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Theoretical Evaluation of Moisture Protection Using Coatings

by J. F. Newill, S. H. McKnight, C. P. R. Hoppel,
G. R. Cooper, and M. S. Berman

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Abstract

A theoretical approach is used to determine important attributes for moisture protection of polymer-matrix composite structures provided by polymeric coatings. Developed solutions are used to evaluate a range of coating materials and thicknesses for thin and thick substrates. The diffusion constants and the saturation levels for the coating material were obtained from the range of available coating materials. The results include typical diffusion patterns for coating materials, saturation of the coating layer, development of moisture through time, and effects of the material properties for the various substrate thicknesses. Moisture diffusion behavior at the substrate-coating interface is also presented. For relatively thick coating layers, the results show that the diffusion constant and the saturation level of the coating must both be low to significantly affect the diffusion process. The study also shows differences in the behavior of the substrate-coating interface.

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1. Introduction

While most polymers are considered “water proof,” that is only partially true in the context of liquid water flowing through the material. However, it is widely recognized that individual water molecules or clusters can be readily absorbed into polymers via diffusion mechanisms. There have been many studies and theories that relate to polymer structure and moisture diffusion behavior. These works have sought to understand the diffusion process from a fundamental level and have provided insight into the mechanisms for moisture diffusion. These works have also guided appropriate simplifications of the diffusion processes in polymers to enable readily usable models to predict moisture transport. This understanding and predictive capability for moisture diffusion in and through polymers is important in a number of materials applications including polymer-matrix composites.

It is understood that combinations of temperature and high humidity cause problems with the performance of plastics and fiber-reinforced plastics (Pipes, Vinson and Chou 1976, Shen and Springer 1976). Moisture absorption is a significant design consideration for polymer-matrix composites. In composites, water causes weight gain, hygrothermal expansion, and degradation of material properties (Pipes, Vinson, and Chou 1976; VanLandingham, Eduljee, and Gillespie 1995). As moisture diffuses into the polymer, it disrupts the local polymer structure and impacts the physical and mechanical properties of the material. Low levels of water in the polymer can also cause a reduction in the glass transition temperature (T_g) and the elastic modulus of the matrix (McCrum, Buckley, and Bucknall 1988). Semi-empirical models have related the reduction of T_g to the moisture content of the polymer. The reduction in T_g is material dependent but can be a serious consideration in the design of composite structure. Hence, the properties at the “wet T_g ” are often used to determine upper-use temperatures for a given polymer in specific applications and design allowables are frequently derived from mechanical characterization of materials that are fully saturated with moisture. Higher levels of moisture can cause cracks and delaminations to form in the composite and further degrade the performance of the composite structure.

Moisture absorption in complex composite parts is controlled by material properties, geometry, and processing. Material properties and processing affect both the moisture diffusion

rate and the amount of moisture that can be absorbed in the structure. The geometry influences the distribution through the surface area to volume ratio of different parts of the structure where a high surface area to volume will allow saturation to occur at a faster rate. Defects such as voids, microcracks, and pinholes affect the rate by providing a transportation mechanism into the part. These defects are also sites for water clusters to nucleate and hold water in the associated liquid state. These factors must be considered if a predictive capability is to be established for quantifying the moisture content in composite structures.

The theory presented in this paper consists of a method for analyzing moisture diffusion in multidomain structures particularly applicable to composites that have been covered with organic coatings. While diffusion in polymers and polymeric coatings can be defect driven, this paper takes a theoretical approach to characterize the effectiveness of ideal (no cracks or pinholes) coatings on composites. This allows the basic material properties of the coating and substrates, as well as the coating thickness, to be evaluated analytically to focus experimental work on the actual materials and processes. The theoretical work can then be used to extend predictions to complicated structures and realistic long-term environments.

2. Kinetics of Fickian Moisture Diffusion

In general, polymers can absorb moisture from their environment due to the nature of their structure. The degree and rate at which polymers can absorb moisture is very dependent on both the chemical and physical structure of the polymer of interest. Certain polymers are more hygroscopic than others and naturally absorb more moisture. Moisture transport in polymeric materials is related to the solubility of water (in the vapor phase) in the polymer and the moisture diffusion rate at a given temperature. The equilibrium of moisture absorption in polymers can be related to the relative humidity of the environment thermodynamically through Henry's law (Springer 1988; Marsh et al. 1988). The rate of diffusion is often described by Fick's second law and is strongly temperature dependent. However, in certain cases, non-Fickian diffusion is observed and more complicated analysis must be used to describe this behavior. For this work, attention is focused on Fickian diffusion mechanisms and state conditions where this assumption may be assumed.

It is convenient to begin moisture diffusion analysis, assuming Fickian diffusion for the moisture uptake for a single homogenous material. Also, if the sample thickness is much less

than the length and width, the diffusion can be considered to be one-dimensional. In this case, Fick's second law is expressed as

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial x^2} \quad (1)$$

For the case of an initially dry coating of thickness $2l$, and a simple constant concentration boundary conditions at $+l$ and $-l$, there is a well-known solution. This familiar series solution is obtained as

$$\frac{C(z, t)}{C_{\infty}} = 1 - \frac{4}{\pi} \sum_0^{\infty} \frac{(-1)^n}{(2n+1)} \exp\left[\frac{-D(2n+1)2\pi t}{4l^2}\right] \cos\left[\frac{(2n+1)\pi z}{2l}\right] \quad (2)$$

In this equation, C represents the concentration at a given z and t , C_{∞} is the equilibrium concentration, and D is the diffusion constant for water in the coating. The assumptions implicit in this solution are that (1) the coating is uniform in properties and composition and initially free from moisture, (2) the equilibrium moisture concentration at $\pm l$ is achieved instantaneously, and (3) the temperature of the system remains constant.

If equation (2) is integrated over z , the total uptake at any time can be obtained. Here, it is common to normalize the uptake with its ultimate value (Comyn 1992). The fractional uptake is expressed by

$$\frac{M(t)}{M_{\infty}} = 1 - \frac{8}{\pi^2} \sum_0^{\infty} \frac{1}{(2n+1)^2} \exp\left[\frac{-(2n+1)^2 \pi^2 Dt}{4l^2}\right] \quad (3)$$

Often, equation (3) is approximated by neglecting the higher order terms in the infinite series. Ignoring these terms, equation (3) simplifies substantially and the familiar square root relationship between mass uptake vs time is revealed as

$$\frac{M}{M_{\infty}} = \frac{2}{l} \left[\frac{Dt}{\pi} \right]^{1/2} \quad (4)$$

This equation has been widely used to determine diffusion coefficients of permeants in polymers. Several excellent studies on moisture uptake of polymers and composites have been published previously (Comyn 1992; Fieldson and Barbar 1993) that have revealed the usefulness of these techniques. In general, the linear behavior exists up to fractional uptakes of 0.6. Thus,

by monitoring the mass of samples exposed to moisture, the diffusion coefficient of water can be readily obtained. The effect of temperature on the rate of moisture diffusion can also be evaluated by conditioning samples and monitoring weight gain at different temperatures. Generally, an Arrhenius relationship will describe the temperature dependence of moisture diffusivity in polymer and polymer-composite samples.

The Fickian model is generally accepted as a reasonable approximation for diffusion of moisture in graphite epoxy exposed to humid air (Springer 1988). However, significant deviations from Fick's law can occur at high temperatures, when materials are immersed in liquids or when diffusion occurs through cracks or voids in the material (Springer 1988; Cai and Weitsman 1994). Immersion of the composite in liquids reduces the activation energy of diffusion, causing moisture to be absorbed at a higher rate than predicted by equation (1) (Woo and Piggott 1987). Cracks and voids facilitate moisture diffusion by increasing exposed surface area and acting as easy paths for moisture transport. Few studies have quantified the effects of voids and other high-permeability pathways on the amount and rate of moisture absorption. One study (Harper, Staab, and Chen 1987) has indicated that an increase of void-volume fraction from 1% to 5% in a graphite-epoxy laminate increased the moisture diffusion rate and saturation moisture content significantly for a range of humidity levels. Furthermore, as expected, the deviation from Fickian behavior was very large for the 5% void-volume fraction laminate.

3. Relative Humidity and Moisture Saturation

Polymer-matrix composites eventually reach maximum moisture content for a given surrounding humidity level. The relation between the maximum moisture content and the external humidity level is given by

$$M_m = \alpha(R.H.)^\beta, \quad (5)$$

where M_m is the maximum moisture content, α and β are material constants, and $R.H.$ is the surrounding relative humidity level (Tsai 1988). For most composite materials, the constant β has a value close to 1.0. Experimental values of the maximum moisture content for several graphite fiber-reinforced composites are shown in Table 1. The uptake values can vary widely, even for apparently similar material systems. For epoxy resins used as matrix materials, the equilibrium uptake is strongly dependent on the curing agent that is selected for that system. It

has been shown that polar curatives, such as diaminodiphenylsulfone (DDS), tend to cause higher saturation levels than similar curing agents with hydrophobic side groups (Springer 1988). The data on the IM7/K3B graphite fiber-reinforced polyimide was included to show the sensitivity of the matrix saturation level to the surrounding relative humidity. The saturation level of the IM7/K3B increases about 13% when the relative humidity is increased from 97% to 100%. VanLandingham, Eduljee, and Gallespie (1995) have also noted that the saturation level of the composites increases with repeated exposure to moisture. If the composites are saturated with moisture, then dried out and resaturated, they will have an increased maximum moisture content.

Table 1. Moisture Saturation Levels for Selected Graphite Fiber-Reinforced Composites

Material System	Exposure (% R.H.)	Saturation (% Moisture)	Reference
T300/1034	Immersed in Water	1.70	Tsai 1988
AS4/3501-5	Immersed in Water	1.90	Tsai 1988
T300/5208	Immersed in Water	1.50	Tsai 1988
T650-35/F584	98	0.62	Smith et al. 1993
AS4/8553-45	98	0.84	Smith et al. 1993
T650-35/1914-4	98	0.63	Smith et al. 1993
T650-35/1914-6	98	0.66	Smith et al. 1993
IM7/K3B	89	0.43	VanLandingham, Eduljee, and Gallespie (1995)
IM7/K3B	97	0.45	VanLandingham, Eduljee, and Gallespie (1995)
IM7/K3B	100	0.51	VanLandingham, Eduljee, and Gallespie (1995)

4. Methodology

This study investigates theoretical diffusion performance of coated substrates. It is intended to help focus experimental work and provide estimates of the “best case” moisture protection that can be afforded by coatings. As was mentioned before, many practical issues, such as quality of the coating, cracks, processing and application, and defects in the coating and substrate, are not addressed here. Figure 1 shows the schematic of the problem. The problem is divided into two regions: the substrate and the coating. The substrate is the material that is being

protected from moisture intrusion. The purpose of this paper is to investigate some of the geometric parameters and material properties that govern moisture diffusion.

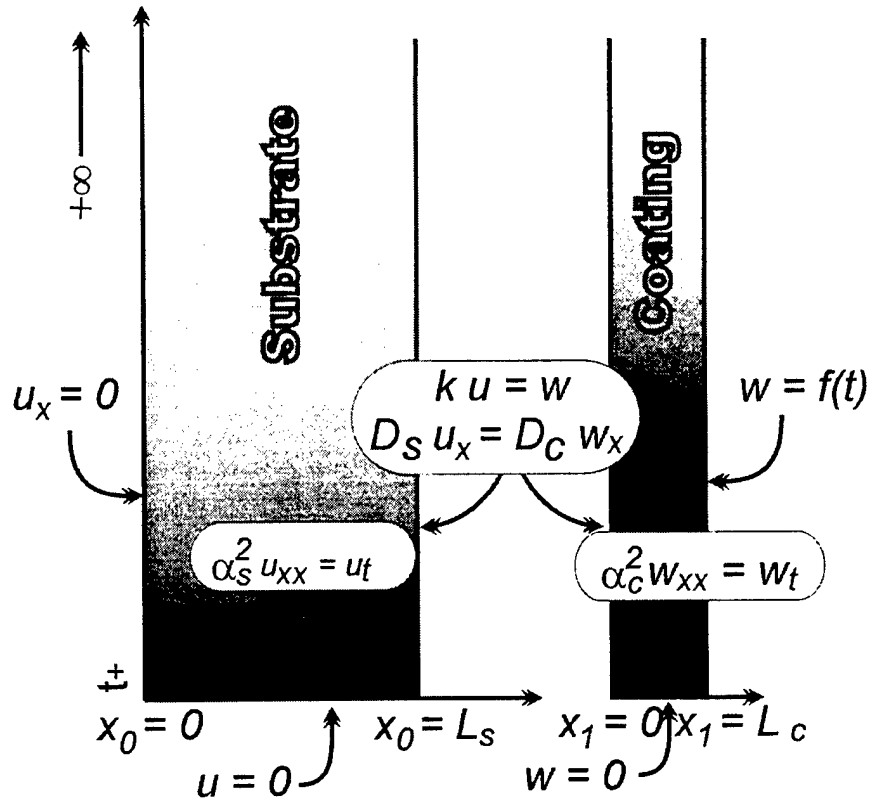


Figure 1. Schematic of Problem Description.

Figure 1 also defines the interface and boundary conditions used for solution. The solution is divided into two domains each with its own local coordinate system. The first domain extends from $x_0 = 0$ to $x_0 = L_s$ and the second domain extends from $x_1 = 0$ at the interface to $x_1 = L_c$ at the outside boundary. It is assumed that both domains start without any moisture. The problem is set up to investigate symmetric one-dimensional loading of the structure. The use of symmetry implies that the midplane boundary condition is represented by zero flux. At the right side of the domain, equation (5) is used where the “ α ” is assumed to be the saturation level of the coating material and “ β ” is assumed to be 1 (Tsai 1988). At the interface a similar condition exists (Carslaw and Jaeger, 1959) two conditions must be met. First, the driving potential of the two materials must be the same:

$$\frac{u}{Sat_s} = \frac{w}{Sat_c}, \text{ or } k u = w, \text{ where } k = \frac{Sat_c}{Sat_s}. \quad (6)$$

The second condition is that the flux at the interface must be continuous:

$$D_s \frac{\partial u}{\partial x_0} = D_c \frac{\partial w}{\partial x_1} \quad (7)$$

There are two important ratios that are present in this problem. The first, the ratio of the saturation levels, is described in equation (6), and the second is the ratio of the diffusion rates of the coating material and the substrate:

$$a = \frac{D_c}{D_s} \quad (8)$$

The range of values used for a and k in this study were obtained from reviewing the moisture literature and data from coating manufacturer. The range of diffusion rates for coating materials was from an order of magnitude below the diffusion rate for the substrate (T650/1914, $6.6 \cdot 10^{-6}$ in²/hr) (Bogetti et al. 1997) to an order of magnitude above. The range for the saturation level of the coating materials was from 0.25 times to 8 times the saturation level for the substrate (T650/1914's saturation level is 0.63 %). This does not imply that there is a coating that represents every combination of a and k used in the study. The aim of the study is to find the key material attributes, not to look at particular systems. The next step in this work is to characterize available coating systems for appropriate use in applications.

The system is solved using both exact and numerical solutions, although the results contained in this paper are from the numerical work. The solution was obtained in the following manner.

Finite difference methods (FDMs) change the differential operators to algebraic operators. The algebraic operators are applied over a discretized approximation of the original domain.

The operators used are implicit Crank-Nickolson algebraic operators. Defining the following for convenience (Greenberg, 1978):

$$r_i = \frac{\alpha_i^2 \Delta t}{2\Delta x^2}, \quad (9)$$

and

$$\Delta x_i = \frac{L_i}{ns}, \quad (10)$$

where $i = c$ or s for the coating or substrate layer, respectively. The governing equation is

$$r_k u_{i+1,j+1} - (2r_k - 1)u_{i,j+1} + r_k u_{i-1,j+1} = -r_k u_{i+1,j} + (2r_k - 1)u_{i,j} - r_k u_{i-1,j}. \quad (11)$$

The subscript $k = s$ for $i \in [1, ns - 2]$ and $k = c$ for $i \in [ns + 1, DOF - 2]$, where ns is the number of nodes in the substrate and DOF is the number of degrees of freedom in the system. Note, for convenience, the primary dependant variable in the coating layer, w , has been renamed as u with subscripts from $ns + 1$ to $DOF - 1$. Four additional equations are needed to complete the system. These equations are the boundary conditions and the interface conditions. The boundary conditions are defined in the next two equations:

$$u_{1,j+1} - u_{1,j} - u_{0,j+1} + u_{0,j} = 0 \quad (12)$$

and

$$u_{DOF-1} = \alpha(R.H.)^\beta. \quad (13)$$

The interface conditions are given in the next two equations:

$$k u_{ns,j+1} - u_{ns+1,j+1} = k u_{ns,j} - u_{ns+1,j} \quad (14)$$

and

$$r_s (u_{ns,j+1} - u_{ns-1,j+1}) + r_c (u_{ns+1,j+1} - u_{ns+2,j+1}) = -r_s (u_{ns,j} - u_{ns-1,j}) - r_c (u_{ns+1,j} - u_{ns+2,j}). \quad (15)$$

This leaves a fully defined system of the following form,

$$\mathbf{A} u^{new} = \mathbf{B} u^{old} + c, \quad (16)$$

which is easily solved and iterated for the appropriate time:

$$u^{new} = \mathbf{A}^{-1} \mathbf{B} u^{old} + \mathbf{A}^{-1} C. \quad (17)$$

5. Typical Results

Figure 2 and Figure 3 show typical results for coated substrates.

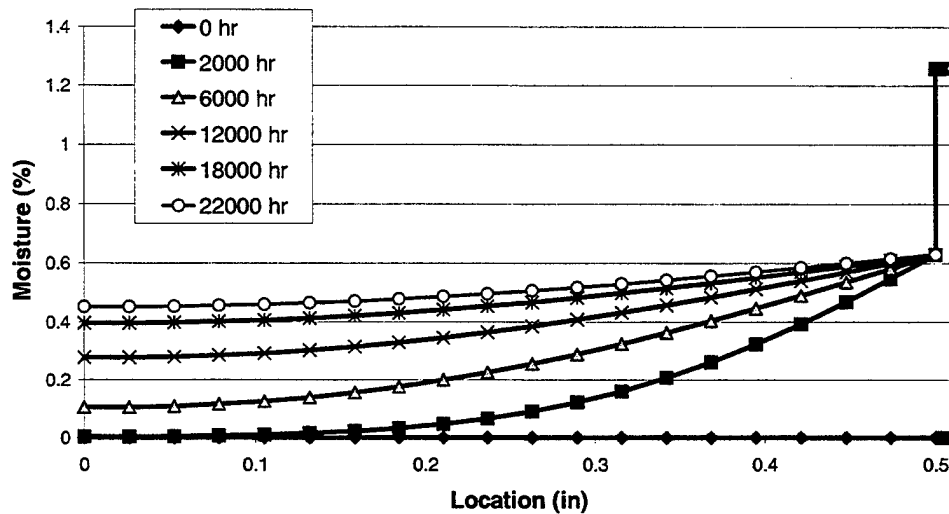


Figure 2. Diffusion Results for a Coated Substrate, $k = 2$, $a = 1$, $L_s = 0.5$ in, and $L_c = 0.01$ in.

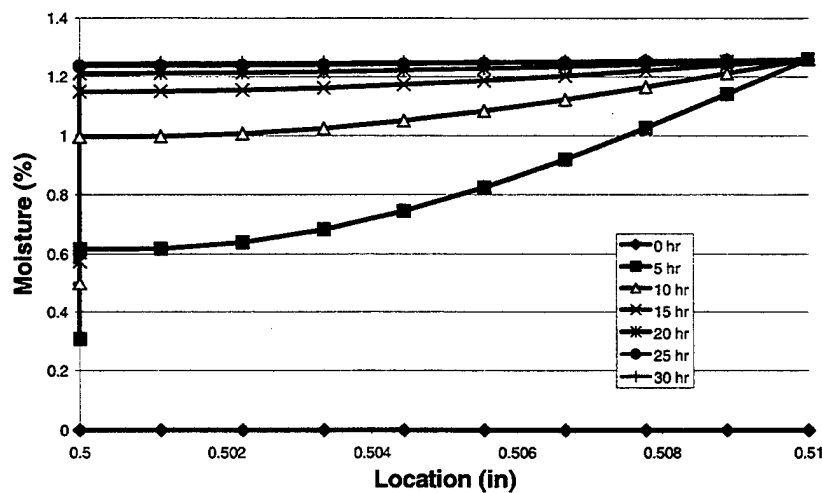


Figure 3. Short Time Behavior of the Coating Layer.

As seen in Figure 2, there are several important features. First, at the interface between the coating and substrate, there is a discontinuity. The discontinuity is a function of the difference in the saturation levels of the coating and substrate. Although, it is difficult to see in

the figure, the flux at the interface is also matched. The development of the solution in the substrate is very similar to the solution in a noncoated layer.

Figure 3 shows that at 30 hr, the coating layer appears to have assumed a quasi-static solution and is almost fully saturated. This implies that any additional moisture entering the coating layer is being transferred to the substrate. It also implies that the material properties of the coating layer should dominate the diffusion process. It is seen in later figures that this is not always the case.

6. Effects of Coating

The next series of figures investigates the effect of the coating thickness and relative materials properties for a thin ($L_s = 0.1$ in) and thick ($L_s = 0.5$ in) substrates. In order to look at moisture penetration into the substrates, the normalized moisture level at the midplane of the substrate was used as a measure. The moisture level was normalized by dividing by the saturation level of substrate and is referred to as the diffusion potential. As a comparison, the equivalent substrate without a coating is shown to gauge relative diffusion performance. The substrate with $a = 1$ and $k = 1$ implies that the coating layer has the same diffusion rate and saturation level as the substrate. For each of the analysis, material properties were chosen for the substrate, then the coating material properties were varied relative to the substrates.

Three types of figures are needed in order to assess the diffusion performance of a coating, time histories, substrate-coating interface plots, and diffusion potential vs the material property ratios' a and k . The time histories show deviation during the diffusion process and ensure that relative comparisons are accomplished at appropriate times. The substrate-coating interface plots provide insight into how the coating works, and the diffusion potential vs material property ratios help isolate effective ranges for the material attributes. On each of the graphs, the diffusion performance for several combinations of the ratios a and k are given to gauge the relative diffusion performance.

Figure 4 shows the general effects of coatings. Since each line in the figure represents a different type of coating, the separations between the lines imply the particular coatings effect.

Each of the lines should be gauged against the case where a and k are equal to 1, which represents the effect of just thickening the substrate without adding a coating.

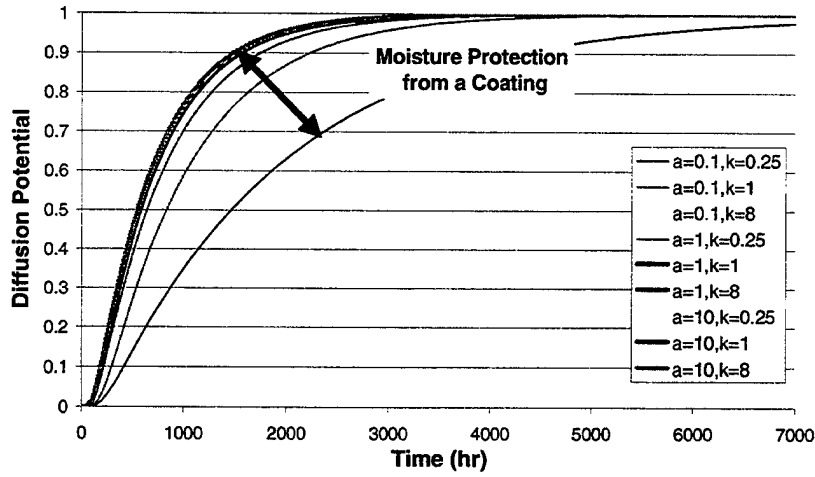


Figure 4. Effect of Coatings shown through the Diffusion Potential at the Midplane ($L_s = 0.1$ in, $L_c = 0.01$ in).

To assess the effectiveness of a coating, the assessment should be made at a time corresponding to the separation near the largest deviation seen in the figure. Figure 5 and Figure 6 show the diffusion potential performance for three different coating thickness on a relatively thin substrate ($L_s = 0.1$ in).

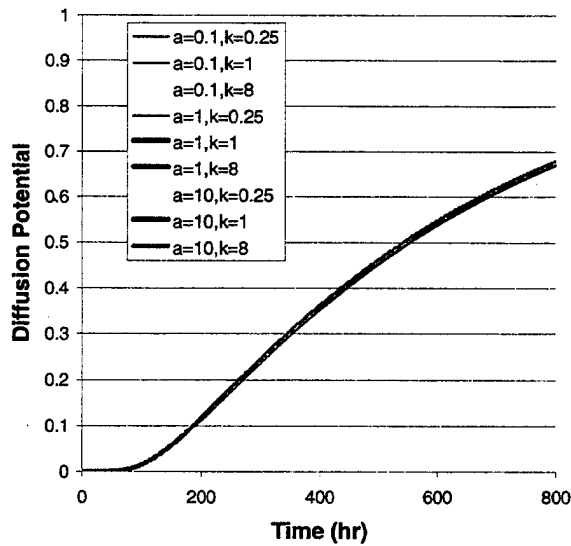


Figure 5. Diffusion Potential at the Midplane for $L_s = 0.1$ in and $L_c = 0.001$ in.

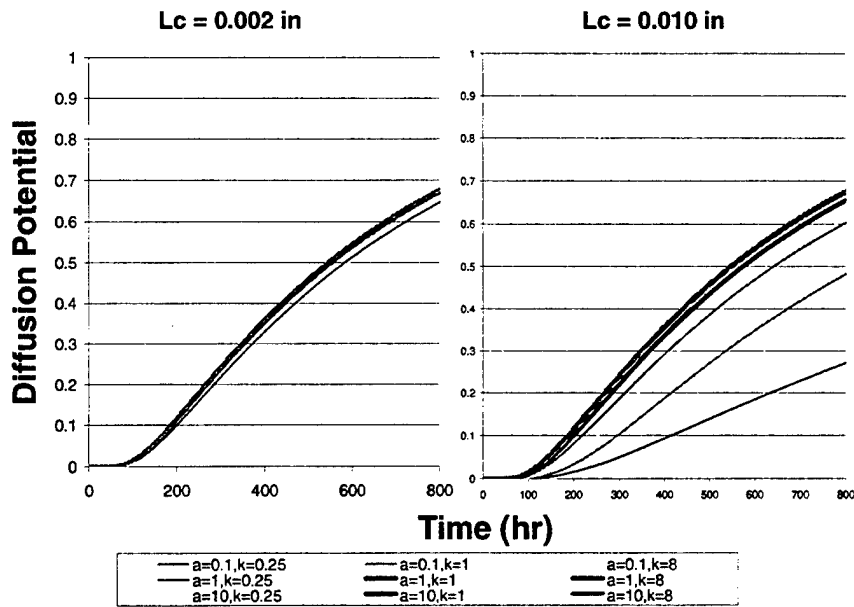


Figure 6. Diffusion Potential at the Midplane for $L_s = 0.1$ in and $L_c = 0.002$ in and $L_c = 0.01$ in, respectively.

Figure 5 shows relative short time (800 hr ~ 1 month) performance of the substrate. The figure shows that, at 800 hr, the diffusion potential is around 0.66, where a value of 1 represents the saturation value. It is very interesting to see that the wide range of material properties does not have any appreciable effect on the diffusion. Figure 6 shows the same results for a coating thickness that is twice as thick ($L_c = 0.002$ in) and ten times as thick ($L_c = 0.01$ in). In this figure, some very small differences can be seen around 800 hr. The maximum value remains near 0.66, and it is important to note that the coated substrates do not appreciably perform differently than the uncoated substrates. In Figure 6, with $L_c = 0.01$ in, a dramatic difference can be seen in the diffusion performance of the different coatings. Even with this relatively thick coating, five of the nine combinations of a and k do not cause the coated substrate to behave differently from the uncoated substrate. The figure shows, that if the material properties of the coating are chosen properly and the coating is applied thick enough, potentially the diffusion can be cut in half. It appears from these figures that control of both a and k are necessary to affect diffusion. This is not surprising as the total permeability of the coating can be expected to be a product of the diffusivity and solution of moisture in the coating. It is also interesting to note that some of the time-history lines cross. This implies that the diffusion rate and saturation level have different effects for short and long time periods.

Figure 7 shows the effects of the same coatings on a thick substrate. The $L_c = 0.001$ in case was omitted since very little diffusion difference is seen in the thicker coatings. As is seen in the figures, for thick substrates, the coatings have very little effect on the moisture diffusion. Only in the case where the coating is 0.01 in thick is there any difference in performance and the performance difference is negligible.

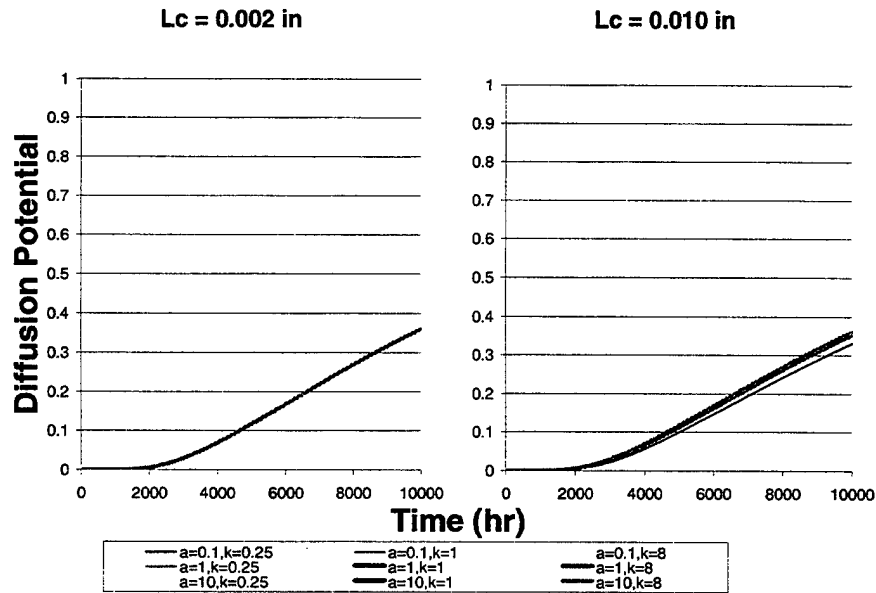


Figure 7. Diffusion Potential at the Midplane for $L_s = 0.5$ in and $L_c = 0.002$ in and $L_c = 0.01$ in, Respectively.

Figure 8 shows the time history of moisture at the interface between the coating and the substrate. The figure shows that, for many of the combinations of coatings, the interface goes to the saturation level very quickly. Again, many of the lines cross each other showing that the saturation level and the diffusion rate operate on different time scales. Finally, the figure also shows that, in order to affect the diffusion rate in the substrate, the interface should not quickly drive to the saturation level of the coating materials. When the saturation level is reached, the maximum amount of moisture is diffusing from the coating into the substrate. If a lower diffusion potential can be held, allowing a smaller amount of moisture to be transferred. This appears to be an effective coating attribute. Later, Figure 9 and Figure 10 directly address combinations of a and k , which offer the best protection.

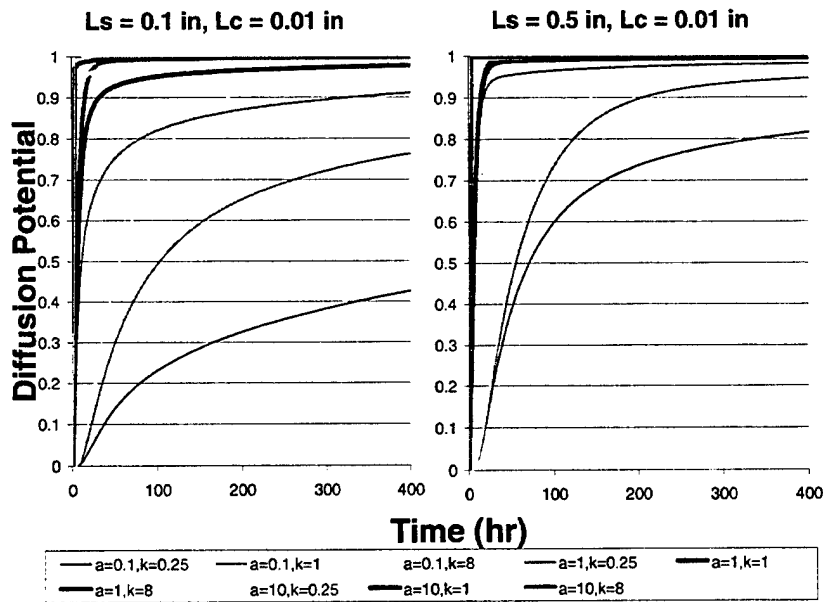


Figure 8. Diffusion Potential at the Interface Between the Coating and Substrate.

Looking at the interface performance for $L_c = 0.01$ in, again there appears to be very little difference in the various coatings. In each case, the diffusion potential very quickly goes to the saturation level of the materials. When compared to the thin substrate results, one of the main differences is that the a and k combinations offering some protection did not saturate quickly and that behavior is absent here. Again, some of the lines cross each other, which implies that the time scales are different for the diffusion rate and saturation levels.

It should also be noted that in Figure 8, some of the lines imply that, with the wrong combination of coating material properties, moisture can be absorbed faster in the substrate than the substrate without a coating.

Figure 9 and Figure 10 investigate the relationships between a , k , and the midplane diffusion potential for various thicknesses of coatings and substrates. These figures, with the exception of Figure 9, show that the coating material properties have very little effect on the amount of moisture uptake in the substrate. For the different material properties, the curves remain flat with very little difference for the midplane diffusion levels. Only when the diffusion rate is very low is the diffusion behavior changing. Figure 9 shows more divergent behavior.

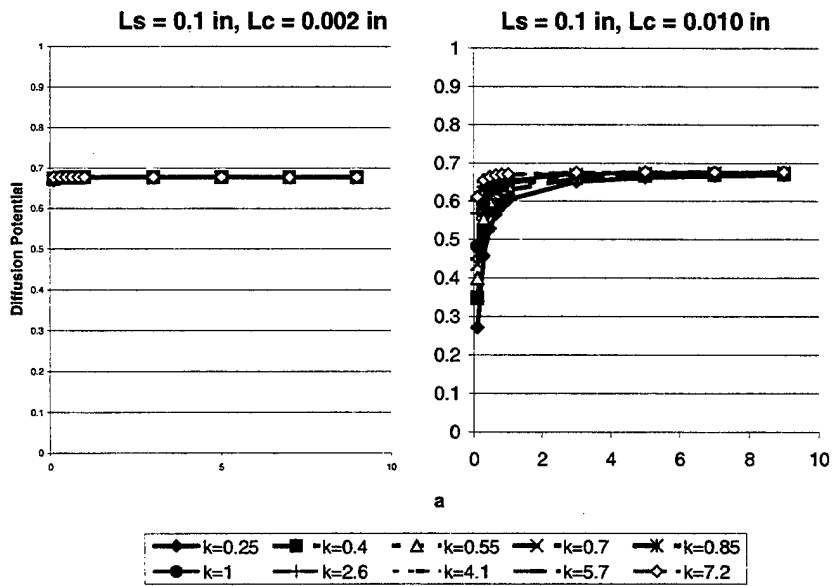


Figure 9. Effect of a and k at $t = 800$ hr.

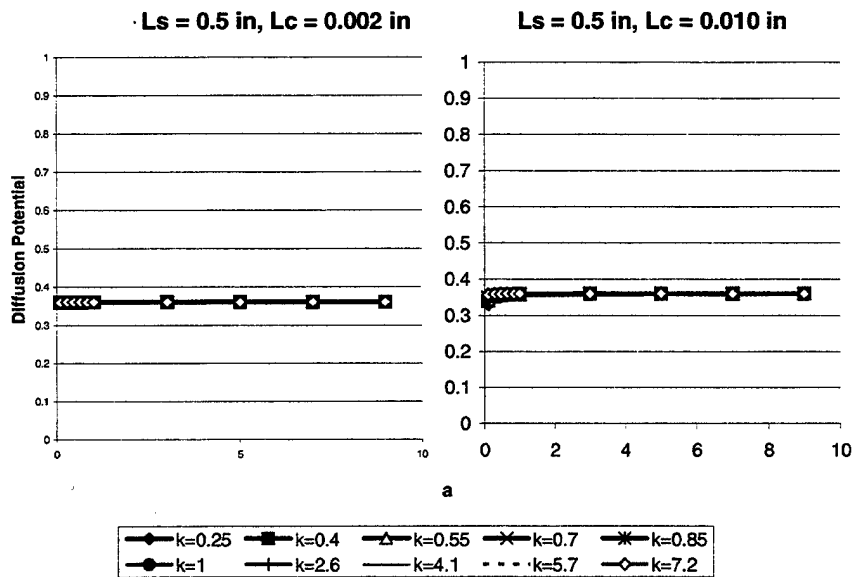


Figure 10. Effect of a and k at $t = 10000$ hr.

Figure 11 shows a portion of Figure 9 for small values of a .

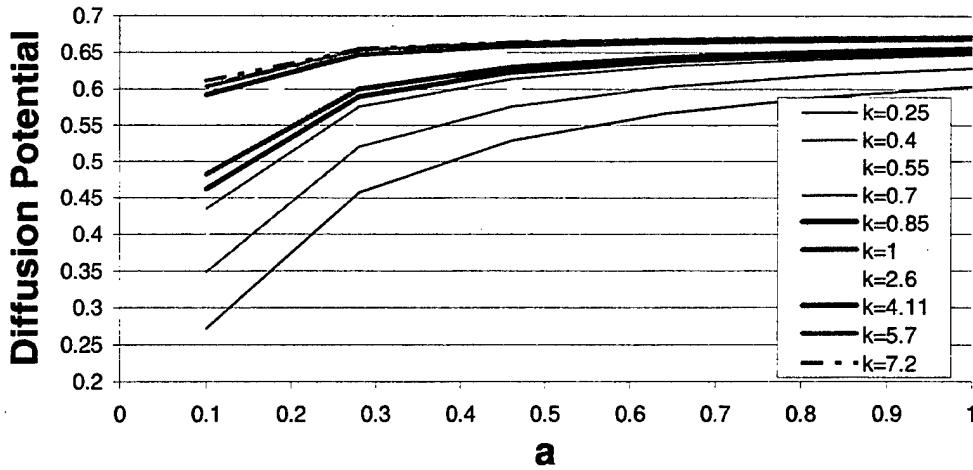


Figure 11. Effect of a and k for $L_s = 0.1$ in and $L_c = 0.01$ in for $t = 800$ hr for Small a .

Figure 11 shows significant reduction in moisture uptake for small values of a . This translates into a diffusion coefficient for the coating around an order of magnitude smaller than the diffusion coefficient for the substrate. It is also clearly shown in this figure that, if the saturation value for the coefficient for the coating is relatively much higher than the saturation value for the substrate, the coating will facilitate moisture uptake in the substrate. This is seen when comparing the curve for $a = 1$ and $k = 1$ to the other curves. It is also seen that this effect can be counteracted by lowering the diffusion coefficient.

Figure 12 shows interface results for a thick coatings ($L_c = 0.05$ in) on a very thin substrate ($L_s = 0.05$ in) and a thick substrate ($L_s = 0.5$ in). In both cases with the right combinations of a and k , the coating are effective. On the thin substrate, similar values to earlier cases for a and k are again effective. For the thick substrate, now that the coating thickness is approach 10%, with the proper values for a and k the coating is protecting the substrate.

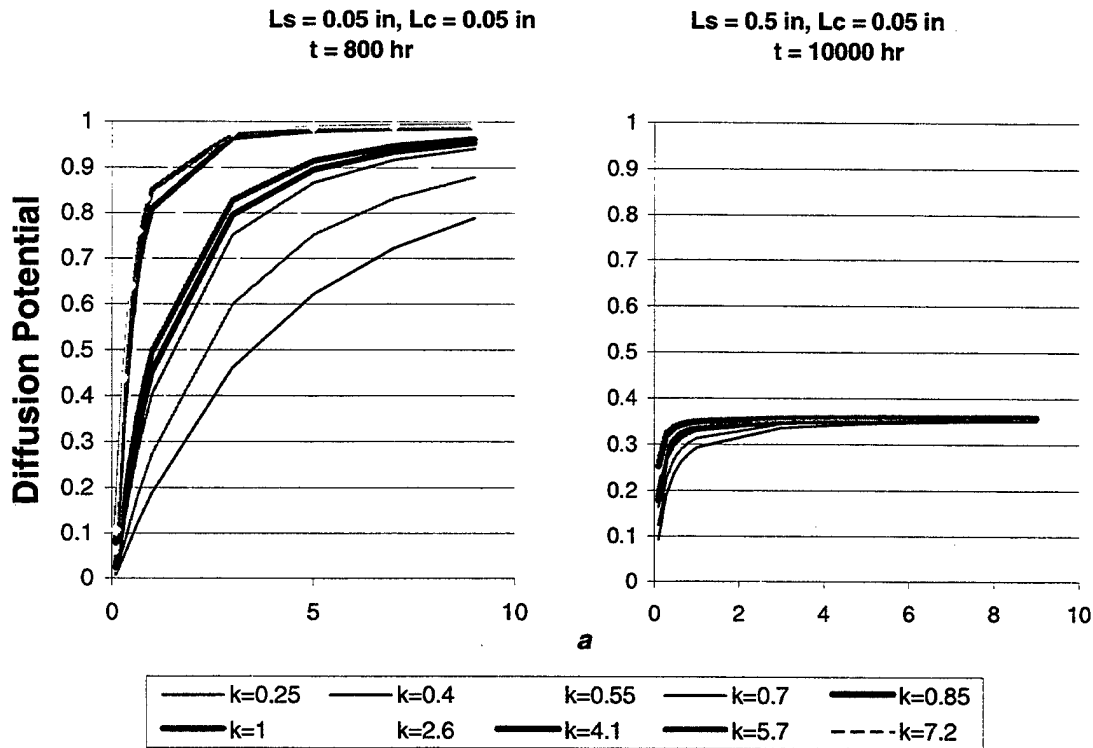


Figure 12. Effect of a and k for a Very Thick Coatings.

7. Conclusions

Polymeric coatings can be used to reduce the moisture absorption in polymer and polymer-composite structures. The analytical approach taken in this paper demonstrates that the thickness, diffusion constant, and saturation level of the coating relative to those of the substrate are essential to the coatings effectiveness. The diffusion rate and the saturation level of the coating must both be very small compared to the substrate for the coating to affect the long-term diffusion behavior. Even when the material constants are optimized, the coating must be relatively thick to affect a substantial reduction of moisture absorption. For the substrates studied in this paper, the thickness of the coating needed to be 10% of the thickness of the substrate to achieve effective protection. Even with coatings 10% of the substrate thickness, many values of a and k are not effective in protecting the structure from moisture ingress. This is partially due to the speed that the coating layer achieves a quasi-steady-state distribution. The coatings that did not saturate quickly were more effective at protecting moisture diffusion. This is not to say that an impermeable coating is the best solution, since it also does not allow

moisture to exit the substrate and could consequently cumulate moisture. This will be investigated in future studies that address temperature effects and fluctuating environmental conditions. It is also interesting to note that the saturation level of the coating and the diffusion rate of the coating can act on different time scales. The results from this study will be used to target the important attributes to allow the selection of appropriate coatings. Experimental evaluation of the coating and the manufacturing process are necessary to ensure that the chosen system can be implemented effectively in a structure.

8. Future Work

Analytical and numerical solutions for moisture performance of coated substrates have been developed. These solutions are being used to develop methods for approximating coating for large complicated models. In addition, these methods are being compared to different coating system to assess the theoretical performance against coatings that are not “perfect” and contain voids, cracks, other defects, or processing differences. The theories developed will be modified to allow realistic prediction of complicated structures protected by coatings with defects.

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