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Characterizing the toughness of an epoxy resin after wet aging using compact tension specimens with non-uniform moisture content

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ABSTRACT

Characterizing the change in toughness of polymers subjected to wet aging is challenging because of the heterogeneity of the testing samples. Indeed, as wet aging is guided by a diffusion/reaction process, compact tension samples (defined by the ASTM D5045 standard), which are relevant for toughness characterization but are somewhat thick, display a non-uniform moisture content over the bulk material. We define here a rigorous procedure to extract meaningful data from such tests. Our results showed that the relation between the moisture uptake of the whole sample and the measured toughness was not a meaningful material property. In fact, we found that the measured toughness depended on the locally varying moisture uptake over the cracking path. Here, we propose a post-processing technique that relies on a validated reaction/diffusion model to predict the three-dimensional moisture state of the epoxy. This makes identification of the variation in toughness with respect to the local moisture content possible. In addition, we analyze the fracture surface using micrography and roughness measurements. The observed variations in toughness are correlated with the roughness in the vicinity of the crack tip.

1. Introduction

Composite materials used in advanced aeronautics and space applications are exposed to severe environmental conditions. One of the most common environmental conditions that leads to degradation of such materials is hygrothermal aging, which takes place when the material is exposed to a wet environment at moderate or elevated temperatures. Interactions between the water in the environment and the polymer backbone of the resin can result in various modifications of the resin's properties [1–5]. Here, we focus on changes in the fracture properties of the epoxy resin, and more specifically on its toughness, when it is exposed to a wet environment.

Various experimental techniques can be used to characterize changes in the toughness of epoxy resins. The ASTM D5045 standard, which relies on compact tension (CT) testing, is a universally accepted method to measure fracture toughness. It has been used previously in the study of wet aging. Alessi et al. [6] used CT testing to study the influence of hydrothermal aging on the fracture toughness of radiation-cured bis(4-glycidyloxyphenyl) methane (DGEBF) with polyethersulfone (PES) as the toughening agent. In their study, the critical stress intensity factor (K_{Ic}) decreased from 1.2 MPa.m$^{0.5}$ to 0.9 MPa.m$^{0.5}$ after 1500 h at 70 °C. Alessi et al. [7] also studied an epoxy/anhydride system and observed a 62% reduction in K_{Ic} after a week in distilled water at 70 °C. Although the ASTM D5045 standard method produces useful results, a problem in using it to identify toughness after hygrothermal aging is that the size of the samples, as defined by the standard, is massive. The moisture state is non-uniform in large samples, making it difficult to draw conclusions about changes in the fracture toughness with respect to the moisture content.

To obtain more representative moisture/toughness relations that could be used in three-dimensional (3D) simulations of moisture-induced failure, other sample configurations can be used, such as those introduced for the essential work of fracture (EWF) method. EWF uses double edge notched tension (DENT) thin specimens in which the water content is much more uniform than in CT samples. EWF has been used to measure plane stress fracture toughness of aged polymers in previous studies [8,9] and is based on the assumption that dissipation occurs either by plastic deformation or by the fracture process. To distinguish between the two processes, several configurations with different neck lengths are needed. This method works well on ductile polymers, but it cannot be applied to brittle epoxy resins, since it requires full yielding of the neck before fracture. The samples considered in this study, based on anhydride cured epoxy, display brittle failure, which

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makes it difficult to utilize the EWF method. Chaléat et al. [8] made use of both ASTM D5045 and EWF techniques to measure the fracture toughness of polymer samples that were in a brittle-to-ductile transition due to hygrothermal aging. In that study, EWF was used to characterize the ductile samples, while ASTM D5045 was used to characterize the brittle ones. Barany et al. [9] used EWF to study the influence of hygrothermal aging on the fracture toughness of polyester sheets although the EWF method is not as robust as the ASTM standard as reported by Clutton [10]. Although they were able to reduce some of the scattering in round robin test results, they also observed that several major uncertainties are inherent in the method.

Since the EWF method is not as robust as ASTM D5045, and cannot be used on our brittle samples, we chose to use the ASTM D5045 method, but with full awareness of the non-uniformity of the moisture state. Our objective was to find a viable post-processing procedure to account for this problem.

Here, we used experimental results as well as an accurate simulation of the heterogeneous moisture content concurrently. The CT samples were subjected to hygrothermal aging inside a climatic chamber and tested at various aging states. After toughness testing, we analyzed the fracture surfaces of the samples by micrography and roughness measurements. To determine the water distribution across the samples, we performed simulations based on a validated diffusion-reaction model of water absorption. Understanding of the spatial distribution of the water content made it possible to identify the relation between the observed toughness and the water content at the point where the cracks started to propagate in an unstable manner.

In the following section, we describe the material, samples, and experimental method related to both fracture toughness identification and surface analysis. In Section 3, we provide details on the simulations of water absorption and crack extension. In the fourth section, we present the experimental and simulation results. Finally, we develop two main points in the discussion: (i) the necessity of considering a local approach for the correct identification of intrinsic material parameters, and (ii) the correlations between the toughness, roughness, and micrography.

2. Materials and methods

2.1. Material formulation and samples

The material used in this study was EPOLAM 2063 (Axson Technologies), a mixture of cycloaliphatic epoxy resin and a diglycidyl ether of bisphenol-A (DGEBA). This formulation was previously characterized [11–13]. Its dry elastic mechanical properties (Young's modulus and Poisson ratio) are $E = 3.1 \pm 0.1$ GPa and $\nu = 0.3$. We fabricated 48 CT samples (Fig. 1) by casting the liquid epoxy system in silicone rubber molds previously coated with an anti-stick agent (MANN EasyRelease). The curing cycle was as recommended by the manufacturer: 6 h at 80 °C and postcuring at 180 °C for 8 h. Afterwards, the samples were sanded and polished with different grit papers (320, 500, 1000, 2000) until the desired thickness was obtained. Finally, pre-cracks were introduced by sliding a fresh razor blade into the root of notches to obtain sharp initial crack tips (details in Fig. 1).

2.2. Aging conditions

We placed the samples in a climatic chamber (Tenney T2RC, Thermal Product Solutions) at a constant temperature of 70 °C and a constant relative humidity of 90% R.H. Periodically, we extracted three samples from the chamber to perform fracture tests for that specific aging time. We made 15 extractions at intervals of 12 h (extractions 1–6), 24 h (extractions 7, 8), and 48 h (extractions 9–15). The baseline properties before aging were obtained separately by performing the fracture test on non-aged dry samples.

2.3. Characterization

For each extraction, we characterized the toughness, the global moisture uptake of the CT samples and the surface morphology of the fracture surface.

The CT samples were tested following the ASTM D5045 [14] standard for measuring plane strain fracture toughness in polymers. We utilized a crosshead speed of 0.3 mm/min (INSTRON 5882 Universal Testing Machine). We calculated K_{Ic} as a function of the geometrical parameters and the maximum load (P_Q) during the test (Eq. (1)):

$$K_{Ic} = \frac{P_Q}{BW^{1/2}} f(x), \tag{1}$$

where $W = 30$ mm, $B = 7.5$ mm and $f(x)$ is defined by the standard as:

$$f(x) = \left(\frac{2+x}{1-x^{3/2}}\right)\left[0.886 + 4.64x - 13.32x^2 + 14.72x^3 - 5.6x^4\right], \tag{2}$$

x is the ratio a_0/W, and $a_0 = 15$ mm is the initial distance between the crack front and the pins (Fig. 1). We conducted the toughness

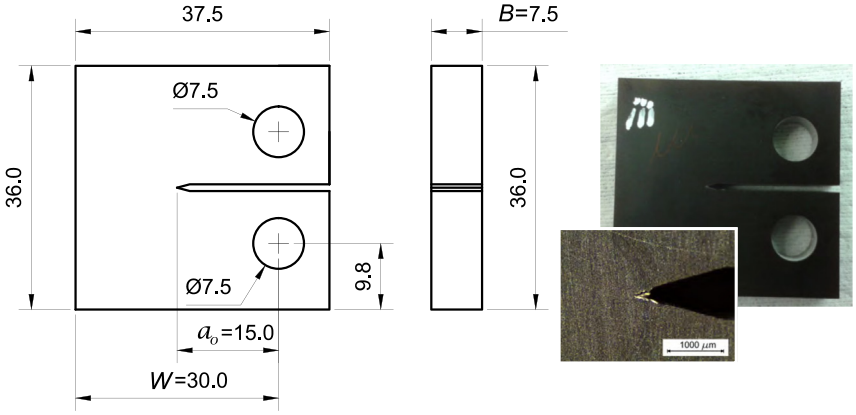

Fig. 1. Geometrical specifications of the standard CT sample used in the study and details of the initial crack tip.