CHARACTERIZATION OF OXIDATION AND DAMAGE IN POLYMER MATRIX COMPOSITE LAMINATES

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Introduction

The anisotropy of oxidation of high temperature polymer matrix composites (HTPMCs) was first documented by Nelson [1] when he observed that the oxidation process was sensitive to the surface area for the different test specimens that he aged. Subsequently, numerous other investigators (e.g., Tandon, et al. [2]) have also observed the anisotropic nature of oxidation in HTPMCs. The thermo-oxidative behavior of the laminated composite is significantly different from that of the constituents as the composite microstructure introduces anisotropy in the diffusion and oxidation behavior. Additionally, in laminated systems, interlaminar residual stresses (besides the fiber-matrix micromechanical stresses at the ply level), further play an important role in the degradation process. Consequently, the thermo-oxidative behavior of a ply within a multidirectional laminated composite is highly dependent on the orientation of the adjacent plies. For example, work by Tsotsis and Lee [3] indicates that residual stresses arising from aging-induced differential resin shrinkage and interaction between plies of different orientations are found to have a strong effect on the degradation process for plies close to the surface and, especially, near free edges. In addition, severe surface oxidation degradation results in the formation of transverse surface cracks coalescing with fiber-matrix disbonds. These cracks not only reduce strength, but also create enhanced pathways for oxygen to penetrate deeper into the composite.

For this work, carbon fiber-reinforced polyimide matrix composite specimens were aged at elevated temperature in air. Isothermal aging was conducted at 177ºC. Five different laminates are considered, namely, $[0]_{16T}$, $[0/\pm 45/90]_{2S}$, $[0/90]_{4S}$, $[\pm 45]_{2S}$**,** and plain-weave laminates. The influence of ply stacking sequence and composite architecture on oxidation growth is investigated. Light microscopy techniques are used to characterize the oxidative process, while oxidation-induced damage development is examined through fluorescence imaging using dye impregnation.

Testing

For each laminate configuration, two rectangular specimens measuring approximately 125 mm × 12.5 mm × 2.3 mm were used to monitor oxidation propagation and damage development along the length and transverse to the fibers. The isothermal aging was accomplished in an air-circulating oven that provided a continuous replenishment of oxygen in the ambient air by convection through the oven inlet. At specified sparse time intervals, specimens were removed from the ovens, and samples were dry-sectioned from the aged larger specimen, potted in epoxy, polished and viewed under an optical microscope. This allowed monitoring of four of the exposed edges of the large specimen. The large sample was subsequently placed back in the oven to continue the aging process.

Results and Discussion

The oxidation layer formation near the exposed specimen edges changes the optical characteristics of the material, and the oxidized layer is observed by light microscopy. Specifically, the oxidation layer appears as a frame around the composite cross section, and is clearly seen as the lighter oxidized material using standard light microscopy in the grayscale mode.

As an example, Figure 1 compares the fluorescence and dark-field optical microscopy images of $[0]_{16T}$ laminate in a cross section parallel to the 0^o direction after 1500 hours of aging. The fluorescent dye image on the left side shows short fiber matrix debonds along the 0º fiber direction. Experimental evidence suggests that these debond cracks will continue to increase in length, while new cracks continue to develop at the free surfaces of the specimen with further aging. The dark-field image on the right showing the oxidation reveals that the oxidation front is nonuniform, and the oxidation has advanced to a greater extent in the region of the specimen corresponding to the location of the debond cracks. Thus, there is a synergistic coupled effect between damage and oxidation development and growth.

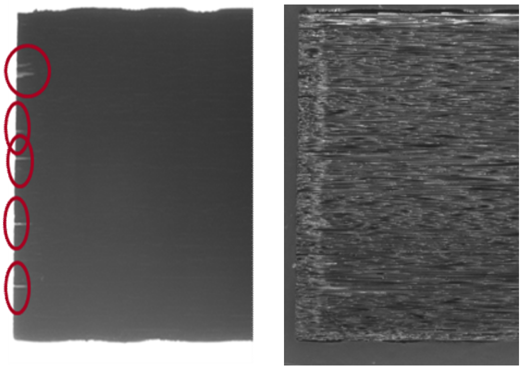
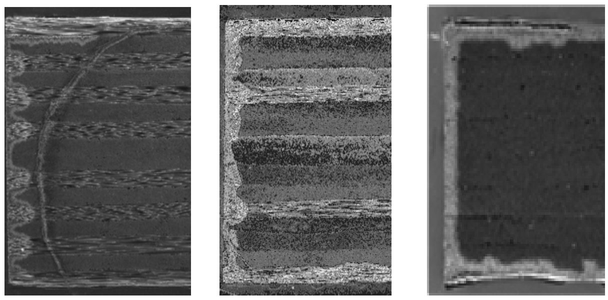
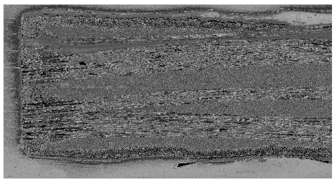


Figure 1: Optical microscopy images in a cross section parallel to the 0º direction in $[0]_{16T}$ laminate

Figure 2 shows the dark-field images of oxidation in (a) cross-ply, (b) quasi-isotropic, (c) angle-ply and plain-weave laminate after 1500 hours of aging. In these images we observe preferential oxidation growth along the fiber paths for the various laminates. The oxidation propagation rate in a ply is strongly influenced by the orientation of its neighboring plies. For example, a 0º ply will tend to increase the oxidation rate in a neighboring off-axis ply as compared to its oxidation rate in a unidirectional composite of the same off-axis orientation. Similarly a 90º ply will tend to decrease the oxidation rate in neighboring plies of different orientation. For the $[0/90]_{4S}$ laminate, the maximum or minimum oxidation extent within an individual ply occurs at the ply midplane due to the fact that the adjacent top and bottom plies in the interior of the laminate are symmetric about the midplane of a ply of interest. However, in a $[0/\pm45/90]_{2S}$ laminate, none of the individual plies (0^o, 45^o or 90^o) have a symmetric distribution of neighboring plies. Hence, the minima or maxima oxidation extent does not occur at the ply mid-plane. In an angle-ply laminate, the oxidized region appears as a picture frame near the exposed free surfaces of the specimen, and the oxidation front quickly becomes non-uniform with aging time because of damage development near the edges. Finally, in a plain-weave laminate, enhanced oxidation is observed along the fiber tows in the warp direction in comparison to the fiber tows in the fill direction. Even though there are some similarities in oxidation growth in a plain-weave and a cross-ply laminate, the tow architecture (namely, tow waviness) introduces some marked differences, as seen in Figure 2.



(a) (b) (c)



(d)

Figure 2: Dark-field images of oxidation in a (a) cross-ply, (b) quasi-isotropic, (c) angle-ply and (d) plain-weave composite

Conclusions

In this work, the influence of ply stacking sequence and composite architecture on oxidation growth is investigated using light microscopy techniques. It is shown that the oxidation propagation rate in a ply is strongly influenced by the orientation of its neighboring plies. It is also shown that alternative pathways for transport of oxygen into the interior of the composite are fiber-matrix debonds and matrix cracks that propagate with the oxidation front. This mechanism for accelerated oxidation is an excellent example of the intrinsic coupling of chemical oxidative aging and damage, which needs to be properly represented in predictive models.

References

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3. Tsotsis, T. K., and S. M. Lee. 1998. “Long-term Thermo-oxidative Aging in Composite Materials: Failure Mechanisms,” *Composites Science and Technology*, 58:355-368.