

**EVALUATION OF MOISTURE DIFFUSION THEORIES IN POROUS
MATERIALS**

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(ABSTRACT)

Moisture transport in building materials is directly responsible for structural damage, as well as poor indoor air quality. For these reasons, the need to understand transfer mechanisms and predict moisture transport through building materials has increased over the last couple of decades.

Although moisture diffusion phenomenon in the isothermal regime has been studied and explained extensively, there is no universally accepted model for predicting the moisture diffusion in a nonisothermal situation. Several diffusion models in the form of "Fick's Law" including ones based on gradients of water-vapor pressure, chemical potential of water, moisture concentration and activated moisture molecules have been proposed for predicting moisture diffusion through porous materials. However, the lack of reliable experimental results, resulting from the complexity of arranging accurate and repeatable measurement techniques and slow moisture movement, has prevented any model from being universally accepted. The present research addresses this modeling problem by evaluating current diffusion models through a series of experiments performed on oriented strand board (OSB), which is a wood-based material.

The present experimental apparatus, developed over the last three years, was designed for the specific purpose of studying and developing an accurate method to measure moisture transfer properties in porous materials under nonisothermal conditions. The apparatus consists of a system of two environmental chambers capable of achieving a wide range of temperatures and relative humidities. Temperature and relative humidity can be independently controlled to within $\pm 0.05^{\circ}\text{C}$ and ± 0.10 per cent R.H. of the set

points. This apparatus is an alternative to the ASTM "cup method" which is limited to isothermal conditions and discrete relative humidities that correspond to those for various saturated salt-in-water solutions. Unlike the cup method, the relative humidity within the chambers is controlled by the direct removal and injection of distilled water. The system has forced recirculating flow which reduces the time to reach steady state. The new forced, direct control measurement procedure is denoted "ASHRAE FDC".

The results obtained from the ASHRAE FDC experiments, show that moisture diffusion under nonisothermal conditions is governed by the gradient of the water-vapor pressure. The moisture transfer must cease when the diffusion potential is the same on both sides of the material for the validation of the diffusion model. The results show that the water-vapor pressure model meets this necessary and sufficient condition. Furthermore, a plot of the diffusion flux versus vapor-pressure difference was linear, within measurement uncertainty bounds. This observation infers that the permeability is approximately constant over the range of temperatures and humidities used in the investigation.

During the ASHRAE FDC experimental procedure a small difference in the static pressure between the chambers was found. This pressure difference which was also observed in ASTM cup tests, is believed to be caused by concurrent air diffusion. The bulk flow of air governed by Darcy's equation balances the diffusion of air in the opposite direction as a result of the gradient in the partial pressure of (dry) air. The air permeability of an OSB specimen was measured and the results presented.

The operation and accuracy of the apparatus was validated by comparing results from a series of isothermal tests to previously published results. The results obtained from the isothermal test allowed the permeability to be compared to results obtained from cup tests during the present investigation and to those previously published using the same method. Good agreement was found between the new data from both FDC and cup experiments and previously published results.

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NOMENCLATURE

A	cross-sectional area (m^2)
c	moisture concentration (kg/kg)
C_p	specific heat ($\text{J/kg}\cdot^\circ\text{C}$)
D	coefficient of proportionality
E_b	activated energy (J/mol)
E_l	difference between the molar heat of vaporization of bound water and free water (J/mol)
E_o	molar heat of vaporization of free water (J/mol)
h	relative humidity (%)
h_v	vapor enthalpy ($\text{J/kmol}\cdot\text{K}$)
K_b	bound water moisture conductivity coefficient
K_m	diffusivity coefficient, based on the chemical potential m
K_{M^*}	coefficient for diffusion of activated moisture molecules
M	permeance ($\text{kg/m}\cdot\text{s}\cdot\text{Pa}$)
M_v	molecular weight of water (kg/mol)
M^*	content of activated moisture molecules in the wood based on oven dry weight
\dot{n}''	moisture flux ($\text{kg/m}^2\cdot\text{s}$)
P_o	standard pressure (kPa)
P_s	static pressure (kPa)
P_{sat}	saturation pressure (kPa)
P_v	water-vapor pressure (kPa)
R	gas constant ($\text{J/mol}\cdot\text{K}$)
R_f	water-vapor resistance ($\text{Pa}\cdot\text{s}\cdot\text{m}^2/\text{kg}$)
RH	relative humidity (%)
s	entropy ($\text{J/mol}\cdot\text{K}$)
s_o	absolute entropy ($\text{J/mol}\cdot\text{K}$)
T	temperature ($^\circ\text{C}$)
T_o	standard temperature ($^\circ\text{C}$)

WVT water-vapor transfer rate ($\text{kg/m}^2\text{-s}$)

Greek

m permeability ($\text{kg/m}^2\text{-s-Pa}$)

m_i air permeability ($\text{kg/m}^2\text{-s-Pa}$)

m chemical potential of water (J/kg)

m_b chemical potential at standard pressure and temperature (J/kg)

f_l glass cup relative humidity (%)

f_u glass vessel relative humidity (%)

g moisture content (kg_w/kg_d)

Dm mass change (kg)

Dt time change (s)

CHAPTER 1

INTRODUCTION

Moisture transport and accumulation in building materials is directly responsible for structural degradation in buildings and also contributes to poor indoor air quality.

Over the last fifty years, a great deal of concern has been placed upon energy use and its conservation. This concern has been driven both by the increase of fossil-fuel prices over the years and by adverse environmental effects. For these reasons, it has become imperative for building operators to be able to control and manage energy use at the highest level. In the effort to improve energy use efficiency, the government has imposed strict energy codes that call for an improvement in the energy performance of buildings. Some of these improvements include a decrease in air leakage through doors and windows and better thermal insulation. The decrease of air infiltration and ventilation within the building can result in an increase in indoor humidity, which is of great importance to human health and comfort. Another consequence of tightly constructed buildings is the possibility of moisture buildup in building components. Decrease in air ventilation may generate moisture buildup along building components. Moisture buildup can lead to the growth of fungus and mold, which in turn can affect the structural integrity of building components. The strength and stiffness of oriented strand-board are substantially reduced as a result of increases in board moisture content, (Wu and Suchsland, 1997).

Moisture diffusion is a phenomenon that has been studied and explored extensively over the last few decades. Moisture diffusion in building materials (wood being among the most common) was believed to be the least expected source of problems in the building industry. However, over the last few decades, it has been determined that extensive failures in building components are often a result of both thermal and moisture

loads, (Chaddock and Todorovic,1991). Failures in many building structures were attributed to the lack of knowledge of the effects of both temperature and humidity in wood. Typically, the building industry has relied on experience for the development and design of new building components. Recently, this traditional design procedure has been updated to include analysis of durability with respect to thermal and moisture loads by laboratory or field experiments. The increased concern of society in general with conservation of energy and human comfort are the main reasons behind the escalation of moisture research.

The purpose of this research is to improve the understanding of moisture diffusion in porous materials under nonisothermal conditions. The analysis of this phenomenon is based upon the evaluation of various existing diffusion models. An experimental apparatus has been designed for this purpose. The experimental apparatus consists of a test specimen and a system of two chambers capable of controlling temperature and relative humidity. The experiments include a series of isothermal and nonisothermal tests. The test specimen consists of a typical hygroscopic building material, known as oriented strandboard (OSB). This material is often used in roof sheathing, I-beam webs, single-layer flooring, and sub-flooring in light-frame construction.

The results obtained from the isothermal tests contribute to validate the operation of the apparatus, while the results from the nonisothermal tests validate the corresponding diffusion models.

The models investigated are diffusion models based on the gradient of water-vapor pressure, chemical potential of water, activated moisture molecules, and moisture concentration. In order for a diffusion model to be valid, the moisture transfer must cease when the diffusion potential is the same on both sides of the specimen.

1.1 MOISTURE DIFFUSION THEORIES

Moisture transfer prediction is a laborious task because of the complexity in devising accurate and repeatable measuring techniques. These difficulties can be attributed mainly to the slow movement of moisture through porous materials, which in turn causes the experimental period to be extensively long. In an attempt to accurately predict moisture diffusion, moisture transport can be categorized in terms of temperature conditions as isothermal and nonisothermal. The first category has been studied extensively since the 1940's. However, a universally accepted model for predicting moisture transfer under nonisothermal conditions does not exist.

1.1.1 Isothermal Moisture Transport

Although most situations encountered by building materials are rarely under isothermal conditions; the majority of moisture transport investigations have been performed at constant temperature.

Diffusion is the flow at a molecular level under the influence of an appropriate property gradient. The steady-state form of Fick's first law states the relationship between the flux of moisture and the concentration gradient. Fick's first law can be expressed as,

$$\dot{n}'' = -D \left(\frac{\partial c}{\partial x} \right) \quad (1.1)$$

where \dot{n}'' is the flux of moisture ($\text{kg}/\text{m}^2 \text{ s}$), $\frac{\partial c}{\partial x}$ is the gradient of concentration c ("driving force") of water in the wood and D is the coefficient of proportionality. The negative sign takes into account that the flux occurs in the direction of decreasing c .

Over the years, a great deal of controversy has surrounded the "driving force" which causes moisture to diffuse across porous materials. A series of driving potentials have been proposed from the very beginning. These models take the form of Fick's law as shown in Table 1.1.

Table 1.1 Tabulation of some moisture transport coefficients used for wood, assumed potentials, and relationships to diffusion coefficient D . (Skaar,1988)

Assumed potential	Symbol	Transport Coefficient	Relationship to D
Moisture Concentration	c	$D = -\dot{n}'' / (\partial c / \partial x)$	
Vapor Pressure	P_v	$m = -\dot{n}'' / (\partial P_v / \partial x)$	$K_p = D / (\partial c / \partial P_v)$
Chemical Potential	m	$K_m = -\dot{n}'' / (\partial m / \partial x)$	$K_m = D / (\partial c / \partial m)$

In the case of isothermal moisture transport, the model most frequently used is based upon the gradient of vapor pressure, and expressed in the form of Fick's first law as,

$$\dot{n}'' = -m \left(\frac{\partial P_v}{\partial x} \right) \quad (1.2)$$

where μ is the moisture permeability of the material, for a given moisture flux rate (\dot{n}'') by virtue of the vapor pressure (P_v) gradient across the sample of thickness x . The permeability of a hygroscopic material is generally a function of the relative humidity and highly nonlinear. Galbraith (1998) argued that vapor permeability results determined under isothermal conditions, can also be used for nonisothermal models.

Several experimental tests have been designed to measure moisture transport under isothermal conditions. The most commonly used test is the ASTM method E 96-80, also known as the "cup" method. A schematic of a typical ASTM cup is shown in Fig. 1.1. The ASTM test method is based on a wet cup and a dry cup. The relative humidity within each cup is maintained constant at a desired value using various aqueous saturated salt solutions.

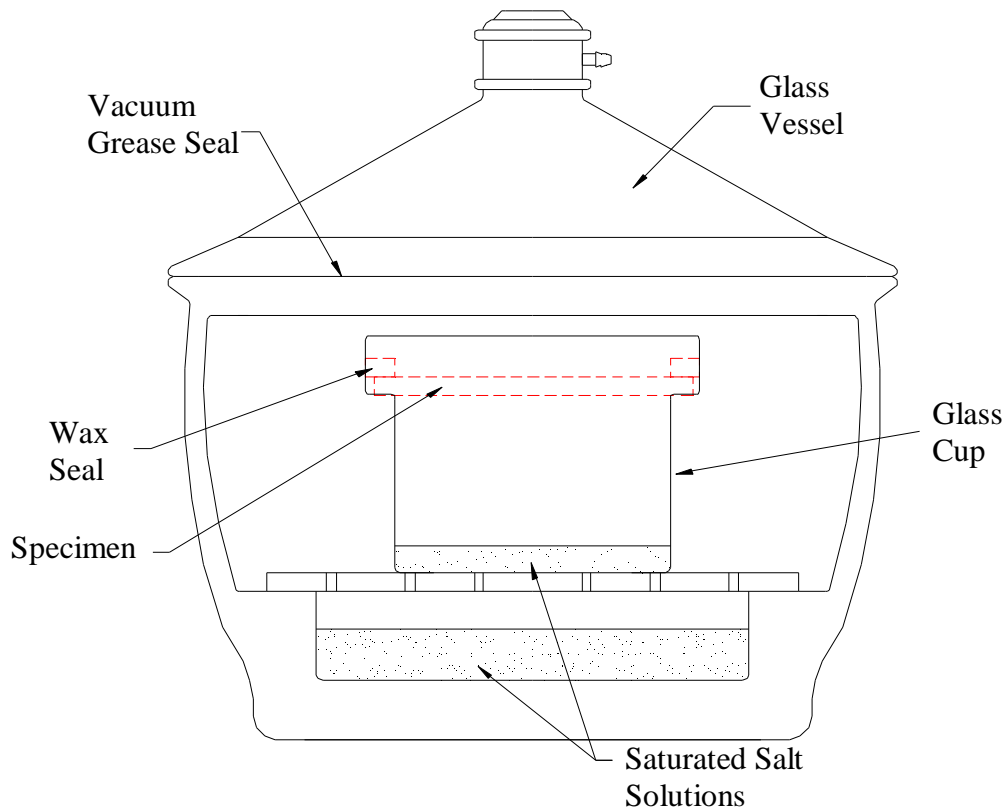


Figure 1.1 ASTM experimental setup.

A modified ASTM method was used in 1991 by Burch and Thomas to obtain permeability data of several building materials. The relative humidities produced by the saturated salt solutions were independently verified by Burch and Thomas (1991) using a precision dew point hygrometer. The saturated salts and the relative humidity corresponding to each of the cups used in the experiment are shown in Table 1.2.

The experiments performed by Burch, Thomas and Fanney (1992) comprised a total of five cup assemblies. These cups covered a range of 11 to 97 per cent relative humidity. The entire system required approximately two weeks to reach steady state conditions. The system was considered to be in steady state, once the moisture gain rate in the cup was constant. The water-vapor transfer rate through the specimen was determined by weighing each cup once a week. The weight of each cup was then plotted as a function of time.

Table 1.2 Relative Humidity and Uncertainties of Saturated-Salt Solutions at 24°C.
(Burch, Thomas and Fanney,1992)

Salt Solution			Relative Humidity (%)		WVT Resistance (10 ⁸ Pa·sec·m ² /kg)
Cup No.	Cup	Vessel	Cup	Vessel	
1	LiCl	KC ₂ H ₃ O ₂	11.30 ± 0.27	22.67 ± 0.32	4.87
2	MgCl ₂	KCO ₃	32.85 ± 0.16	43.16 ± 0.39	5.23
3	NaBr	KCl	57.92 ± 0.40	69.09 ± 0.24	7.58
4	NaCl	KCl	75.33 ± 0.12	84.51 ± 0.26	4.27
5	Sr(NO ₃) ₂	K ₂ SO ₄	85.46 ± 0.38	97.36 ± 0.45	4.39

The water-vapor transfer rate (*WVT*) was then computed directly from the slope of the resulting plot. Permeance is equivalent to the permeability per unit thickness. The permeance (*g*) of the specimen was determined by the following relation,

$$g = \frac{1}{\frac{AP_{sat} [f_u - f_l]}{WVT} - R_f} \quad (1.3)$$

where f_u and f_l are the relative humidity generated by the saturated salts within the glass vessel and the glass cup respectively, and P_{sat} is the saturation pressure at the tested temperature. The cross-sectional area of the specimen is denoted (*A*), and R_f denotes the water-vapor transfer resistance of the two air layers at both sides of the specimens. This resistance causes the relative humidity to vary at the interface with the specimen during moisture transfer process. Burch and Thomas (1990) studied this phenomenon, and their results were used in the present experiment. The water-vapor transfer resistance values used on this experiment are shown in Table 1.2.

Isothermal moisture diffusion is used extensively to obtain information on material properties such as permeance and permeability. This information is extremely valuable to the building and construction industry. This section has been largely inspired by Burch, Thomas and Fanney (1992).

1.1.2 Nonisothermal Moisture Transport

In most cases, building components are exposed to a combination of temperature and humidity gradients; these gradients are directly responsible for moisture diffusion across the material. It is for this reason that nonisothermal moisture prediction is an essential tool in the analysis of building components. Despite many attempts to describe the moisture diffusion mechanism, moisture movement in wood is neither well defined nor fully understood. Many analytical models have been developed over the years, but none entirely accepted. Among the models proposed are those based upon the gradient of chemical potential, activated moisture content, vapor pressure, and moisture concentration.

Chemical Potential

Chemical potential, also known as Gibbs free energy, determines the thermodynamic equilibrium state of a system. A system is said to be in chemical equilibrium if the chemical potentials of its components are equal. Chemical potential is a function of the vapor pressure, temperature, and general composition. For a pure substance, it can be written in terms of the enthalpy (h_v) and entropy (s_v) as,

$$m(P_v, T) = h_v(P_v, T) - Ts_v(P_v, T) \quad (1.4)$$

Entropy in turn is derived using the second law of thermodynamics. For an ideal gas the entropy can be expressed as,

$$s_v(P_v, T) = s^o(P_o, T_o) + \int_{T_o}^T \frac{C_{pv}(T)}{T} dT - R_v \ln \left(\frac{P_v}{P_o} \right) \quad (1.5)$$

where C_{pv} is the specific heat and s^o is the absolute entropy at the reference conditions P_o and T_o .

Equation 1.5 can be simplified assuming the specific heat (C_{pv}) is constant over the interval of interest,

$$s_v(P_v, T) = s_o(P_o, T_o) + C_{pv} \ln\left(\frac{T}{T_o}\right) - R_v \ln\left(\frac{P_v}{P_o}\right) \quad (1.6)$$

where the vapor pressure (P_v) is a function of the relative humidity (f) and temperature (T).

$$P_v = f P_{sat}(T) \quad (1.7)$$

The gas constant is given in terms of the universal gas constant (R) and the molecular weight of water (M_v).

$$R_v = \frac{R}{M_v} = 0.4615 \frac{\text{kJ}}{\text{kg K}} \quad (1.8)$$

This information is used to calculate the chemical potential difference across the material.

Chemical potential models based on irreversible thermodynamics have been proposed both by Siau and Skaar. Siau (1983) stated that when the chemical potential is assumed to be the driving force, Fick's law can be expressed as,

$$\dot{n}'' = -K_m \frac{d\mathbf{m}_v}{dx} \quad (1.9)$$

where K_μ denotes the diffusivity coefficient, based on the chemical potential (\mathbf{m}_v) of water vapor. Since the chemical potential is a function of temperature (T) and relative humidity (f), its gradient can be expressed as,

$$\frac{d\mathbf{m}}{dx} = \left(\frac{\partial \mathbf{m}}{\partial T}\right)_f \frac{dT}{dx} + \left(\frac{\partial \mathbf{m}}{\partial h}\right)_T \left(\frac{\partial h}{\partial f}\right)_T \frac{df}{dx} \quad (1.10)$$

where h denotes the per cent relative humidity in equilibrium.

Equations 1.9 and 1.10 can be combined and simplified by introducing a coefficient K_M , derived from K_μ as a function of the moisture content M .

$$K_m = K_M \left(\frac{\partial M}{\partial \mathbf{m}} \right)_T = K_M \left(\frac{\partial h}{\partial \mathbf{m}} \right)_T \left(\frac{\partial M}{\partial h} \right)_T \quad (1.11)$$

Fick's law can then be written as,

$$\dot{n}'' = -K_M \left[\left(\frac{\partial h}{\partial \mathbf{m}} \right)_T \left(\frac{\partial M}{\partial h} \right)_T \left(\frac{\partial \mathbf{m}}{\partial T} \right)_M \frac{dT}{dx} + \frac{dM}{dx} \right] \quad (1.12)$$

In order to further simplify this form of Fick's law, Siau and Skaar use the definition of the chemical potential of water vapor as,

$$\mathbf{m}(T, P) = \mathbf{m}^\circ(T) + RT \ln \left(\frac{h}{100} \right) \quad (1.13)$$

where \mathbf{m}° denotes the chemical potential of water vapor at one atmosphere and temperature (T). This definition is then used to evaluate the corresponding derivatives:

$$\left(\frac{\partial h}{\partial \mathbf{m}} \right)_T = \frac{h}{RT} \quad (1.14)$$

$$\left(\frac{\partial \mathbf{m}}{\partial T} \right)_M = \frac{d\mathbf{m}^\circ}{dT} + RT \left(\frac{\partial \ln(h/100)}{\partial T} \right)_M + R \ln \left(\frac{h}{100} \right) \quad (1.15)$$

According to Skaar, the second term in Eq.1.15 can be replaced by E_i/T , where E_i is the difference between the molar heat of vaporization of bound water and that of free water (E_o). Siau approximated this difference by:

$$E_i = 6000 \exp(-0.16M) \quad (1.16)$$

Equation (1.12) may be expressed in the form:

$$\dot{n}'' = -K_M \left[\frac{h}{RT} \left(\frac{\partial M}{\partial h} \right)_T \left(\frac{d\mathbf{m}^o}{dT} + \frac{E_l}{T} + R \ln \left(\frac{h}{100} \right) \right) \frac{dT}{dx} + \frac{dM}{dx} \right] \quad (1.17)$$

Equation (1.17) is used by Siau (1983) to predict moisture movement in wood under nonisothermal conditions.

Activated Moisture Molecules

Skaar and Siau (1981) proposed a nonisothermal moisture diffusion model based on the gradient of activated moisture molecules. Once again Fick's law was modified to the form

$$\dot{n}'' = -K_{M^*} \frac{dM^*}{dx} \quad (1.18)$$

where K_{M^*} is the coefficient of diffusion, and M^* is the content of activated moisture in wood based on oven-dry weight (kg_m/kg_d). The activated moisture content can be calculated from the Boltzmann distribution as

$$M^* = \mathbf{g} \exp \left(\frac{-E_b}{RT} \right) \quad (1.19)$$

where \mathbf{g} (kg_w/kg_d) is moisture content and E_b is the activated energy, which can be calculated with the use of,

$$E_b = [38,500 - 290\mathbf{g}] \left(\frac{\text{J}}{\text{mol}} \right) \quad (1.20)$$

This relation gives good values over the moisture range between 5 and 25 per cent. Since activated moisture (M^*) is a function of both \mathbf{g} and T , its gradient can be expressed as,

$$\frac{dM^*}{dx} = \left(\frac{\partial M^*}{\partial T} \right)_{\mathbf{g}} \frac{dT}{dx} + \left(\frac{\partial M^*}{\partial \mathbf{g}} \right)_T \frac{d\mathbf{g}}{dx} \quad (1.21)$$

The partial derivatives can be evaluated assuming that E_b is independent from temperature.

$$\frac{\partial M^*}{\partial T} = \frac{gE_b}{RT^2} \exp\left(\frac{-E_b}{RT}\right) \quad (1.22)$$

and,

$$\frac{\partial M^*}{\partial g} = \left(1 - \frac{g}{RT} \frac{\partial E_b}{\partial g}\right) \exp\left(\frac{E_b}{RT}\right) \quad (1.23)$$

This last formulation can be simplified by using the derivative of Eq.1.20 with respect to the moisture,

$$\frac{\partial M^*}{\partial g} = \left(\frac{RT + 290g}{RT}\right) \exp\left(\frac{-E_b}{RT}\right) \quad (1.24)$$

Fick's law based on activated moisture molecules, as shown in Eq.1.18 may be written as

$$J = -K_M \left[\left(\frac{g}{RT + 290g}\right) \frac{E_b}{T} \frac{dT}{dx} + \frac{dg}{dx} \right] \quad (1.25)$$

where K_M is the coefficient used in Fick's law for isothermal diffusion calculated for a given temperature T and g Moisture content of wood is generally given as a fraction of m or percent M of its dry weight.

$$m = \frac{W_w}{W_o} \quad (1.26)$$

$$g = 100 m \quad (1.27)$$

where the weight of water is W_w and the weight of the dry wood is W_o . The weight of water in the wood (W_w) is obtained from the difference between the moist (W_m) and dry (W_o) weight of the wood (Skaar, 1988).

$$m = \frac{(W_m - W_o)}{W_o} \quad (1.28)$$

An accurate use of this proposed model requires the use of extensive information in the properties of the material being tested, i.e., sorption isotherm which is a function of relative humidity. The sorption isotherm relates the equilibrium moisture content (g) and the relative humidity (f) at a constant temperature for a given type of wood.

$$g(f)|_T \quad (1.29)$$

Another difficulty encountered while using this model lies upon the uncertainty in the value of the activation energy (E_b) for different materials, (Skaar and Siau, 1981).

Water Vapor Pressure

The water vapor pressure model is perhaps the most widely used as a result of its simplicity and practicality. This model takes the form of Fick's law as previously stated in Eq.1.2

$$\dot{n}'' = -m \left(\frac{\partial P_v}{\partial x} \right) \quad (1.2)$$

where the vapor pressure (P_v) is given by the product of the relative humidity (f) and the saturated pressure ($P_{sat}(T)$) at a given temperature. Brahall (1976) reported that this model was the correct driving force for moisture diffusion, and since then numerous articles have been published based on this model.

Moisture Concentration

Many scientists have believed that moisture diffusion as a result of a temperature gradient is significant. However, many still believe that moisture is driven by the

difference in its concentration. In the case of the concentration gradient, Fick's law takes the form of,

$$\dot{n}'' = -K_b \left(\frac{\partial M}{\partial x} \right) \quad (1.29)$$

where K_b is the bound water moisture conductivity coefficient and M is the moisture (Choong, 1963).

1.2 DISCUSSION

The main objective of this chapter was to present a general overview of moisture diffusion phenomenon, as well as, some of the existing predicting models for both isothermal and nonisothermal conditions.

The ASTM standard method presents a simple and reliable method of measuring permeability under isothermal conditions. However, the use of saturated salt solutions and the lengthened experimental procedure makes the "cup" test an inconvenient method. The nonisothermal models proposed by Siau and Skaar are elegantly presented, but further research is needed as a result of their complexity and reliability on material properties. The vapor pressure and moisture concentration models are simple, practical, and the potential or "driving force" is independent of material properties.

The existing apparatus designed and developed over the last several years was used to further analyze moisture diffusion in porous materials and to evaluate the models discussed earlier. This evaluation consisted of creating conditions such that the potential difference across the test specimen corresponding to each model is zero. The models based on Fick's first law require no moisture transfer when the gradient is zero.

A detailed description of the apparatus and its operation is given in the following chapter.