desorption curve predicted by the model is also included in the same plot. It is apparent that the derived model which uses the new form of the phenomenological coefficient  $L_{mq}$ , is in excellent agreement with the experimental data. This agreement is better compared to the one obtained from a previously tested similar form model where  $L_{mq}$  was derived based on the assumption that water chemical potential  $\mu$ , was the driving force for diffusion (Avramidis et al. 1992).

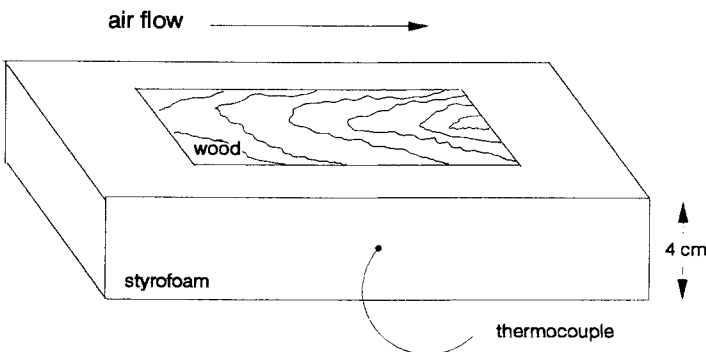


Fig. 2 Wood-styrofoam experimental assembly

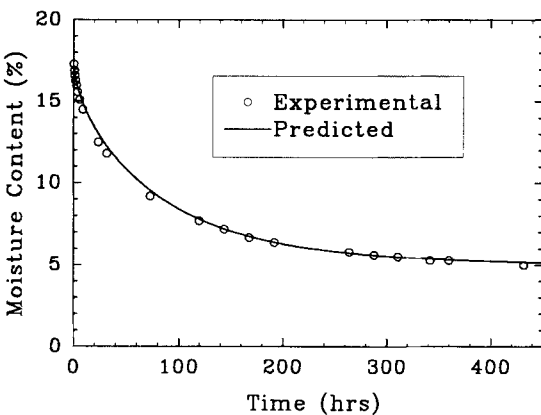


Fig. 3 Experimental and model predicted average moisture content as a function of time

The moisture profiles predicted by the model are shown in Fig. 4. It is seen that the specimen's surfaces reached the equilibrium moisture content of 5% during the first ten minutes of the desorption experiment. This was expected since a high value of convective mass transfer coefficient was used. At the beginning of the experiment, it is noted that the moisture content just under the surface at which evaporation takes place, increases and a moisture front begins to form. This front is seen to move towards the specimen's center as desorption progresses resulting therefore, in an increase of its moisture content by about 0.43 percentage points at 380 minutes (Fig. 5), approximately. Beyond that point, the moisture content of the center continuously decreased until it reaches the final equilibrium of 5%. This phenomenon is the result of the steep thermal gradient that existed in the initial stages of the run (Soret effect). It was experimentally proven and theoretically predicted in the past under steady-state and dynamic non-isothermal conditions, respectively (Avramidis, Siau 1987; Avramidis et al. 1992).

The thermal diffusion phenomenon is more evident in Fig. 5 where the moisture content changes at the center and three other locations within the specimen during the initial stages of desorption, are plotted against time. The initial increases and later decreases of moisture are very prominent.

Figure 6 shows the measured as well as the predicted temperatures at the center of the specimen during desorption. It is evident that thermal equilibrium within the specimen was reached much faster than moisture equilibrium. It is apparent from the graph that theory and experiment are in very close agreement particularly in the first 40 minutes and 160 minutes (beginning of thermal equilibrium), until the end of the experiment.

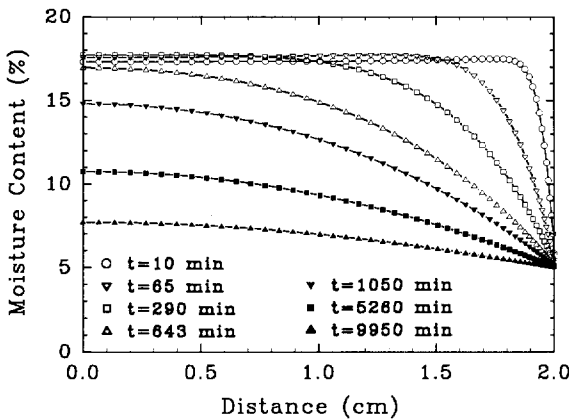


Fig. 4 Predicted moisture profiles within the specimen ( $x = 0$  is the center)

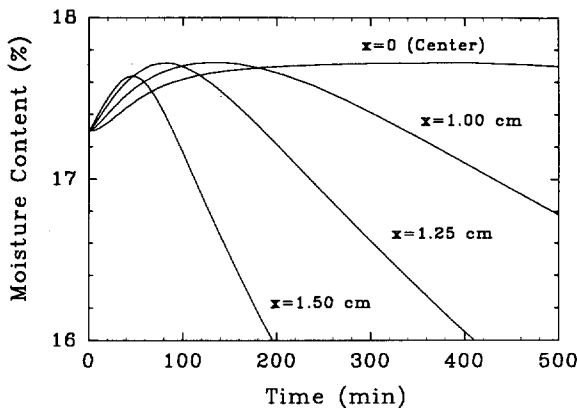


Fig. 5 Predicted moisture contents at four locations within the specimen during the initial desorption stages

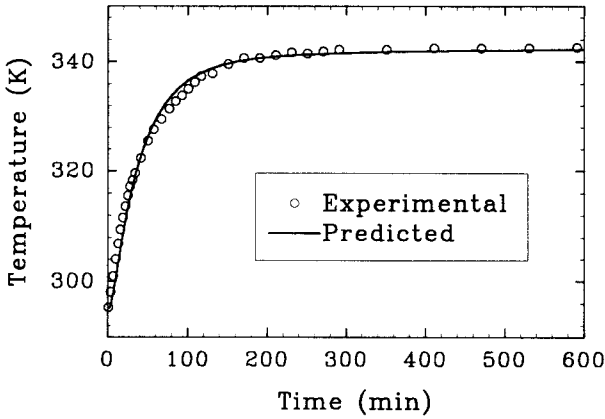


Fig. 6 Experimental and predicted temperatures at the center of the specimen

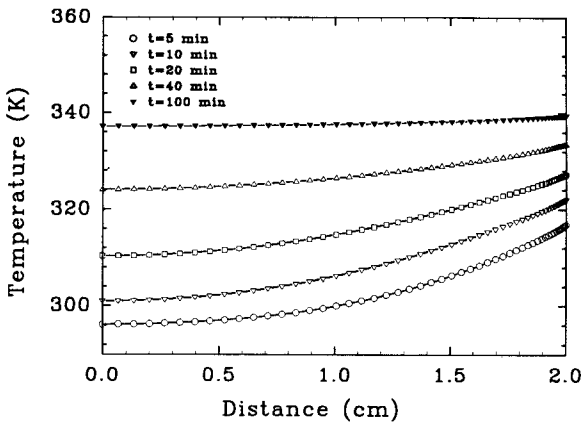


Fig. 7 Predicted temperature profiles within the specimen ( $x = 0$  is the center)

The predicted temperature profiles during the first 100 minutes of desorption, are shown in Fig. 7. Steep temperature gradients are developed in the initial stages of desorption, but the temperature reached its equilibrium profile in a very short period of time.

### Conclusions

A model which predicts heat and moisture transfer through wood under unsteady-state non-isothermal desorption conditions, and whose temperature gradient phenomenological coefficient was based on the principles of nonequilibrium thermodynamics, was described and evaluated with experimental data. A very good agreement was found between the predicted and experimental average moisture contents as well as, the temperature rise at the center of the specimen. The model also predicted a flux of moisture from the surface towards the center of the specimen in the initial stages of the desorption experiment (Soret effect).

### Appendix

The diffusion coefficient for gross wood in the direction transverse to the longitudinal (fiber) axis is given by:

$$D = 0.07 \exp[-(9200 - 70M/RT)/(1 - V_a)(1 - \sqrt{V_a})]$$

where  $V_a$  is the porosity of the material given by the empirical equation:

$$V_a = 1 - 0.46(0.667 + 0.01M)$$

The thermal conductivity for wood is given by:

$$K_T = [0.45(5.18 + 0.096M) + 0.57V_a]E^{-4}$$

and  $c_p$  and  $\rho_a$  are the specific heat and density of wood, respectively, given as

$$c_T = (-0.0323 + 0.0011T + 0.01M)/(1 + 0.01M)$$

$$\rho = G(1 + 0.01M)$$

where oven dry specific gravity ( $G_o$ ) is 0.46. The water activation, vaporization and differential energies are given by:

$$E_o = 10730 + 10(273-T)$$

$$E_b = 9200 - 70M$$

$$E_L = 5000\exp(-0.14M)$$

The slope of the sorption isotherm is given by:

$$\partial M/\partial H = (A + CH^2)/(A + BH - CH^2)^2$$

where

$$A = (W/18)[1/K_2(K_1 + 1)]$$

$$B = (W/1800)[(K_1 - 1)/(K_1 + 1)]$$

$$C = (WK_1K_2)/[180000(K_1 + 1)]$$

$$W = 1591 - 9.4T + 0.0185T^2$$

$$K_1 = -49.74 + 0.32T - 0.000501T^2$$

$$K_2 = -0.176 + 0.001697T - 5.64E - 6T^2$$

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