Turkish J. Eng. Env. Sci.
32 (2008) , 253 – 263.

An Uncertainty Method for Moisture Concentration Assessment in Composite Material Plates T300/5208 under Hygrothermal Conditions Effects

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Received 06.04.2007

Abstract

Matrix polymer composite materials are increasingly required especially for high-performance applications. Sadly, these materials are subjected to degradation in a given environment. Indeed, the aromatic structure of the matrix makes it not only sensitive to the effects of certain solvents, but also susceptible to the reorganization of chains over time. This last phenomenon is known as ageing and leads to the loss of initial physical and mechanical properties of the matrix.

Degradation problem due to the exposure to the environment is one of the most critical affecting phenomena in the use of composite materials in various structures.

The objective of this work is to present a study of temperature and humidity effects on plates of polymer matrix materials. The hygrothermal effects are analysed in a study of water concentration evolution through the plate thickness (Fick's model). The second part of this work deals with the determination of water concentrations in composite material plates using a probabilistic method. It also aims to demonstrate the importance of probabilistic methods in the field of composite materials and to study uncertainty regarding the unpredictable variables.

Key Words: Composite materials, Hygrothermal behavior, Aging, Reliability analysis, Environmental cyclic conditions, Fick's Model, Moisture diffusion.

Introduction

Under cyclic environmental conditions, temperature and humidity produce considerable stresses, which may cause the deterioration of the polymeric matrix composites due to the increase of humidity concentration and temperature variations. Recently, in particular for the aeronautical application; Loverich et al. (2000) investigated the lifespan for polymer composites subjected to cyclic loading at high temperatures.

Ellyin et al. (2003) studied the influence of humid environment, with temperature and cyclic loading, on the laminated composites (Glass/Epoxy). Vaddadi et al. (2003) evaluated the effect of humidity absorption for composite materials exposed to a humid environment. His results provide detailed numerical analyses, which strongly simulate the real heterogeneous microstructure of the composite. Qi and Hertzberg (1999) present a technological approach for investigating the residual stresses after the impact and cyclic hygrothermal conditions for the carbon/epoxy laminated composites.

Gopalan et al. (1988) determined the effect of humidity absorption in the hybrid glass/carbon /epoxy composites using Fick's model to measure the degradation level induced by the humidity in matrix and the interface between fiber and matrix.

Ishai et al. (1995) presented a work on the long-term durability for hybrid sandwich composite plates under various hydrothermal conditions, representing ambient environmental extreme conditions and the evaluation of the durability on the damage tolerance for the proposed composite.

The composite material considered in this study is the graphite/epoxy, which is particularly used in aeronautical and space structures, exposed to accidental operation or environmental heating leading to the degradation of these materials and thus affecting the integrity of the composite structures (longitudinal and transverse strengths, modulus of elasticity, etc.) (Griffis et al. 1981; Colling and Mead, 1988).

In fact, the intensive hydrothermal behavior deteriorates the physicochemical state of the resin. This phenomenon is transformed into microscopic-damages in the form of interlaminated cracks. From another point of view, this leads to non-coherence of the interface between fibers and matrix caused by conditional thermal stresses (Chang, 1987). Several studies showed the importance effects of humidity on the mechanical properties of composite materials and on their long-term behavior (Shen and Springer, 1977; Springer, 1981). A large part of this work is devoted to the characterization of the static residual properties or the performances in fatigue after ageing in wet atmosphere or immersion.

In fact, the polymeric resins absorb the humidity of the atmosphere; water exists in the form of molecules or forms grouping of molecules of the hydrogen bonds with polymer. Thus, in many cases, the kinetics of water absorption can be described in a correct way using Fick's model for humidity diffusion in the composite materials (Shen and Springer, 1976; Shirrell, 1978). Consequently, the material should generally be considered as complex structures subjected to a uniform temperature rather than non-uniform distribution of humidity. Additionally, a detailed attention must be lent to the calculation of the non-uniform concentration in humidity. Actually, it seems that there are few studies which have investigated the hygroscopic effects in composite material plates due to the non-uniform distribution of humidity. Hahn and Kim (Adda-Bedia et al. 1992) studied the distribution in 3 cases of stacking order; Benkeddad et al. (1995) developed a numerical procedure to calculate the hygroscopic components of a plane, specifically in the laminated plates.

In the present work, a numerical model is built for the prediction of moisture diffusion under cyclic environmental conditions on the basis of the classical assumptions for non-uniform transient concentration, as described by Springer, (1981). This model is then applied to evaluate the scatter of the moisture concentration in terms of input parameter uncertainties, especially for those related with material diffusivity. It is shown that the moisture distribution is far from being normally distributed, and extreme distributions should be considered to better represent the scatter of the graphite/epoxymoisture concentration. This result is very useful for reliability analysis of composite plates under climatic conditions. In the following sections, Fick's model is presented, followed by the numerical solution under environment cycles. Then, Monte Carlo simulations are described and applied to graphite/epoxyplates with different thickness and model uncertainties.

Fick's Model for Water Absorption

The majority of diffusion models used to estimate the kinetics of humidity absorption relies on Fick's model. This model represents a simple approach for humidity diffusion (phenomenon of water absorption). It is based on the assumption that there is proportionality between mass transfer diffusion through a surface and its concentration gradient perpendicular to this surface.

The equations of diffusion (Springer, 1981) suggested for modeling the water absorption phenomenon were obtained starting from a comparison made between heat transfer by conduction and mass transfer (Eq. (1)):

$$\vec{F} = -\bar{\bar{D}}.\vec{\nabla} C << \text{Fick's model} >> \qquad (1)$$

with $\vec{F}$: Mass flow (flow of diffusing phase per unit of area), C (water concentration in the equation of the diffusion)

The general formulation of Fick's model can be written as (Eq. (2)):

$$\frac{\partial C}{\partial t} = div\left(D.\vec{\nabla} C\right) \tag{2}$$

where $C = C\left(\vec{X}, t\right)$ and $\vec{X} = (x,\, y,\, z)$

In the case of anisotropic solid and diffusivity independent of the concentration, Eq. (2) is written in the following way in Cartesian co-ordinates (Eq. (3)):

$$\frac{\partial C}{\partial t} = D_x.\frac{\partial^2 C}{\partial x^2} + D_y.\frac{\partial^2 C}{\partial y^2} + D_z.\frac{\partial^2 C}{\partial z^2} \tag{3}$$

For isotropic case we can write:

$D_x = D_y = D_z = D$

Solution of Fick's law for Composites Plates

For infinite composite plates, Eq. (3) becomes one-dimensional and Eq. (4) is written:

$$\frac{\partial C}{\partial t} = D_x.\frac{\partial^2 C}{\partial x^2} \tag{4}$$

Equation (4) is generally solved by supposing that the plate is exposed to a wet environment at both sides. It is supposed that in the initial state, the concentration in the plate is constant and equal to C*i*. Also, it is supposed that the concentration on both sides of the plate remains constant. The initial boundary conditions are as following:

For $-h/2 < x < h/2$ and $t = 0$ $C = C_i$

For $x = \pm\, h/2$ and $t > 0$ $C = C_\propto$

By assuming constant diffusivity of the material equal to D, the analytical solution of Eq. (5) developed by Crank (1983) is given by:

$$\frac{C - C_i}{C_\infty - C_i} = 1 - \frac{4}{\pi}\sum_{n=0}^{\propto}\frac{(-1)^n}{2n+1}\cos\frac{(2n+1)\,\pi x}{h}$$
$$\exp\left[-\frac{D\,(2n+1)^2\,\pi^2 t}{h^2}\right] \tag{5}$$

with C = C (x, t): concentration with x-coordinate and time t; $C_\propto$: water concentration with saturation (after an infinite time); h: thickness of the plate; D: diffusivity of the plate in direction x, depending only on the presumed constant temperature of the medium.

Consequently, Eq. (6):

$$G = \frac{m - m_i}{m_m - m_i} = 1 - \frac{8}{\pi^2}\sum_{n=0}^{\propto}\frac{1}{(2n+1)^2}$$
$$\exp\left[-\frac{D\,(2n+1)^2\,\pi^2 t}{h^2}\right] \tag{6}$$

with G: mass fraction of absorptive water; m: absorptive water mass at the moment t; m_m: absorptive water mass at saturation; m_i: initial water mass in material.

In practice, the percentage of humidity contained in the plate at time t is defined by Eq. (7):

$$M = \frac{W - W_i}{W_i}.100 = \frac{m}{m_i}.100 \tag{7}$$

where M: increase in weight (quantity of absorptive water) in %; W: material weight at the moment t; W_i: initial material weight (dry); m: absorptive water mass at the time t; m_i: initial water mass in the material.

To facilitate the use of the model, Shen (1981) proposed an approximate expression given by Eq. (8):

$$G = 1 - \exp\left(-7.3.\left[\frac{D.t}{h^2}\right]^{0,75}\right) \tag{8}$$

Concerning diffusivity D, which seems independent of the relative humidity and only a function of the temperature according to Arrhenius's model (Eq. (9)):

$$D = D_0\exp\left(-\frac{E}{R.T}\right) \tag{9}$$

with D_0: index of permeability; R: ideal gas constant equals 8.32 $J.mol^{-1}.K^{-1}$; E: activation energy; T: temperature; D_0 and E are material constants (Loos and Springer, 1981).

For a given material, the maximum quantity of water M_m retained at saturation depends on the temperature and the humidity content of the environment. For materials exposed to humid air, M_m seems independent of the temperature and depends only on the relative humidity according to a power model (Eq. (10)):

$$M_m = a\,(H)^b \tag{10}$$

where A and B are material constants; H: relative humidity of the medium in %.

The preceding relationship is a simple approximation. Actually, M_m varies with the temperature;

this variation is often about 20% compared to the approximated value.

In different cases, it is noticed that the variations of M_m are asymptotically increased to reach a maximum value.

The increase in M_m is a consequence of the microscopic cracks developed in material and, probably, with a process of diffusion “non-Fick’s model”. The decrease of M_m value results from the loss of material weight due to the erosion of the matrix.

The mass D (diffusivity) characterizes the speed at which the concentration in humidity changes inside the material. The value of D depends on the material, the surrounding fluid, temperature, and humidity concentration.

Water Absorption in Polymer Plates

A plate made of graphite/epoxyT300/5208 is studied under cyclic environment actions of temperature and humidity, applied on both faces (Figure 1). Three plate thicknesses have been considered: 11.7, 12.7, and 13.7 mm. The characteristics of T300/5208 are given in Table 1 (Shirrell, 1978; Adda-Bedia et al. 1998):

Table 1. Characteristics of the T300/5208 material.

ρ(kg/m^3)	C_∞	DA1 (mm^2/S)	DA2 (K)	$CC1$	$CC2$
1600	1.23	0.57	4993	0.015	1

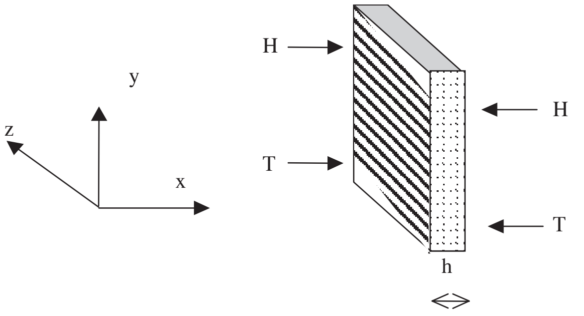

Figure 1. Plate subjected to temperature and moisture in the x-axis direction.

The studied plates are subjected to the cycles of temperature and relative humidity, as illustrated in Figure 2. For a period of 144 h, the temperature varies between 27 °C and 142 °C while the relative humidity oscillates between 14% and 82%. The selected cyclic variations of temperature T and relative humidity RH are presented as a function of time t before and after flight. These conditions are the same ones as described by Springer (1981). It gives a test plane where temperature and moisture are considered as cyclic variables during 144 h (i.e. 6 days); the normal atmospheric conditions are fixed during the first 123 h, followed by 11 h flight, and once more the normal atmospheric conditions are fixed during the last 10 h. The ambient relative humidity increased when the temperature decreased, except during the flight where moisture decreased to zero. Also, it can be said that the relative humidity is constant (82%) except during the flight time. The plate is exposed to 2000 periods reproducing the cycles in Figure 2,;hich makes 288,000 h (i.e. 32 years and 11 months). The analysis aims to compute the moisture concentration through the plate thickness, as a function of time. This can be carried out by numerical integration of Fick’s laws for water absorption in composite plates (Shen and Springer, 1976; Shirrell, 1978).

For 3 constant temperatures: 27, 37, and 47 °C, Figure 3 shows the moisture concentration in graphite/epoxyplate in humid air subjected to constant relative humidity of 82%. It can be observed that, in the first stage, the water absorption increases monotonically until saturation, where the increase of weight is fast and regular. The second stage is rather constant, which characterizes a “quasi-nil” diffusion. The moisture concentration curves present an inflection point, which becomes more visible for thick plates. After an acceleration part, the moisture rate is almost linear (up to 40% of the saturation moisture at temperature 47 °C). Then, the water absorption rate decreases progressively until the maximum moisture saturation. From these curves, it can be observed that higher temperature increases the moisture concentration proportionally. In addition to the diffusivity increase (slope of the curve), the long-term water concentration increases with temperature, from 0.39% at 37 °C to 0.52% at 47 °C when $h=$ 11.7 mm (i.e. 13% of increase). This is explained by the fact that diffusion is a phenomenon, which was driven by high temperatures.

For $h=$ 12.7 mm, the influence of temperature is visible in reducing the time to saturation: at 47 °C the material is saturated at 1000 cycles (i.e. 17 years) while at 27 °C, saturation occurs at 600 cycles (i.e. 10 years). Concerning the saturation level, it is 0.28% at 27 °C, 0.39% at 37 °C, and 0.52% at 47 °C. We can conclude that the moisture concentration is around 0.39%, which can be expressed as the concentration at equilibrium increases with time and diffusivity increases with temperature.

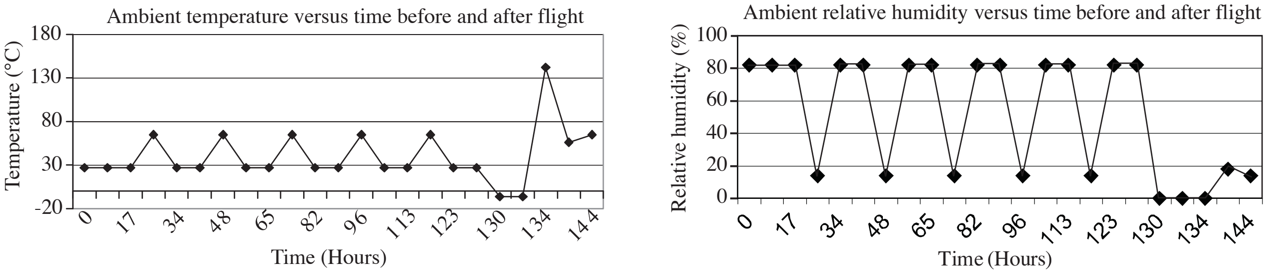

Figure 2. Applied environmental temperature and relative humidity cycles.

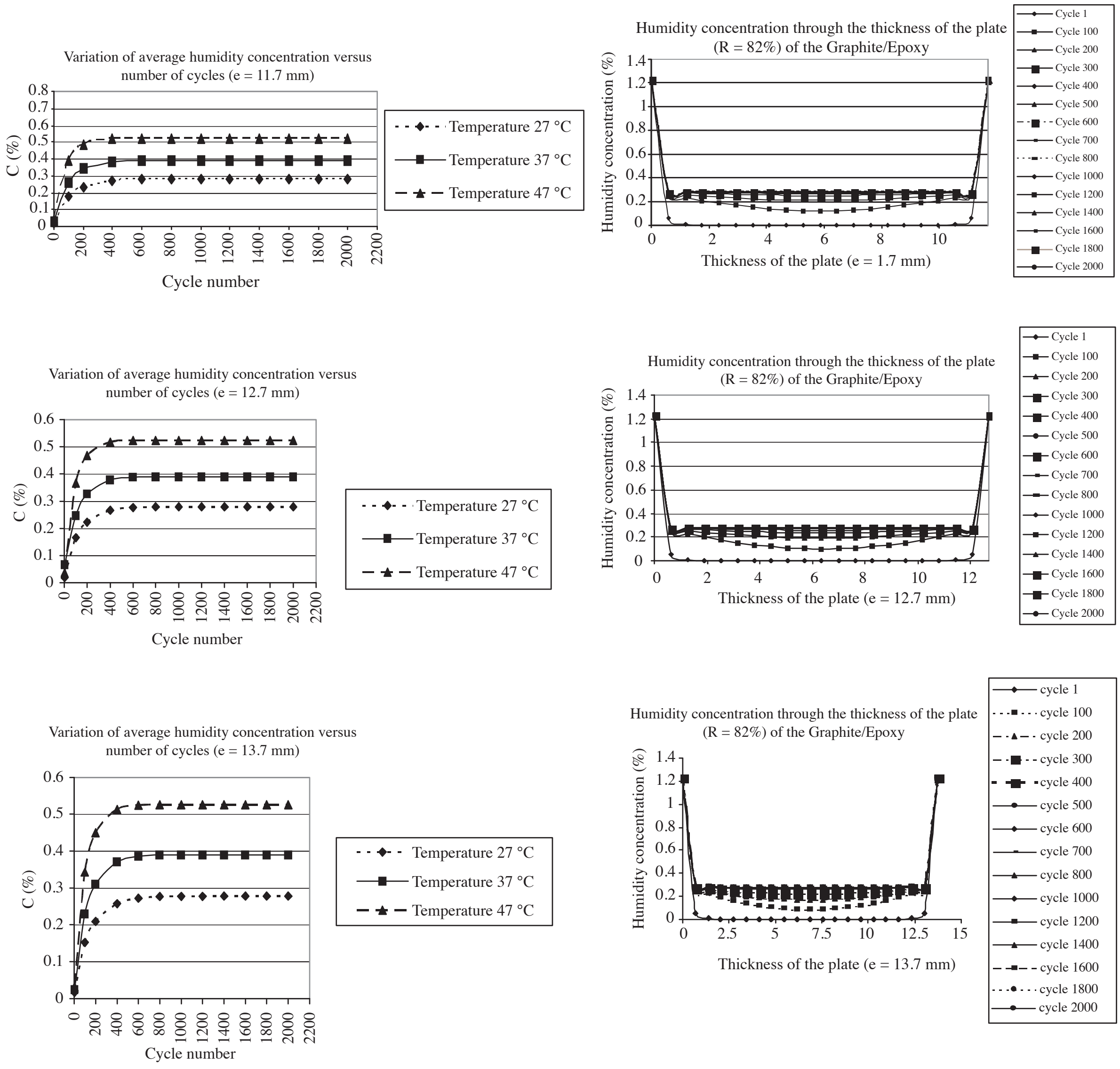

Figure 3. Moisture concentration as a function of cycles and thickness.

The water concentration in graphite/epoxy plates increased by 0.28% at 27 °C, which is less than the concentrations at 37 °C and 47 °C. These results show that the importance must be given to the evolution of the concentration according to the number of cycles and time.

Concerning the 3 thicknesses at 27 °C, the Figure 4 shows the moisture concentration inside the plate at different loading periods. It can be observed that, for the studied material, the moisture concentration does not exceed 0.30% regardless of the ageing time.

Up to 27 °C temperature, we noticed that the necessary time to arrive to the equilibrium is very long for *h*= 13.7 mm. The profile at equilibrium is obtained after approximately 1200 cycles. This result confirms the need for using a method of accelerated calculation.

Also we noticed, after 2000 cycles, that the water concentration does not exceed 0.52%. This value is much lower than the maximum concentration that is equal to 1.23%.

Moisture Scatter in Graphite/Epoxy Plates

In order to study the scatter of the moisture concentration, the graphite/epoxy plates with the thickness of 13.7 mm were subjected to 20 cycles of 144 h, as illustrated in Figure 1, which makes 120 days of exposure. The thermal conductivity is set to 250 W/m.K. The sensitivity study was carried out by introducing uncertainties in the input variables of the model. In other words, the selected input variables were considered as normally distributed random variables with a specified coefficient of variation (COV). For each configuration, 1.000 random simulations have been carried out, leading to 1.000 realizations of the model output, given by the moisture concentration. The statistical analysis of the response allows us to determine the influence of each parameter on the concentration scatter.

The analyzed configurations are related to the thickness, the thermal conductivity, and the Fick's law parameters DA1 and DA2.

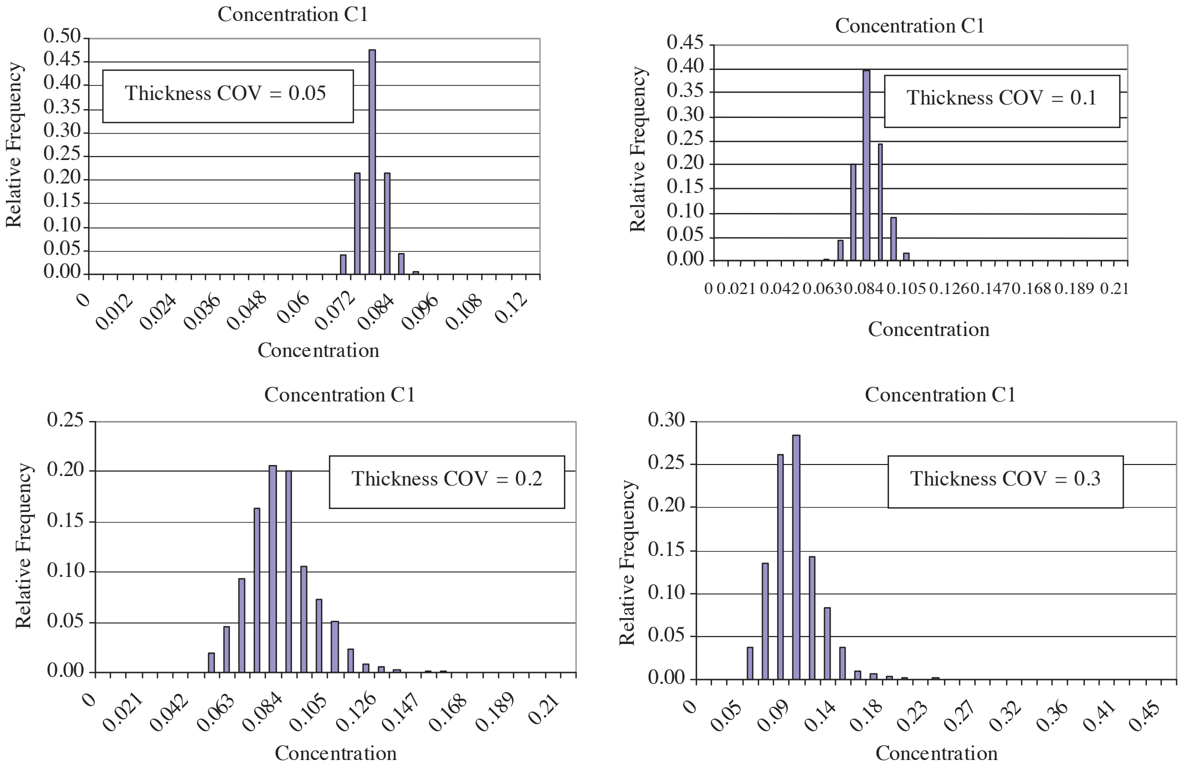

Figure 4. Distribution of moisture concentration for different COV of plate thickness.

Plate thickness

In this section, the influence of thickness uncertainties is considered. As indicated in Table 2, the coefficient of variation (COV) of the moisture concentration is practically equal to the COV of the plate thickness. The mean moisture is slightly increased with the COV of the thickness. The comparison of the distributions in Figure 4 shows that, although the distribution is symmetrical for low thickness dispersion, it becomes significantly skewed for large thickness dispersion. In this case, the use of normal distribution to model the moisture is not valid even when the thickness itself is normally distributed.

Parameter DA1

The scatter is now considered for the parameter DA1. As shown in Table 3 and Figure 5, the dispersion of the parameter DA1 has insignificant effect on the mean moisture concentration and very low effect on its COV. Hence, this parameter is not critical for material characterization. However, the observation of the skewness values shows a slight lost of distribution symmetry for large dispersion of DA1.

Parameter DA2

As it could be expected, the parameter DA2 has a very large influence on the mean moisture concentration. The present study shows that the impact of this parameter is even larger for the dispersion and for the shape of the moisture distribution, which is mandatory for a reliable design of such a plate. When the parameter COV varies from 0.01 to 0.07 (Table 4), the mean moisture increases from 0.089 to 0.122 and its COV is multiplied by 7. It is noticed that the COV of the moisture concentration is almost 10 times more than the COV of DA2. The observation of the distributions in Figure 6 (and the values of the kurtosis and the skewness) shows that the distribution becomes significantly non-symmetrical with a very long tail on the right-hand side. This is very important for reliable design as it increases significantly the probability to have high concentration values, leading to material deterioration. For a COV of DA2 equal to 0.07, even with a mean moisture of 0.122, concentrations over 0.6 have been observed among the 1,000 samplings; which means that even with a safety factor of 5, the probability to exceed 0.6 is about 10^{-3}, which is very high for practical engineering design. This analysis shows that even that deterministic computation can give indications on the parameter sensitivity, it is not sufficient to

Table 2. Influence of the thickness dispersion.

Thickness COV	Mean	COV	Kurstosis	Skewness
0.03	0.0739	0.0287	0.3598	0.0748
0.05	0.0741	0.0481	0.3928	0.1182
0.06	0.0742	0.0579	0.4045	0.1480
0.07	0.0743	0.0676	0.4208	0.1774
0.08	0.0745	0.0774	0.4488	0.2099
0.10	0.0748	0.0968	0.5176	0.2773
0.20	0.0771	0.1938	1.1385	0.6231
0.30	0.0810	0.2907	2.2987	0.9897

Table 3. Influence of the dispersion of DA1.

COV of DA1	Mean	COV	Kurtosis	Skewness
0.02	0.0885	0.0099	0.1848	0.1229
0.03	0.0885	0.0147	0.2103	0.1319
0.04	0.0885	0.0196	0.2545	0.1485
0.05	0.0885	0.0245	0.2886	0.1612
0.06	0.0885	0.0294	0.2952	0.1785
0.1	0.0883	0.0491	0.3045	0.2416

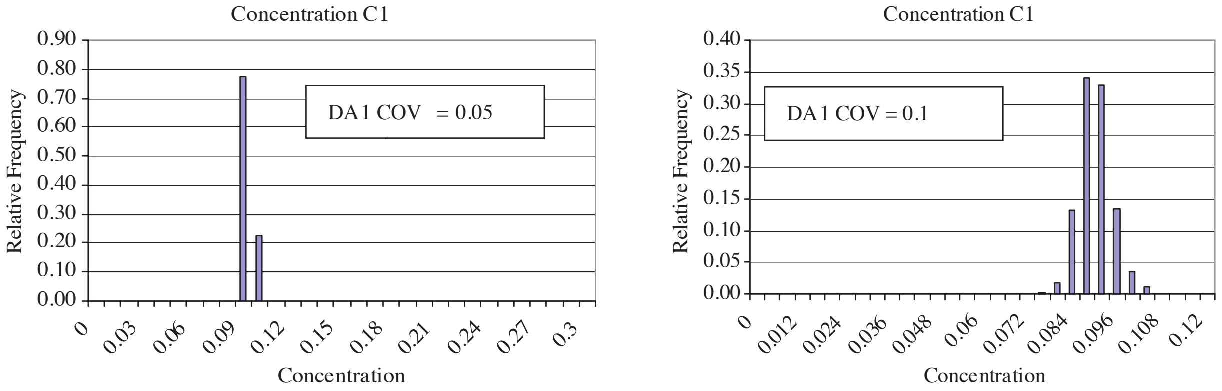

Figure 5. Impact of **DA**1 on the moisture concentration distribution.

Table 4. Influence of the dispersion of DA2.

COV of DA**2**	**Mean**	**COV**	**Kurstosis**	**Skewness**
0.01	0.089	0.118	0.501	0.298
0.02	0.092	0.237	1.296	0.694
0.03	0.095	0.357	2.644	1.101
0.04	0.100	0.480	4.085	1.467
0.05	0.106	0.597	4.642	1.689
0.06	0.114	0.703	4.336	1.759
0.07	0.122	0.790	3.458	1.708

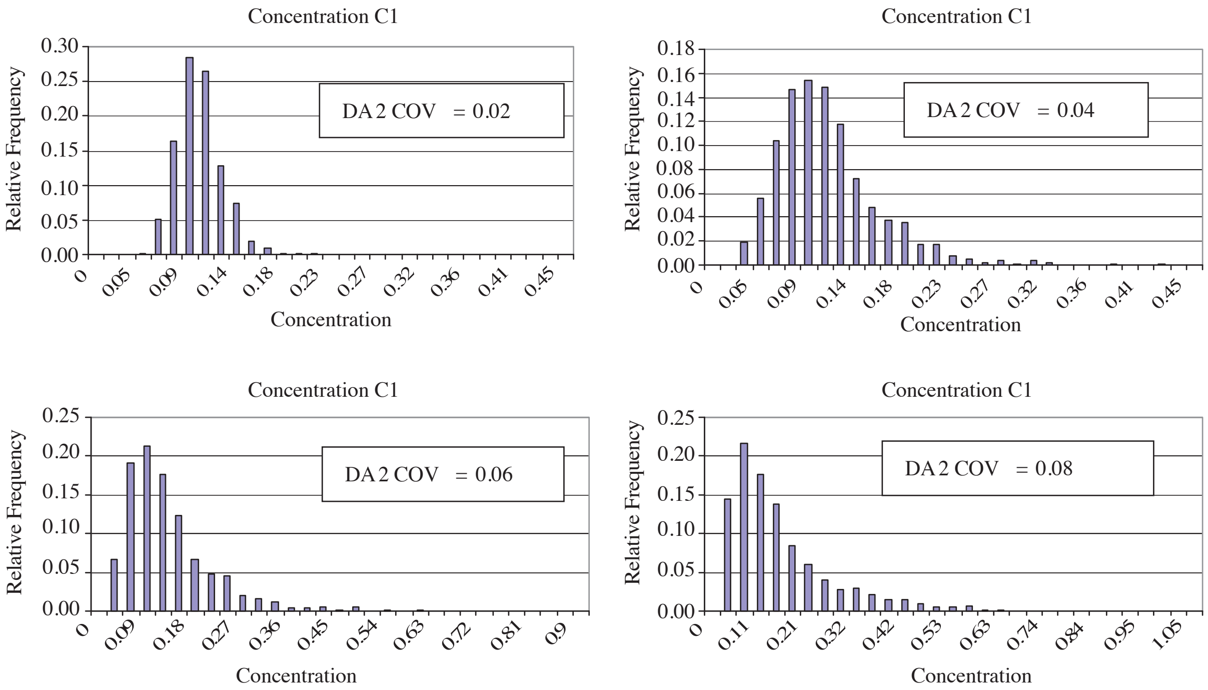

Figure 6. Impact of DA2 on the moisture concentration distribution.

predict the dispersion of the moisture, and hence cannot guarantee the required reliability level. The probabilistic analysis is the only way allowing a realistic modeling of the structural reliability, because it takes account for the uncertainty propagation in the physical behavior.

Coupled uncertainty analysis

Table 5 gives the interaction of the dispersions of the 3 parameters respectively: the plate thickness, the thermal conductivity, and the DA1 parameter. The parameter DA2 has been discarded at this stage due to its highly dominant role, which does not allow us to understand the coupled roles of the other parameters. The observation of Table 5 leads to the following remarks:

- The influence of thermal conductivity is still insignificant, when compared with the mean value of 0.77 for the thickness alone and with the mean of 0.883 for DA1 alone.

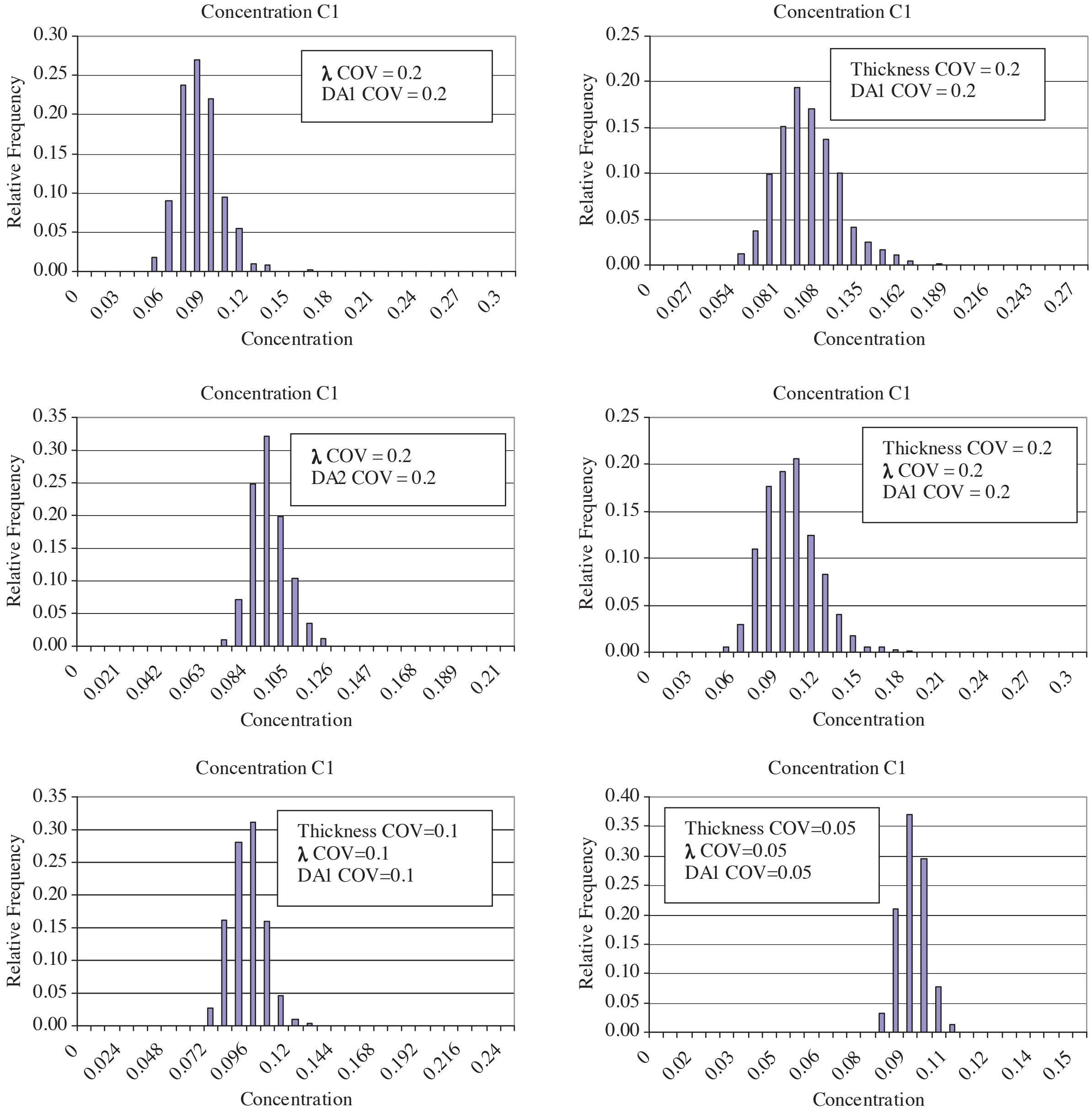

Figure 7. Coupled effects of parameter dispersions.

Table 5. Influence of coupled dispersions.

Thickness	Thermal conductivity conductivity	DA1	Mean	COV	Kurtosis	Skewness
0.2	0.2	0	0.0769	0.1894	0.9426	0.6153
0.2	0	0.2	0.0922	0.2164	0.7676	0.6345
0	0.2	0.2	0.0884	0.0996	0.0043	0.3793
0.2	0.2	0.2	0.0906	0.2158	0.7604	0.6195
0.1	0.1	0.1	0.0889	0.1084	0.1648	0.2773
0.05	0.05	0.05				

- The thermal conductivity, even low level of it, plays a role of a reducer for the concentration phenomenon (for the mean and for the dispersion).

- The coupled effect of thickness and DA1 increases both the mean value and the coefficient of variation of the moisture concentration. In this coupled effect, DA1 gives a lower bound for the mean and the thickness gives the lower bound for the COV. The coupled parameters are higher than the maximum of each parameter for the 2 effects separately.

Figure 7 shows the moisture distribution where it can be easily observed that the coupling of the thickness and DA1 dispersions increases significantly the moisture dispersion. The distribution skewness is higher when the thickness dispersion increases. It seems appropriate to model the moisture concentration by lognormal or Weibull distributions.

Conclusion

This study achieved its objectives; the water concentrations in the studied plates do not exceed 0.7%, which is still less than the maximum concentration C_∞. Also it is noticed that the lifespan of the graphite/epoxy material can reach up to 33 years of service, which is achieved by a simulation about 2000 cycles. In every cycle we had 144 h, i.e. 6 days. Therefore, the sum of them gives the duration of about 33 years of life span. Higher life span is continuously demanded as claimed in the applications of high performances, and, more particularly, in aeronautics applications, as the growth of the composites in this sector is increased to approximately 5% in volumes of materials per year.

Under cyclic environmental conditions, the model shows the influence of the temperature and plate thickness on the rate and the limit of concentration. From the probabilistic point of view, the obtained results underline the sensitivity of the moisture concentration to the scatter of the diffusion parameters DA1 and DA2.

This observation leads to orient the test measurement on the parameter DA2, rather than DA1 or the thickness. Even with a very low coefficient of variation of DA2, the scatter of moisture concentration is highly amplified.

An important practical conclusion is that increasing the plate thickness leads to exponential decrease of the high moisture concentrations probability, which leads to a significant increase of lifetime reliability.

List of principal symbols

t	time
T	temperature
H	humidity
D	diffusion coefficient
DA1	the permeability index of the layer (mm^2 s^{-1})
DA2	constant (°K)
R	the constant of perfect gases
E	the activation energy of the diffusion process
M	fractional (or percent) weight gain
c	concentration of diffusant
D	diffusion coefficient for the material (diffusivity)
W	the weight of the wet specimen
W_i	the weight of the dry specimen
m_i	the initial weight of the moisture in the material
m_m	the weight of the moisture in the material when fully satured
Cov	coefficient of variation

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