

# Modeling of Diffusion Through Optical Fiber Coatings

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## ABSTRACT

In earlier work, diffusion of moisture through polymer coatings was modeled by using an analytical solution to the diffusion equation and so was only applicable to the simplest cases, *e.g.* cylindrically symmetric Fickian diffusion. In this work the limitation of the analytical approach is avoided by the use of finite element analysis. However, finite element programs do not usually implement matter diffusion, and therefore it has been modeled by analogy with thermal conduction.

**Keywords:** diffusion, optical fiber reliability, moisture, coatings, and finite element analysis

## 1. INTRODUCTION

### 1.1 Diffusion

Silica glass optical fibers are normally coated with dual layers of UV curable acrylate. The inner, primary coating, has a lower modulus than the outer, secondary coating. These coatings serve to reduce microbending loss, protect the glass surface from handling damage, and can affect the rate of chemical attack of the surface.<sup>1</sup> In order to properly perform mechanical testing of coated optical fiber, equilibration with the test environment, usually humid air, must be achieved. Previous work has shown that the diffusion of moisture through coatings can be monitored by measuring the strength of the fiber.<sup>2-4</sup> In that work it was shown that the diffusion coefficient can be calculated by using the following analytical equation:

$$1 - \frac{C(r, t)}{C_1} = \pi \sum_{n=1}^{\infty} \left\{ \exp(-D\alpha_n^2 t) J_1(a\alpha_n) J_0(b\alpha_n) \frac{Y_0(r\alpha_n) J_1(a\alpha_n) - J_0(r\alpha_n) Y_1(a\alpha_n)}{J_0(b\alpha_n)^2 - J_1(a\alpha_n)^2} \right\}, \quad (1)$$

where  $C(r, t)$  is the concentration at a distance  $r$  from the center of the fiber at a time  $t$ ,  $D$  is the diffusion coefficient,  $a$  is the glass fiber radius,  $b$  is the coating outer radius,  $J_i$  and  $Y_i$  are Bessel functions of order  $i$ , which may be approximated by polynomials as given by Abramowitz and Stegun,<sup>5</sup> and  $\alpha_n$  is the  $n$ th positive root of:

$$J_1(a\alpha) Y_0(b\alpha) - J_0(b\alpha) Y_1(a\alpha) = 0. \quad (2)$$

The results of this work<sup>2-4</sup> and some work performed by Bouten,<sup>6</sup> showed that the diffusion of moisture through the polymer coatings on optical fibers occurred in times on the order of minutes. Bouten's results showed that the diffusion of moisture out of the coating into low ambient humidity ( $\ll 20\%$ ) occurred more than 10 times slower than diffusion at nominal humidities. This could be explained by the fact that a monolayer of moisture forms on a silica surface at 18% RH.<sup>7</sup> Neither of these experiments takes into consideration the polymer reacting with the diffusing species, *i.e.* non-Fickian diffusion. Finite element analysis will be used here to determine if diffusion along the ends of the fiber is significant and to model dual coated systems and a case where the solubility of water into the polymer is varied.

### 1.2 Finite element analysis

Finite element analysis (FEA) consists of modeling a part or specimen by breaking it down into small discrete pieces, which are called elements. The quantity of interest (concentration in this case) is assumed to vary simply across each element (*e.g.* linearly) to form a piece-wise approximation to the behavior across the whole specimen. ANSYS 5.3<sup>†</sup> was used to perform

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the analysis in this paper. In order to model moisture diffusion into polymer coatings with the ANSYS package, an adaptation of the heat flow equations had to be used.

Fick's first and second laws for diffusion in one dimension are as follows:

$$F = -D \frac{\partial C}{\partial x} \quad \text{and} \quad \frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} \quad (3)$$

where  $F$  is the heat flux,  $C$  is the concentration of the diffusing species,  $x$  is the distance,  $D$  is the diffusion coefficient, and  $t$  is time.<sup>8</sup> The two corresponding heat transfer equations are

$$F = -k \frac{\partial \theta}{\partial x} \quad \text{and} \quad \frac{\partial \theta}{\partial t} = \left( \frac{k}{c\rho} \right) \frac{\partial^2 \theta}{\partial x^2}, \quad (4)$$

where  $\theta$  is temperature,  $k$  is thermal conductivity,  $\rho$  is density, and  $c$  is the specific heat per unit mass. By comparing these equations diffusion can be modeled by equating temperature,  $\theta$  to concentration,  $C$ , and the diffusion coefficient,  $D$  to thermal diffusivity,  $k/c\rho$ . If it is assumed that the moisture forms an ideal solution in the polymer then relative humidity can be equated to the concentration in the polymer *i.e.* temperature in heat transfer, then without loss of generality we can take  $c\rho$  equal to 1, and  $D$  equivalent to  $k$ .

If materials with different solubilities for water are to be modeled then the solubility needs to be incorporated into the model. This can be done by recognizing that the diffusion is really controlled by activity gradients, rather than concentration; it is activity which is matched across boundaries. In the diffusion experiments relative humidity has been equated to concentration. In reality the relative humidity is only equal to the molar concentration when the activity coefficient is equal to one. Activity,  $a$ , is defined as follows:

$$a = \frac{p_{\text{H}_2\text{O}}}{p_{\text{sat}}} = RH = \gamma^* x \quad (5)$$

where  $\gamma^*$  is the activity coefficient and in this case is assumed to be Henrian *i.e.* a constant, and  $x$  is the mole fraction of water in the polymer,  $p_{\text{H}_2\text{O}}$  is the partial pressure of water in the air and  $p_{\text{sat}}$  is the saturated vapor pressure. The activity can be related to the concentration of moisture,  $C$ , by

$$a = \gamma_{\text{eff}} C, \quad (6)$$

$$\text{where } \gamma_{\text{eff}} = \gamma^* f, \quad (7)$$

where  $\gamma_{\text{eff}}$  is the effective activity coefficient and  $f$  is a conversion factor between concentration in the polymer and vapor pressure in the surrounding air. The activity is equal to the relative humidity, so the relationship between the relative humidity and concentration is as follows:

$$RH = \gamma_{\text{eff}} C \quad (8)$$

Since the diffusion is driven by activity gradients, Fick's first law can be written:

$$\text{Error! Objects cannot be created from editing field codes.} \quad (9)$$

By comparison with (3) and (6) we find:

$$D' = \frac{D}{\gamma_{\text{eff}}}. \quad (10)$$

The solubility of water in the polymer,  $C_\infty$ , is the concentration of moisture in equilibrium with 100% humidity in the surrounding air, *i.e.* from (8)

$$C_{\infty} = \frac{1}{\gamma_{eff}} . \quad (11)$$

Therefore, different solubilities can be accounted for by varying  $\gamma_{eff}$ , *i.e.* a polymer with high solubility will have a low  $\gamma_{eff}$ .

In order to model different solubilities in ANSYS, equivalence is drawn between equations 4 and 9. Table I shows the equivalence between the diffusion and heat transfer parameters.

**Table I. Equivalent between diffusion and heat transfer parameters**

Diffusion	Heat transfer
$RH$	$T$
$D/\gamma_{eff}$	$k$
$D$	$k/c\rho$
	$\rho=1$ (for convenience)
	$c=1/\rho\gamma_{eff}$

## 2. EXPERIMENTAL

The shape of coated optical fibers is cylindrical, but by utilizing the axisymmetry option, a two-dimensional model can be used in ANSYS to solve this problem. The element chosen for this model was the thermal solid PLANE 55, which has four nodes and can be used in axial symmetry. The two-dimensional shape used here was a rectangular slab, which can be seen in Figure 1. The dimensions of this slab were 125  $\mu\text{m}$  ( $x$  direction) and 500  $\mu\text{m}$  ( $y$  direction). In reality the fiber sample dimensions are radius,  $r = 125 \mu\text{m}$  (coated) and length,  $l \gg 500 \mu\text{m}$ . A small length was used in this model to reduce computational time. For all cases considered here, the outer diameter of the coating is 250  $\mu\text{m}$ . If there is no evidence of significant end effects at this length, then there will be no end effects at longer lengths. For the single polymer coated case the section was divided in half, where the silica glass is material 1 and the polymer coating is material 2 (see Figure 1a). In the dual coated case, which is shown in Figure 1b, the polymer coating was divided into two parts of equal thickness (31.25  $\mu\text{m}$ ) where material 2 is the secondary coating, and material 3 is the primary. The material properties of the three regions can be specified separately from each other. Previous research has found the diffusion coefficient for moisture into glass at 90°C to be  $10^{-19} \text{m}^2/\text{s}$ .<sup>9</sup> In the current work, the interest lies in how fast the moisture gets to the glass surface, not in the diffusion of the moisture into the glass which is negligible at temperatures and on time scales of interest here. Therefore, as long as the assigned diffusion coefficient for water into glass is several orders of magnitude lower than that in polymers, the time it takes the moisture to get to the glass surface can be studied. To avoid a ratio error in ANSYS, the diffusion coefficient for the glass material was set to  $10^{-17} \text{m}^2/\text{s}$ , which is  $\sim 10^5$  times lower than the values used for the polymer.

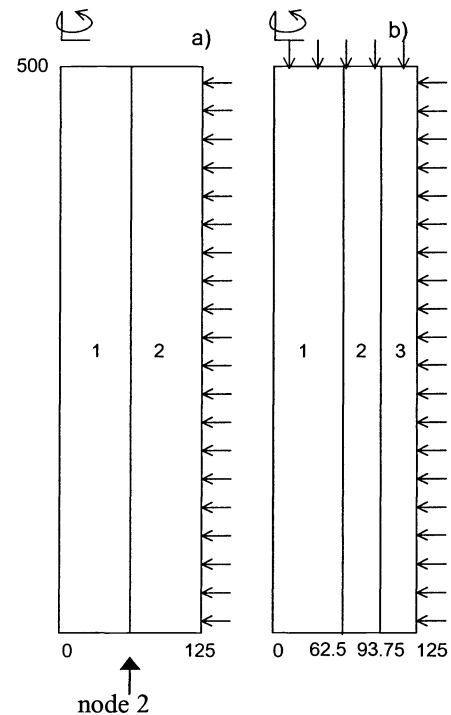


Figure 1. Model for finite element analysis a) single coated with surface loads b) dual coated with end and surface loads. Dimensions are in microns.

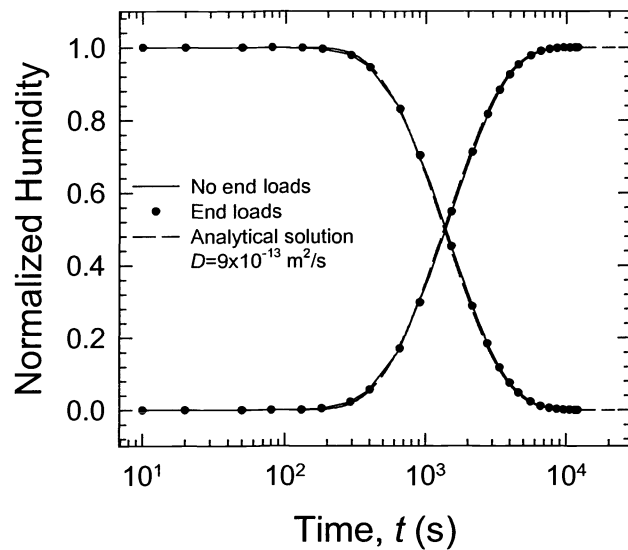


Figure 2. ANSYS diffusion data for a single coated fiber with and without end loads.

The total time specified was 12,000 s with a minimum time step of 10 s and a maximum time step of 100 s.

### 2.3 Dual coating

For the dual coated model three different cases were studied which are summarized in Table II. The model for the dual coated fiber is shown Figure 1b, except that the length in the  $y$ -direction was set to be  $62.5 \mu\text{m}$  and there were no end loads applied. This smaller area allowed for finer meshing to be used while also reducing computation time. Since the diffusion rates and solubilities are varied for the primary coating in this set of models, the total time had to be increased from 12,000 s to 50,000 s to ensure that the diffusion process was completed. The time steps used in this set of experiments were a minimum of 10 s and a maximum of 1,000 s.

Table II: Three different dual coated cases studied.

Case 1	Case 2	Case 3
Same solubility but higher primary diffusivity	Same diffusivity but higher primary solubility	Higher primary solubility and diffusivity
$D_{\text{primary}} = 10 D_{\text{secondary}}$	$D_{\text{primary}} = D_{\text{secondary}}$	$D_{\text{primary}} = 10 D_{\text{secondary}}$
$\gamma_{\text{eff,primary}} = \gamma_{\text{eff,secondary}}$	$\gamma_{\text{eff,primary}} = \gamma_{\text{eff,secondary}} / 10$	$\gamma_{\text{eff,primary}} = \gamma_{\text{eff,secondary}} / 10$

## 3. RESULTS AND DISCUSSION

Figure 2 shows the results for the single coating normalized humidity *versus* time calculated at node 2 (see Figure 1a) for cases both with and without end loads. Since the with and without end load curves fall right on top of each other, the effect of the end load is negligible at this ratio of  $l/d$ , where  $l$  is the length along the  $y$ -axis and  $d$  is the thickness of the coating. This implies that at higher  $l/d$  ratios, diffusion into the ends of the fiber has a negligible effect on the water activity at the glass surface. This figure also shows a comparison between the finite element analysis (solid line or dots) and the analytical solution, equation 1 (dashed line). The diffusion coefficient used in the finite element analysis was  $1 \times 10^{-12} \text{ m}^2/\text{s}$ ; a diffusion

### 2.2 Single coating with and without end loads

Two different load schemes or boundary conditions were investigated, and for all cases top to bottom symmetry was assumed. The first load scheme, which can be seen in Figure 1a, was to only apply a load through the coating not along the ends *i.e.* the temperature ( $\equiv$  humidity) on the outside surface is fixed. The second load scheme, shown in Figure 1b, includes the end load which was only applied to the top surface in this model because the rectangular section is only one half the total length of fiber modeled. The humidity applied at these nodes depends on which diffusion was being modeled *i.e.* diffusion in or out. For example, for diffusion in, the initial uniform humidity of the body was defined to be 20% RH and the external loads applied to the body were 95% RH. The reverse was applied for diffusion out. In order to obtain the relative humidity as a function of time at the glass/polymer interface a time step and total time had to be defined.

coefficient of  $9 \times 10^{-13} \text{ m}^2/\text{s}$  in equation 1 was found to give good agreement with the modeling results. Employing finer meshing and smaller time steps in the FE model could eliminate this slight difference.

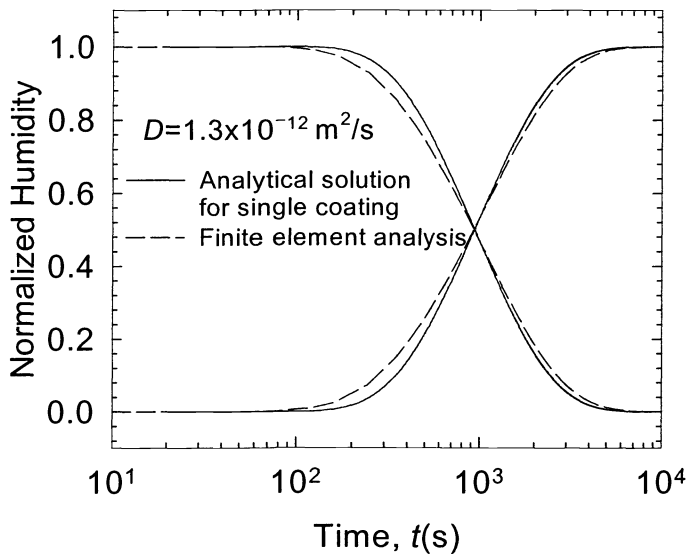


Figure 3. Case 1: dual coated fiber with a primary that has a diffusion coefficient that is tens times higher than the secondary.

constant and varies with humidity. However, this interpretation would not be correct since the broadening is readily explained by having two Fickian materials with different diffusivities.

Figure 4 shows the results from case 2, which is a dual coated system where the primary has the same diffusion coefficient, but in this case the solubility of water in the primary is set to be ten times the solubility in the secondary. The best fit of equation 1 to these curves results in an effective diffusion coefficient,  $D_{eff} = 2.2 \times 10^{-13} \text{ m}^2/\text{s}$ . So when the primary has a higher solubility the overall diffusion process is slowed. This makes sense because the primary needs to absorb more water. Changing the solubility results in apparent deviations from linear Fickian diffusion behavior, since linear Fickian diffusion assumes that the diffusion coefficient is not dependent on the concentration of the diffusing species. Again, broadening of the diffusion curve need not indicate that non-Fickian diffusion is occurring.

The results for case 3, where the primary coating has a higher diffusion coefficient and a higher solubility, are shown in Figure 5. The best fit diffusion coefficient for this case was  $D_{eff} = 2.3 \times 10^{-13} \text{ m}^2/\text{s}$ . This cumulative effect of higher solubility and higher diffusion has resulted in curves that are slightly broader than Figure 4, but increasing the diffusion coefficient of the primary did not greatly impact the overall speed of the diffusion process. Therefore changes in solubility broaden the diffusion profile more than changes in diffusion coefficient.

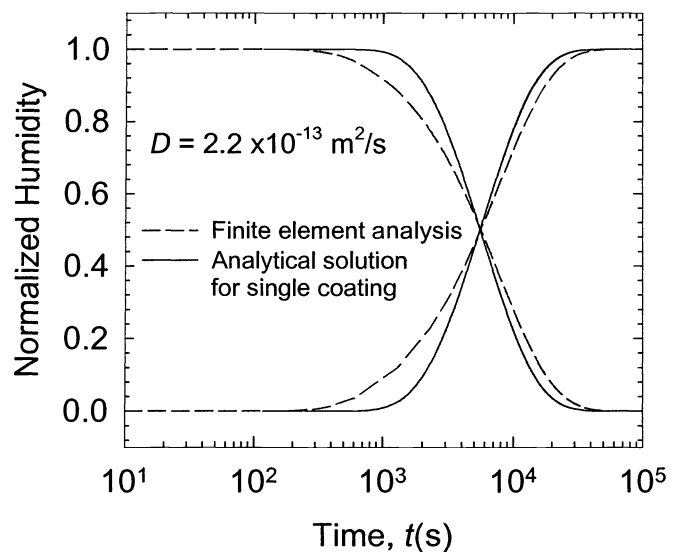


Figure 4. Case 2: dual coated where both coatings have the same diffusion coefficient, but the primary has a higher solubility (10 $\times$ ).

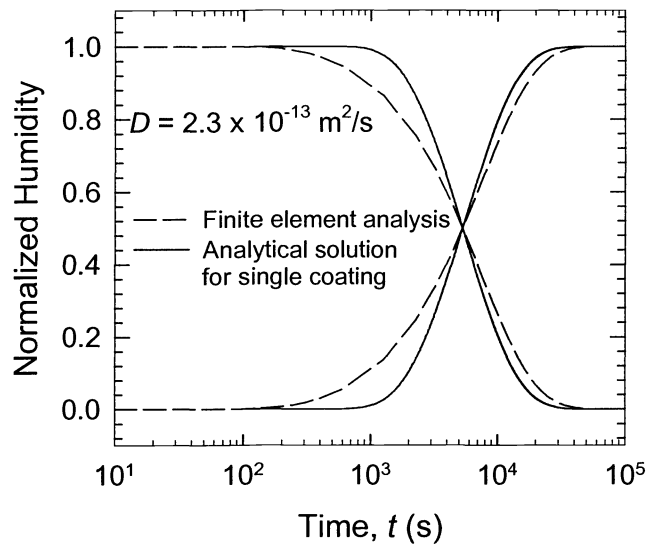


Figure 5. Case 3: Primary coating has a higher diffusion coefficient and solubility.

The FEA results show that increased solubility changes the rate of diffusion and the shape of the concentration *versus* time behavior. These results can be used to help determine if an accumulation layer, which would have an effectively very high local solubility, has formed between the glass and polymer surface and could also help to describe the results that Bouten<sup>6</sup> observed when drying fibers to very low humidity.

#### 4. CONCLUSIONS

Finite element analysis was used to model the diffusion of moisture through polymer coatings giving comparable results to the analytical solution for cylindrical symmetry. This analysis also verified the expected result that the diffusion into the ends of the coating does not significantly affect the diffusion rate because the ends only affect concentration on a length scale comparable with the coating thickness.

For dual coated fibers it was found that if the primary coating has a different diffusion coefficient than the secondary, or different solubility for moisture, the diffusion curve broadens. This erroneously appears to indicate a deviation from linear Fickian behavior. Thus FEA is a useful tool for modeling diffusion through multilayer coatings and is important for correct interpretation of experimental results.

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