

## 2D Experimental and Numerical Analysis of Hygroscopic Behavior of a Polymer Material

Wael Mokhtar Ph.D., John Witte  
Grand Valley State University, Allendale, MI, 49401  
mokhtarw@gvsu.edu, john.m.witte@gmail.com

### Abstract

A study was completed to improve the ability for the designer to account for two-dimensional hygroscopic effects in design analysis while considering a variety of applicable temperature and humidity environments. A literature review was performed to better understand published ISO and ASTM testing specifications and other established methodologies for determining the necessary material properties for performing hygroscopic analysis. The relevant theory was outlined including a review of field problems, of which both thermal and moisture absorption are a subset of this category. The analogy between the underlying equations describing both thermal and moisture sorption problems was addressed for the purpose of justifying the usage of commercial thermal Finite Element Analysis (FEA) software in simulating moisture sorption problems. A two dimensional experiment and analysis were performed on two polymer materials, 33% glass filled nylon and polypropylene. The FEA results showed a 1.6% discrepancy with regards to mass change due to moisture absorption and a 7.3% discrepancy with regards to hygroscopic expansion in the direction normal to the glass fibers. The FEA software capabilities may be somewhat limited in simulating anisotropic behavior due to hygroscopic effects, particularly if the fiber material is suspected to be arranged in a relatively uniform manner.

### Introduction

A literature review has indicated that the study of hygroscopic mismatch stresses which occur between polymer and other substrate materials has been widely researched for application in the electronics industry. However, when polymer components are used in mechanisms, dimensional variation due to hygroscopic effects may also be of concern as design performance may be affected. At the time of this research, no such documented process has been found to exist which describes the complete process for obtaining material properties and using them to simulate transient dimensional variation for arbitrary shapes in mechanisms as a result of hygroscopic effects.

Some steady state material data regarding moisture uptake are published as a result of testing per ISO62:2008 and ASTM D570. The measured values which are typically published following these experimental guidelines have limited use in the context of this study because they are only valid at the temperature and humidity condition at which the data was taken. Additionally, the maximum moisture uptake at a specific condition is typically the only value presented on the data sheets. In order to simulate hygroscopic effects, additional data is required including the diffusion coefficient and the relationship between moisture uptake and strain.

The goal of this research was to better understand the material properties of a particular polymer material for the purpose of simulating 2-dimensional hygroscopic behavior. The water submersion test procedure was utilized because it avoided the requirement for costly environmental test equipment and greatly reduced the time required to run the experiment and obtain material data. Water submersion conditions typically result in the largest potential moisture uptake, or saturated moisture condition, when compared to ambient conditions. Therefore, the water submersion method also results in the most severe deformation at a saturated moisture condition. Methodology utilized in this study may be repeated with

access to environmental test equipment if a simulation at ambient conditions was determined necessary in future studies.

A literature review of existing research relevant to this study has been performed and is included in this proposal. Moisture diffusion is a subset of a physical phenomenon known as field problems for which the relevant theory has been outlined in the Field Problems section. As part of understanding field problems in general, the correspondence between thermal and moisture analysis was outlined to justify the use of commercial Finite Element Analysis (FEA) software to simulate moisture problems. The relevant theory necessary to develop an analytical model follows the description of field problems in general, which allows for comparison of numerical simulations to direct calculation of ideal moisture uptake.

## Literature Review

As previously mentioned, a number of studies have been performed which pertain to application in the electronics industry. A large amount of effort has been committed to attaining the ability to predict hygroscopic mismatch stresses between various components in an integrated circuit board.

Hsiang-Chen Hsu et al. [1] evaluated the coefficient of moisture expansion by means of Thermal Gravitational Analysis (TGA) and Thermal Mechanical Analysis (TMA) in polymeric packaging materials used in complimentary metal-oxide-semiconductor (CMOS) image sensor image packages (CIS). Diffusivity was determined under Arrhenius behavior for both absorption and desorption. The overall transient moisture distribution was simulated utilizing commercial FEA software. Moiré Interferometry (MI) was also used to compare the experimental deformation to finite element simulation results.

M.H. Shirangi and B. Michel [2] studied the diffusion of moisture in plastic encapsulated devices. Moisture uptake as a function of time was measured and is shown in Figure 4. Samples were dry baked for 24 hours at 125°C and then exposed to 85° C / 85% relative humidity. The test samples consisted of molded discs 100 mm in diameter and 2 mm thick.

M.H. Shirangi and B. Michel also proposed a method for evaluating non-Fickian, or dual stage behavior in polymer materials [2]. It was found that when the  $\sqrt{t}/l$ , or square root of time divided by the thickness of the sample ratio, exceeded 13, the moisture absorption curves for two samples of the same material but different thickness deviated from each other as shown in Figure 3. The curve for the 2 mm thickness sample in Figure 5 displays a prominent feature of most non-Fickian types of diffusion where there is no characteristic equilibrium mass uptake.

One proposed explanation for non-Fickian behavior is the process of relaxation in polymer molecules and/or irreversible reaction between the polymer and moisture such as the formation of hydrogen bonds [2]. Swelling of the polymer matrix can theoretically increase the available empty volume in the material allowing a greater maximum amount of moisture to be absorbed. As the swelling follows a different time based phenomenon when compared to general diffusion, the characteristics of the moisture uptake will tend to show a different pattern. Likewise, if the explanation for the non-Fickian behavior is hydrogen bonding, this transfer of water molecules to a bound state with a different energy level requires overcoming some energy barriers and occurs relatively slowly [2]. Of course, all of these physical means of moisture absorption may occur simultaneously, and it is not until the initial rapid uptake by means of diffusion begins to decrease that the moisture uptake by other means becomes apparent [2].

While materials showing Fickian diffusion behavior may be assumed to have the same diffusion coefficient regardless of sample thickness, variation in the external conditions in which the samples are exposed to will result in a different value. To support the simulation of non-Fickian behavior, M.H. Shirangi and B. Michel calculated the diffusion coefficient over a range of average moisture concentrations as shown in Figure 6. The apparent diffusion coefficient was found to decrease with average moisture concentration. The simulation was then carried out utilizing a transient finite element analysis.

Some types of experimentally measured non-Fickian behavior could be closely modeled using the approach proposed by M.H. Shirangi and B. Michel. The results of this study suggested using a first Fickian diffusion with the diffusion coefficient  $D_1$  and saturated content level  $C_\infty$  is followed by a second Fickian utilizing a second diffusion coefficient  $D_2$  and saturated moisture content  $C_{2\infty}$  at a specific time  $t_2$ . M.H. Shirangi and B. Michel also outline a thermo mechanical analysis and thermo gravimetric (TMA / TGA) analysis approach to experimentally capture material properties.

X.J. Fan and S.W.R. Lee found that saturated moisture concentration was a measure of the moisture content under given temperature and humidity conditions [2].  $C_{sat}$  in the studied polymers was found to be linearly proportional to the ambient relative humidity. For most polymer materials,  $C_{sat}$  was found to be temperature independent below the glass transition temperature. Above the glass transition temperature, the  $C_{sat}$  for the studied samples showed strong dependency on temperature.

The porosity, called the free volume fraction, in polymers was found to be a critical material property related to moisture absorption. The volume of a sample of polymer material may be divided into three elements: occupied (van der Waals) volume, interstitial free volume, and hole free volume. Macroscopic changes to the material volume are largely governed by the hole-free volume. X.J. Fa and S.W.R. Lee also found that the free volumes, which are occupied by the absorbed moisture, are typically in the nanometer range [2]. Moiré interferometry (MI) was proposed as a method for analyzing hygroscopic strain. A separate control sample was used which was exposed to constant thermal and humidity conditions to determine if strain due to thermal aging was a factor in the hygroscopic strain measurement.

Onur Sayman [3] presented an analytical approach to performing a general stress analysis for thick or thin multi-layered composite cylinders under hygrothermal loading. The composite epoxy-glass fiber materials studied showed different mechanical properties in the principal material directions. Analytical calculations were compared to FEA simulations.

Weide-Zaage et al. [4] studied moisture diffusion in printed circuit boards. Concentration distribution was compared based on FEA simulations, taking into consideration the effects of copper layers acting as a barrier to diffusion. Hygroscopic stress which develop as a result of unequal moisture saturation levels between laminated layers were studied because of the effect of these stresses on delamination which occurs during the soldering process.

Derrien and Gilormini [5] were able to show that a composite may have a nonlinear sorption isotherm even though the polymer matrix has a linear one, as a result of the internal stresses induced by swelling. The composite reinforcing material is assumed to not absorb moisture and consequently not swell. As a result, the composite material will tend to absorb less moisture than a comparable sample composed purely of the matrix material for two reasons. First, the reinforcing material takes up a component of the volume and does not absorb moisture. Second, the reinforcements induce internal stresses that restrict the potential absorption of the matrix material.

Poenninger and Defoort [6] described a test facility for the measurement of the length variation of polymer composites. Commercial laser interferometers with a resolution of 10 nm working under vacuum conditions were found to yield a total accuracy of 0.1  $\mu\text{m}$ . The length variation was determined by using two laser interferometers. Each laser was focused on a reflector attached to opposing ends of the sample.

Celik et al. [7] studied non-Fickian diffusion in electronic packages. Non-Fickian diffusion may be observed in polymer materials due to high relative humidity and temperature of the environment. A power law relation was presented to describe the variation of diffusivity in substances as a function of average moisture concentration. This variable coefficient of diffusion was utilized in commercial FEA software to simulate moisture absorption in materials found to exhibit this behavior.

Yoon et al. [8] proposed a technique for analyzing the combined hygroscopic and thermo-mechanical effect with FEA based simulations and experimental validation using Moiré Interferometry. The study was performed to investigate the mechanisms of popcorn cracking and delamination in plastic encapsulated microelectronics.

Yu [9] presented a study of moisture diffusion in polymeric packaging materials at high temperatures. A comparison of conventional testing technique TGA was compared with Gas Chromatography/Mass Spectrometry (GC/MS) as well as Karl Fisher Titration (KFT) on molding compound, underfill, and die attach materials used in the electronics industry. Results obtained using TGA were found to be affected by the outgassing of solvent at high temperatures where Karl Fischer Titration was found to be more reliable in this application.

Hu et al. [10] studied the effect of thermal and hygroscopic stresses on delamination in Light Emitting Diode (LED) packages. Commercial FEA software was used to simulate the moisture absorption in an electronics assembly and correlated to a micrograph of a delaminated lead frame.

Zhang et al. [11] performed hygro-thermo-mechanical modeling and testing for an electronics reliability application. Dynamic Mechanical Analyzer (DMA), TMA, and TGA techniques were used to determine material properties.

Li et al. [12] presented an analytical model for effective mass diffusivity in composites. Square and hexagonal arrays of cylindrical filler materials were considered. The models allowed for the prediction of mass diffusivity based on geometry and fiber volume fraction.

Wei et al [13] studied the hygroscopic dimensional changes in resin-matrix composites during water sorption/desorption cycles. Disk shaped specimens with a 15 mm diameter and 2 mm thickness were prepared according to ISO 4049. The mean diameter of each specimen was measured with a custom laser micrometer as shown in Figure 9 with a resolution of 200 nm. This method was found to be highly accurate for measuring dimensional changes of material specimens over time.

A preliminary study was completed by Mokhtar and Witte to show that the use of transient thermal and static structural FEA could be combined with available measurement techniques to produce acceptable results, prior to studying the automotive joint in greater depth [14]. The results were found to be acceptable and further study was carried out.

### Field Problems and Formulation of Thermal Moisture Analogy

Moisture diffusion is considered a field problem. Other examples of engineering field problems include torsion of a non-circular section, ideal irrotational fluid flow, seepage, heat transfer and electrostatic potential [15]. The general field equation which may be used to describe these types of problems is given as:

$$D_x \frac{\partial^2 \phi}{\partial x^2} + D_y \frac{\partial^2 \phi}{\partial y^2} - A\phi + B = 0 \quad [1]$$

where  $\phi$  is the field variable and  $D_x$ ,  $D_y$ ,  $A$ , and  $B$  represent physical properties which are specific to the field problem being analyzed. For example a two-dimensional, steady state thermal problem with no internal heat generation can be represented by the equation:

$$D_x \frac{\partial^2 T}{\partial x^2} + D_y \frac{\partial^2 T}{\partial y^2} = 0 \quad [2]$$

where  $T$  is the temperature and  $D_x$  and  $D_y$  represent the thermal conductivity in the  $x$  and  $y$  directions, respectively. In a three-dimensional case, the transient heat conduction through a material may be modeled with the following [1]:

$$\frac{\partial T}{\partial t} = \alpha_T \left( \frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} + \frac{\partial^2 T}{\partial z^2} \right) \quad [3]$$

Where  $T$  is the absolute temperature,  $x$ ,  $y$ ,  $z$  are the spacial coordinates,  $t$  is the time, and  $\alpha_T$  is the thermal diffusivity. The thermal diffusivity is calculated with:

$$\alpha_T = \frac{k}{\rho c} \quad [4]$$

where  $k$  is the thermal conductivity,  $c$  is the specific heat, and  $\rho$  is the density of the material.

### General Diffusion

Diffusion of moisture in polymer materials obeying Fick's Second Law is described by the following equation:

$$\frac{\partial C}{\partial t} = D \left( \frac{\partial^2 C}{\partial x^2} + \frac{\partial^2 C}{\partial y^2} + \frac{\partial^2 C}{\partial z^2} \right) \quad [5]$$

where  $C$  is the moisture concentration and  $D$  is the moisture diffusivity calculated with the Arrhenius equation:

$$D = D_0 e^{-\frac{E_d}{kT}} \quad [6]$$

where  $D_0$  is the diffusivity coefficient,  $E_d$  is the activation energy,  $K$  is the Boltzmann constant, and  $T$  is the absolute temperature. While equation [5] is adequate for a single material analysis, moisture problems often require the consideration of multi-material interfaces. Because the concentration potential is not identical between materials, a wetness field variable is utilized to avoid differential difficulty and to allow for a single equation to describe the problem:

$$\frac{\partial w}{\partial t} = D \left( \frac{\partial^2 w}{\partial x^2} + \frac{\partial^2 w}{\partial y^2} + \frac{\partial^2 w}{\partial z^2} \right) \quad [7]$$

where  $w$  is the wetness defined as:

$$w = \frac{C}{C_{sat}} \quad [8]$$

where  $C$  is the moisture concentration at a particular point and  $C_{sat}$  is the saturated moisture concentration of the material under the specific conditions being analyzed.

## Fickian versus Non-Fickian Diffusion

Yu [9] presented the four models which are shown in Figure 1 regarding the characteristics of moisture absorption, Fickian, Sigmoidal, Two Stage, and Case II. ISO 62:2008(E) describes procedures for verifying Fickian absorption, but the key recognizable characteristics may include by the following [9]:

1. Mass of absorbed moisture versus time<sup>1/2</sup> is initially linear.
2. As time increases, the absorption curve levels to a saturated level of moisture content.
3. A plot of  $M_t/M_{sat}$  versus  $t^{1/2}/L$  is identical for different values of material thickness.

Sigmoidal sorption may occur when a material that would typically display Fickian behavior is exposed to a variable surface concentration. This indicates that the actual transport through the material is Fickian, but the initial curve is not linear due to the slow establishment of a constant boundary condition at the surface of the material.

Two stage sorption consists of two distinct segments, the first composed of fast Fickian absorption, the second is composed of slow non-Fickian absorption. Instead of leveling off at the completion of the Fickian absorption, the curve continues to show increased moisture absorption with time. This behavior is typically attributed to diffusion-relaxation, resulting in increased moisture holding capacity of the material. Case II sorption is essentially a linear model which may be accompanied by a significant amount of material swelling.

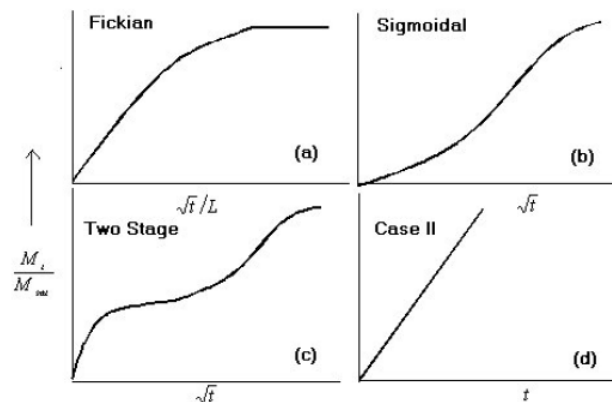


Figure 1: Characteristic curves for (a) Fickian, (b) Sigmoidal, (c) Two Stage, and (d) Case II [9]

## 1D Moisture Diffusion

For a one-dimensional case involving a single material, equation [5] may be reduced to:

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} \quad [9]$$

Figure 2 describes a one-dimensional case where a material of thickness  $L$  is exposed to a moisture concentration at the boundaries  $X = L/2$  and  $X = -L/2$ .

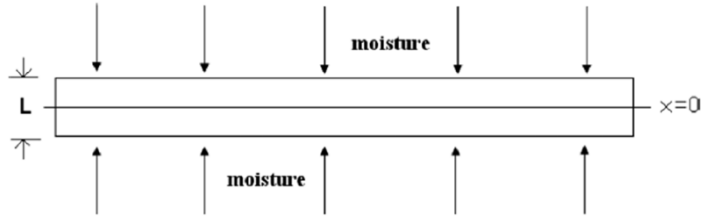


Figure 2: One-dimensional moisture diffusion in an isotropic material [1]

The initial and boundary conditions for the problem may be described as:

Initial Condition:

$$C = 0, \text{ for } -\frac{L}{2} \leq x \leq \frac{L}{2} \quad [10a]$$

Boundary Condition 1:

$$\frac{\partial C}{\partial x} = 0, \text{ at } x = 0, t \geq 0 \quad [10b]$$

Boundary Condition 2:

$$C = C_{\infty}, \text{ at } x = \frac{L}{2} \text{ and } x = -\frac{L}{2}, t \geq 0 \quad [10c]$$

Using Laplace transformation method and separation variable method, equation [9] may be expressed as:

$$C(x, t) = C_{\infty} - \frac{4C_{\infty}}{\pi} \sum_{n=0}^{\infty} e^{\left(\frac{-D(2n+1)^2\pi^2 t}{L^2}\right)} \cos\left(\frac{(2n+1)\pi x}{L}\right) \quad [11]$$

where the moisture concentration at any particular depth of the on-dimensional moisture diffusion model may be determined. Integrating equation [11] yields:

$$\frac{M_t}{M_{\infty}} = 1 - \sum_{n=0}^{\infty} \frac{8}{(2n+1)^2\pi^2} e^{\left(\frac{-D(2n+1)^2\pi^2 t}{l^2}\right)} \quad [12]$$

where the absolute weight gain of the sample may be calculated as a function of time. The use of 20 summands is generally considered sufficient [17].

### Determination of Diffusion Coefficient (ISO)

ISO 62:208(E) and ASTM D570 address the required practices to conduct a controlled experiment for the purpose of evaluating moisture absorption in polymer materials. The standards each have a slightly different description of the required test sample size and experimental procedures. A brief summary is included but the standards themselves should be reviewed prior to conducting sample testing.

## ISO 62:2008(E)

The ISO standard was developed for the purpose of determining the equilibrium moisture content in polymer materials as a result of being exposed to humidity, immersion, and boiling water. The standard also provides guidelines for determining the diffusion constant of a material under carefully controlled non-equilibrium conditions. For non-equilibrium analysis, this standard assumes the Fickian model as valid for homogeneous materials and for reinforced polymer-matrix composites tested below their glass transition temperature. The standard is not applicable to cellular plastics, granulates, or powders which can show additional absorption and capillary effects.

## ASTM D570

The ASTM standard is similar in nature to ISO 62:2008(E). The purpose of this standard is to determine the relative rate of absorption of water at specific conditions, and the steady state concentration of moisture at saturation [18].

### *Method 1*

ISO 62:2008(E) states that at temperatures well below the glass transition temperature of the wet polymer, the water absorption of most polymers correlates well with Fick's laws and a water diffusion coefficient independent of time and concentration can be determined. To verify that a polymer follows Fickian diffusion behavior, the experimental data must be taken up to the saturation concentration [17]. The diffusion coefficient may be calculated with equation [12] at the time required for 70% moisture concentration, or  $t_{70}$ .

### *Method 2*

The initial stage of moisture absorption, meaning when the moisture concentration is less than 50% of the saturated moisture concentration, can be determined by simplifying equation [12] to the following [2]:

$$\frac{M_t}{M_\infty} = 4 \left( \frac{Dt}{\pi l^2} \right) \quad [13]$$

where  $M_t$  is the moisture concentration at the time of interest,  $M_\infty$  is the saturated moisture concentration,  $D$  is the diffusion coefficient, and  $l$  is the thickness of the sample.

### *Method 3*

Two applicable dimensionless numbers may be used to fit empirical data with ideal Fickian diffusion. The dimensionless values are given by:

$$\varphi = \frac{D\pi^2 t}{d^2} \quad [14]$$

where  $\psi$  is the dimensionless value,  $D$  is the diffusion coefficient,  $t$  is the current time, and  $d$  is the sample thickness. The second dimensionless value is the wetness calculated with equation [8]:



The dimensionless values for ideal Fickian diffusion are listed in ISO62:2008(E). These values may also be utilized as described in the ISO standard to validate the agreement of the experimentally measured material behavior with Fick's diffusion laws.

### Thermal Moisture Analogy

Because moisture diffusion through a material may be described as mathematically similar to thermal diffusion, the correspondence of material properties which are relevant when considering these behaviors may be highlighted. A comparison of various physical properties is shown in Table 3.

In a thermal analysis as described by equation [3], the primary variable is temperature which represents the potential for heat transfer whenever a thermal gradient exists. The density, thermal

Table 3: Corresponding material properties for thermal moisture analogy

Property	Thermal Analysis	Hygroscopic Analysis
Primary Variable	T	Wetness, w
Density	$\rho$	1
Thermal Conductivity	k	$D \cdot C_{sat}$
Specific Heat	c	$C_{sat}$
Coefficient of Thermal Expansion	$\alpha_{te}$	$\beta \cdot C_{sat}$

conductivity, and specific heat are all utilized in equation [4] to determine the thermal diffusivity of the material. A relatively high thermal conductivity, low specific heat, or low density will result in a high thermal diffusivity as shown by equation [4] and will result in heat being more readily transferred through a particular medium. The coefficient of thermal expansion describes the linear relationship between material temperature change from a starting reference point and the resulting relative strain.

Comparatively, in a hygroscopic analysis the primary variable is the wetness. Equation [7] describes the behavior of moisture sorption. Although the true driving potential through a material is the moisture concentration gradient, the absolute moisture absorption potential at a specific humidity and temperature condition is not the same for all materials. Therefore, to allow for a multi-material analysis the moisture concentration is normalized by the absolute moisture potential. In a hygroscopic analysis, the density has no physical meaning so a value of 1 is used in the field equation. This is in contrast with a thermal analysis where all parts in an assembly have equal potential to reach the external temperature.

Because the wetness is used, the diffusivity must be multiplied by the saturated moisture concentration of the material at the external temperature and humidity conditions of interest. This value is comparable to the thermal conductivity through a material. The saturated moisture concentration is also used in place of the specific heat for material, which when comparing to equation [4] essentially means that the diffusivity is the only factor driving the rate of moisture diffusion. A large diffusivity will indicate that the moisture will be absorbed at a relatively faster rate than a material which exhibits a low diffusivity.

Similar to the coefficient of thermal expansion, hygroscopic strain may be calculated using a coefficient of hygroscopic expansion. The amount of hygroscopic strain is generally assumed to be linearly proportional to the moisture concentration of a material and may be expressed as:

$$\varepsilon_h = \beta C \quad [13]$$

where  $\varepsilon_h$  is the hygroscopic strain,  $\beta$  is the coefficient of hygroscopic strain, and  $C$  is the moisture concentration.

Water molecules may be found in polymer materials in either the unbound state, where the water molecules occupy voids and nanopores, or in the bound state where the water molecules react with polymer chains. Current belief is that the hydrogen bonding state is the primary physical cause for hygroscopic swelling [2]. This is partially implied by the experimental observation that the ratio of hygroscopic volume expansion to the volume of absorbed water is typically measured to be less than 1 [2].

The diffusion coefficient shall be determined using two applicable dimensionless numbers to fit empirical data with ideal Fickian diffusion. The dimensionless values are given by:

$$\varphi = \frac{D\pi^2 t}{d^2} \quad [16]$$

where  $\psi$  is the dimensionless value,  $D$  is the diffusion coefficient,  $t$  is the current time, and  $d$  is the sample thickness. The second dimensionless value is the wetness calculated with equation [8]:

The dimensionless values for ideal Fickian diffusion are listed in ISO62:2008(E). These values may also be utilized as described in the ISO standard to validate the agreement of the experimentally measured material behavior with Fick's diffusion laws.

## 2D Rectangular Sample Experimental Results and Simulation

The outline of the CAD model and a photo of the samples used in this experiment are shown in Figures 12 (a) and (b), respectively. A 1 mm notch was placed at the center of each edge to ensure that the periodical measurement points were repeatable for length and width.

The eight glass-filled nylon samples were submerged in 23° C water for a duration of 530.5 hours. The experiment termination time was determined by plotting the mass versus log time and determining that the absorption behavior was no longer linear on this scale as shown in Figure 13. The experimental versus published values for saturated moisture content of the glass-filled nylon are shown in Table 4.

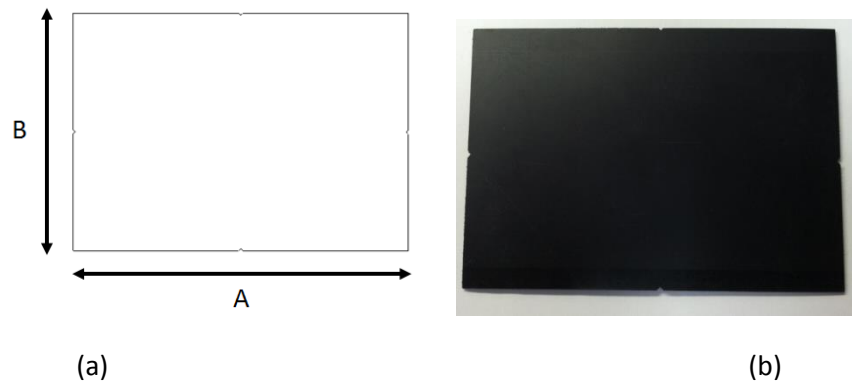


Figure 12: (a) Full CAD model and (b) photograph of experimental sample with notched edges for measurement point indication

Table 4: Experimental versus published saturated moisture content for glass-filled nylon material

	Moisture Absorption (Percent Dry Mass)
Experimental	5.88%
Published	5.70%

The mass versus time for each of the individual glass-filled nylon samples versus the average mass is shown for reference in Figure 14. As can be seen by the individual results, the individual samples behaved very similar to the average thus the usage of the cumulative mass to determine material properties and ensure best use of measurement scale resolution was appropriate. Figures 13 and 14 also indicated that the glass-filled nylon followed Fickian moisture absorption as there was no secondary moisture uptake phase following the initial Fickian behavior as described by Yu [9].

The dimensional change versus moisture concentration for the glass-filled nylon samples is shown in Figure 15. The material clearly behaved in a non-isotropic manner which was not surprising for a glass filled composite. Oner [3] and Derrien and Gilormini [5] indicated that hygroscopic behavior in composite materials may be affected by the reinforcing material. As shown in the plot, the coefficient of hygroscopic expansion was found to be  $0.161 \text{ mm}^3/\text{mg}$  in the longer “A” direction of the rectangle and  $0.017 \text{ mm}^3/\text{mg}$  in the shorter “B” direction. A second submersion experiment was performed with the samples cut from the parent material at a  $90^\circ$  orientation relative to the original experiment. These samples showed similar behavior indicating that the sample shape was not influencing the difference in expansion.

The moisture absorption properties were determined by fitting the experimental data to the dimensionless curve for ideal Fickian behavior. As shown in Figure 16, the parameters used in Table 5 allowed for close correlation with ideal Fickian behavior.

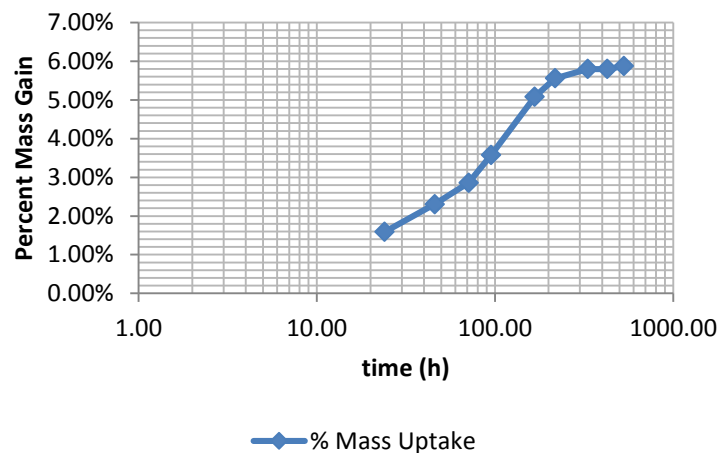


Figure 13: Moisture gain versus logarithmic time plot for glass-filled nylon indicating that samples reached full saturation

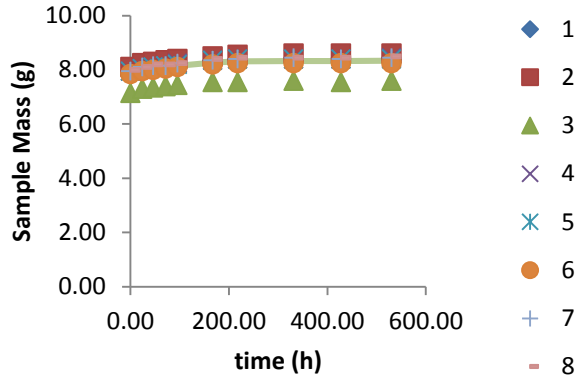


Figure 14: Mass versus time for each of eight glass filled nylon samples

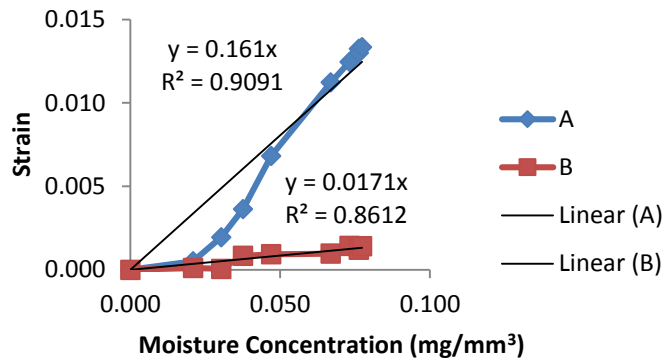


Figure 15: Determination of  $\beta$  for longer dimension "A" and shorter dimension "B" on rectangular sample

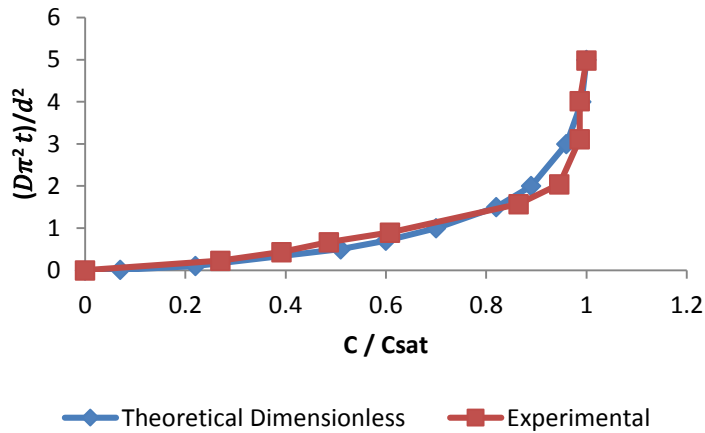


Figure 16: Glass filled nylon material dimensionless parameters plotted against theoretically ideal Fickian absorption parameters

Table 5: Experimentally determined hygroscopic parameters for glass filled nylon

$\beta$ (mm <sup>3</sup> /mg)	0.161
D (mm <sup>2</sup> /s)	3.20E-07
C <sub>sat</sub> (mg/mm <sup>3</sup> )	0.07734

The ¼ symmetry FEA model which was used to simulate the glass-filled nylon material is shown in Figure 17. A comparison of the experimental and simulated moisture uptake versus time is shown in Figure 18. As can be seen in Figure 18, the simulation closely approximated the experimental mass uptake, with an error of 1.6% at 511.4 hours of exposure. The moisture contours at 50 and 530 hours are shown in Figures 19 and 20, respectively.

The isotropic hygroscopic expansion is shown in Figure 21 and the directional deformation which corresponds to the experimental measurements is shown in Figure 22. The comparison between FEA and experimental expansion in the “A” direction is shown in Table 6, with a 7.3% discrepancy between results. The coefficient of hygroscopic expansion was determined by the “A” direction growth so these values agreed well with the experimental data. The predicted deformation in the “B” direction as indicated by Figure 21 did not compare well to the experimental 0.085 mm of measured growth. This was not expected to match well as a comparison between  $\beta$  values for the A and B directions of the glass filled nylon material (Figure 15) clearly indicated the anisotropic behavior.

Table 6: FEA and experimental hygroscopic expansion in “A” direction

FEA (mm)	1.056
Experimental (mm)	1.139
Discrepancy	7.3%

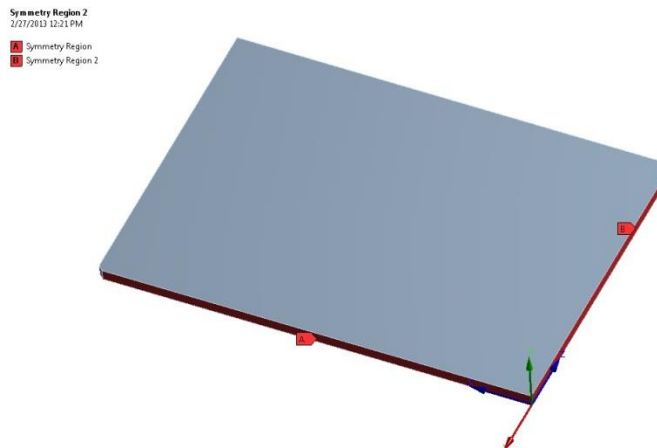


Figure 17: ¼ symmetry model showing symmetry planes

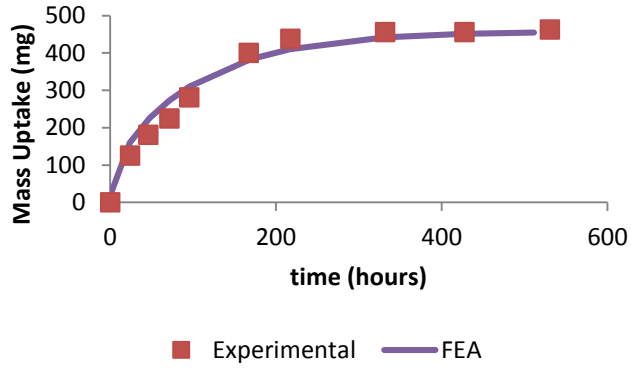


Figure 18: 2D Experimental moisture uptake versus FEA simulation

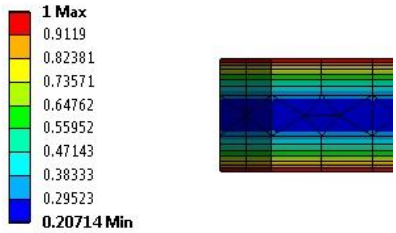


Figure 19: Wetness at 50 hours

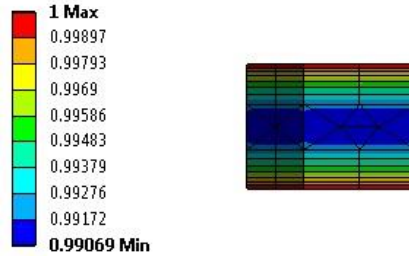


Figure 20: Wetness at 530 hours

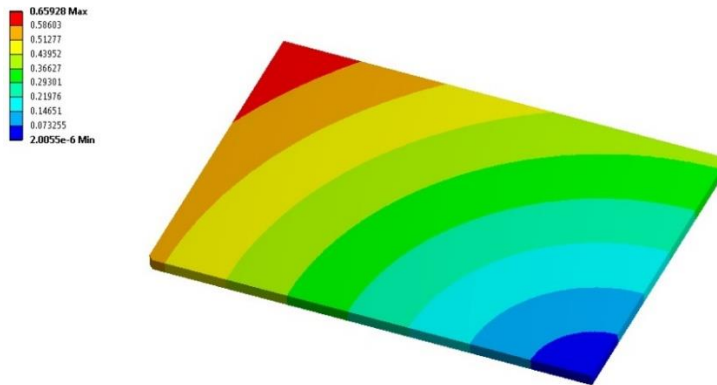


Figure 21: 1/4 symmetry model showing isotropic deformation



Figure 22: 1/4 symmetry model directional deformation at point of experimental measurement

## Conclusion

A literature review was performed to better understand the mechanism of moisture diffusion and to research established testing methods. Several research examples were identified where moisture diffusion was studied in electronic packages as related to delamination of assembled components, but none were found to be specifically aimed at analyzing dimensional change that could potentially affect the performance of mechanism interfaces such as sliding joints, cams, or pivots. Some published steady state material data was identified but it was determined that there was not enough relevant information available to calculate the necessary parameters for defining material properties in a hygroscopic FEA. The guidelines for published testing procedures were taken into consideration when designing the experiments that were used to determine hygroscopic material properties. Developing the ability to conduct experiments to determine material properties was crucial due to the fact that the saturated moisture content and diffusion coefficient are environmentally dependent, and need to be measured for the specific conditions that the designer intends to simulate on more complex geometries.

A published testing standard for determining the coefficient of hygroscopic expansion was not identified. However, existing research describing this procedure was found and replicated. The results from the two-dimensional FEA simulation showed that the procedures used to determine this value allowed for very accurate correlation between simulation and experimental results for hygroscopic strain. The nylon material was tested to full saturation as verified by a plot of percent moisture content versus the log of time. The experimentally determined saturated moisture content was comparable to the published value for the material under identical conditions.

The analytical model describing ideal Fickian moisture absorption was discussed and found to be suitable for calculating diffusion in a wide range of polymer materials. A comparison of the analytical model to the mathematical formulation for thermal FEA software was used to justify the adaptation of thermal FEA software for simulating hygroscopic behavior.

Due to the suspected fiber direction in the glass-filled nylon material, the hygroscopic strain was found to be highly anisotropic with most material growth only occurring in one direction. A hygroscopic FEA was run using the material properties determined from the two-dimensional glass-filled nylon samples. This simulation was found to correlate very closely with experimental data in terms of mass gain and expansion in the direction of primary growth, with 1.6% discrepancy with respect to mass gain and 7.3% discrepancy with respect to hygroscopic expansion. Further experimental procedures indicated that the rectangular shape of the samples was not the cause of the anisotropic behavior.

The FEA software capabilities were found to be somewhat limited in modeling anisotropic hygroscopic growth, particularly if the fiber material is suspected to be arranged in a relatively uniform manner. The FEA and experimental method did provide reasonable results for isotropic behavior, and also for anisotropic behavior in the direction for which  $\beta$  was determined.

Further study should seek to obtain material properties in a humidity controlled environment instead of the worst case water submersion conditions used in this research. There would also be useful value in proceeding with further investigation of the relationship between composite fiber direction and the anisotropic coefficient of hygroscopic expansion.

## References

1. Hsu, Hsiang-Chen, Hui-Yu Lee, Yu-Cha Hsu, and Shen-Li Fu. "Thermo-Hygro-Mechanical Design and Reliability Analysis for CMOS Image Sensor." *Journal of Thermal Stresses* 31:22 Apr. (2008): 917-34.

2. Fan, X.J., and E. Suhir, eds. *Moisture Sensitivity of Plastic Packages of IC Devices*. New York: Springer Science + Business Media, 2010.
3. Sayman, Onur. "Analysis of Multi-Layered Composite Cylinders under Hygrothermal Loading." *Composites Part A* 36 (2005): 923-33.
4. Weide-Zaage, Kirsten, Walter Horaud, and Helene Fremont. "Moisture Diffusion in Printed Circuit Boards: Measurements and Finite Element Simulations." *Microelectronics Reliability* 45 (2005): 1662-67.
5. Derrien, K., and P. Gilormini. "The Effect of Applied Stresses on the Equilibrium Moisture Content in Polymers." *Scripta Materialia* 56 (2007): 297-99.
6. Poenninger, A., and B. Defoort. "Determination of the Coefficient of Moisture Expansion (CME)." *Proceedings of the 9th International Symposium on Materials in a Space Environment* (2003).
7. Celik, Emrah, Ibrahim Guven, and Erdogan Madenci. "Experimental and Numerical Characterization of Non-Fickian Moisture Diffusion in Electronic Packages." *Electronic Components and Technology Conference* (2007): 1069-73.
8. Yoon, Samson, Bongtae Han, Seungmin Cho, and Chang-Soo Jang. "Experimental Verification of Non-Linear Finite Element Analysis for Combined Hygroscopic and Thermo-Mechanical Stresses."
9. Yu, Shi. "A Study of Moisture Diffusion in Polymeric Packaging Materials Especially at High Temperatures." *Graduate Thesis* (2002).
10. Hu, Jianzheng, Lianqiao Yang, and Moo Whan Shin. "Mechanism and Thermal Effect of Delamination in Light-Emitting Diode Packages." *Tima Editions / Therminic* (2005): 249-54.
11. Zhang, Xueren, Tong Yan Tee, Hun Shen Ng, Jerome Teyseyre, and Shane Loo, Subodh Mhaisalkar, Fong Kuan Ng, Chwee Teck Lim, Xinyu Du, Eric Bool, Wenhui Zhu, Spencer Chew. "Comprehensive Hygro-thermal-mechanical Modelling and Testing of Stacked Die BGA Module with Molded Underfill." *Electronic Components and Technology Conference* (2005): 196-200.
12. Li, Shoujie, James Lee, and Jose Castro. "Effective Mass Diffusivity in Composites." *Journal of Composite Materials* 36.14 (2002): 1709-24.
13. Wei, Yong-jie, Nick Silikas, Zhen-ting Zhang, and David C. Watts. "Hygroscopic Dimensional Changes of Self-Adhering and New Resin-Matrix Composites During Water Sorption/Desorption Cycles." *Dental Materials* 27 (2011): 259-66.
14. Mokhtar, Wael, and John Witte. "Experimental and Numerical Analysis of Hygroscopic Behavior of a Polymer Material." *ASEE 2013 North Central Conference* (2013).
15. Madenci, Erdogan, and Ibrahim Guven. *The Finite Element Method and Applications in Engineering Using ANSYS*. New York: Springer Science + Business Media, 2006.
16. Samson Yoon, Bongtae Han and Seungmin Cho, Chang-Soo Jang "Experimental Verification of Non-Linear Finite Element Analysis for Combined Hygroscopic and Thermo-Mechanical Stresses." *Research Thesis* (2002).
17. ISO. *ISO 62:2008(E)*. International Organization for Standardization, 2008.
18. ASTM. *ASTM D570*. American Society for Testing and Materials, 2010.