

EXPERIMENTAL STUDY OF THE PROPAGATION OF DELAMINATION UNDER OXIDIZING ENVIRONMENT AND MECHANICAL LOADING

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Abstract

Oxidation of the polymer plays a crucial role in the degradation of composites in high temperature applications. Even in the absence of external mechanical loads, cracks can develop and allow the oxygen to penetrate deep into the composite. A new experimental method, allowing oxidation-driven crack propagation under different external mechanical loads, is proposed here to gain some insight on the chemo-mechanical coupled behavior of polymer matrix composites.

1. Introduction

The use of polymer matrix composites for high temperature applications (100-300° C) is limited by the polymer oxidation. Oxidation aging is initially confined to a fine surface layer. Even in the absence of external mechanical loads, oxidation eventually leads to cracking, which allows the oxygen to penetrate deep into the composite. The oxidation/cracking coupling is detrimental to the long term resistance of polymer matrix composites under high temperatures.

To the authors' knowledge, the only experimental information currently available on the oxidation/cracking coupling within a composite are self-cracking tests [1, 2]. In these tests, all the mechanisms involved in thermal oxidation (diffusion, reaction, modification of the polymer's properties [3, 4]) are active at the same time, thus it is not possible to identify their different contributions. In particular, the chemical reaction decreases the material toughness and, at the same time, it provides the differential shrinkage which is the driving force for crack propagation. The introduction of an external mechanical load to provide a supplementary driving force for propagation would constitute an additional source of information for the understanding of the oxidation/cracking coupling.

In this paper, a new experimental method is proposed, allowing oxidation-driven crack propagation under different external mechanical loads. The test setup is based on the Climbing Drum Peel (CDP) test, in which a mode I delamination propagates under a constant applied force. Through a simple energy balance, the applied force can be related to the energy available for crack propagation. The classical CDP test is carried out under displacement control and crack propagation occurs when the strain energy release rate G equals its critical material value G_c . In

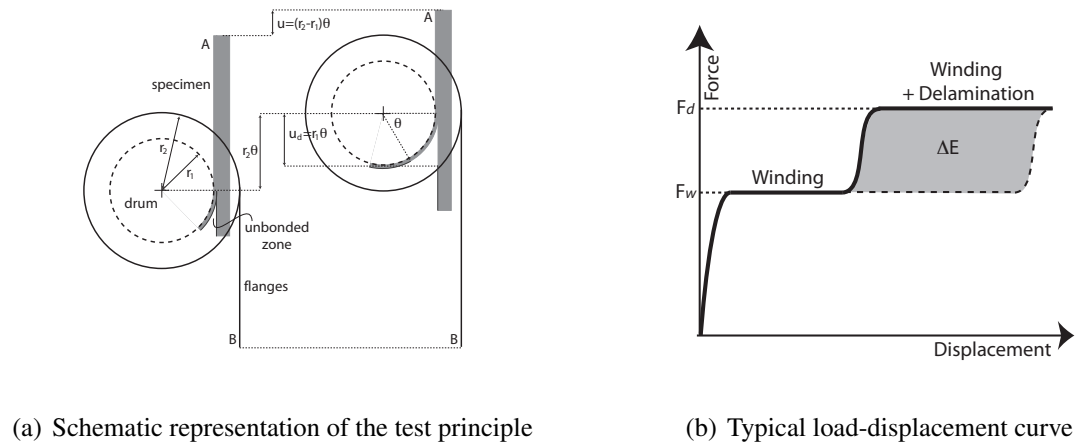


Figure 1. The Climbing Drum Peel (CDP) test.

the present version, a lower constant force is applied to the setup, which is placed in an oxidizing environment. Since the available energy is lower than the one required for crack propagation in a healthy material, the oxidation and consequent degradation of the critical strain energy release rate on the crack front is what drives the crack propagation.

2. The oxi-delamination test

2.1. The Climbing Drum Peel test

The original CDP test (ASTM D1781 [5]) allows to test the mode I bonding strength between a flexible and a rigid adherend. The flexible adherend is wound around a drum having a constant radius, propagating an initial debonding. The applied force is measured and the strain energy release rate is calculated [6]. The CDP is classically used to evaluate the skin-core bonding strength in sandwich structures. To the authors' knowledge this work (partly presented in [7]) is the first example of its adaptation to the delamination of monolithic composites.

The test principle is illustrated in Fig. 1(a). Two radii are defined with respect to the drum's center of mass: an internal radius r_1 , around which the flexible part of the specimen is wound, and an external radius r_2 , along which are placed two flanges which apply the torque required for the drum progression along the specimen. The specimen is made up of two zones: an unbonded zone, whose flexible end is attached to the internal radius of the drum, and a bonded zone along which the crack will propagate during the test. For global equilibrium, the system is generally oriented with a small angle α with respect to the vertical direction, usually neglected, but which increases as the drum climbs along the specimen.

In the classical test setup, a relative displacement u is applied between the bonded part of the specimen and the flanges (points A and B in Fig. 1(a)) and the reaction force F is measured. The load-displacement plot (Fig. 1(b)) displays two constant load portions. At $F = F_w$, the work of the external force is used to lift the drum and to wind the flexible part of the specimen to the constant radius r_1 , while at $F = F_d$ the energy increase is used to create the new delaminated surfaces. If the flexible part of the specimen conforms to the radius r_1 , the energy difference ΔE

is the energy required for delamination. The mode I critical strain energy release rate is then $G_{Ic} = \frac{\Delta E}{\Delta A}$, where $\Delta A = u_d \cdot w$ is the cracked surface, u_d is the length of the delaminated portion and w is the specimen width.

An interesting feature of the CDP setup is that the cracked surface can be determined by geometric considerations. For small values of α , the displacements involved can be parametrized by the angle θ of rotation of the drum:

$$u_d = r_1\theta, \quad u = (r_2 - r_1)\theta \quad (1)$$

thus the critical energy release rate can be calculated as

$$G_{Ic} = \frac{\Delta E}{\Delta A} = \frac{(F_d - F_w)u}{u_d w} = \frac{F_d - F_w}{w} \frac{r_2 - r_1}{r_1} \quad (2)$$

This calculation requires only the measure of the two constant forces F_w and F_d and no monitoring of the crack front, unlike in the standard mode I test, the Double Cantilevered Beam (DCB).

2.2. Crack propagation driven by the oxidation

In the test proposed here, a setup similar to the CDP test is used to provide a constant mechanical contribution to the strain energy release rate of the system, while letting the oxidation drive the crack propagation. To this end, the setup is submitted to a constant force F_{ext} so that $F_w < F_{ext} < F_d$ and placed in an oxidizing environment. The mechanical strain energy release rate, G_{ext} , can be calculated from Eq. (2) by replacing F_d with F_{ext} . Crack propagation occurs once the oxidation has sufficiently fragilized the material, that is once one has, on the crack tip, $G_{Ic,ox} = G_{ext}$. The crack propagation as a function of time is monitored and the propagation velocity is determined for different applied external forces. The oxi-delamination test proposed here is fundamentally different from self-cracking tests.

In self-cracking tests, the oxidation-degradation develops within the plies and it involves a volume of the material, starting from the external surface and increasing its depth with time. Isolated cracks initiate at the fiber-matrix interface and propagate to create an uneven front, which is measured in different locations for different aging times. The propagation of the oxidation-degradation front depends on the balance between the fragilization induced by oxidation, which leads to the decrease of the critical strain energy release rate, and the driving force, constituted by the differential shrinkage. There is no simple way to upset the balance between these mechanisms, since they are both related to the local oxidation level.

In oxi-delamination tests, crack propagation occurs along an interface between two plies. A single, straight crack front is kinematically imposed, thus the local behavior is averaged out across the specimen width. The propagation velocity is measured along the same crack and it depends both on the oxidation level (fragilization and shrinkage) and on the external imposed load, which provides a supplementary mechanical driving force. The possibility to upset the balance between the mechanical and oxidative contributions to crack propagation makes this setup interesting, since the aim is to separate the contributions of the different mechanisms.

2.3. Design of the test setup

A new test setup for the oxi-delamination test was designed and manufactured (see [7] for details), with $r_1 = 50$ mm, $r_2 = 75$ mm and the drum mass $M = 2.1$ Kg.

The tests were carried out within a temperature controlled chamber to accelerate oxidation. Another way to accelerate the test is to increase the partial pressure of oxygen, but unfortunately no experimental means to provide such an acceleration were available. Based on self-cracking tests, the expected crack propagation velocities are of the order of the micron per hour, which implies a number of difficulties for the setup design.

Multiple tests should be carried out in parallel to reduce the time frame needed to measure the long term specimens behavior. Thus, a less bulky CDP setup was conceived and manufactured in three copies, which were simultaneously set within the temperature chamber. Linear actuators allowed to control the displacement during initial pre-delamination, then they were replaced by dead weights, which impose the constant forces during oxidation-controlled propagation.

During oxidation-driven propagation, the key quantity to be monitored is the crack velocity. An accurate displacement measure, allowing to detect extremely small propagation velocities, is the most critical point of the modified setup. In order to reduce the dependence of the displacement measure on the flexibility of the testing machine, we chose to introduce a differential measure in the form of linear displacement transducers positioned outside the temperature chamber and attached, on one side, to the flanges, and, on the other, to the specimen. Thus, the relative displacement u is measured and the delaminated length u_d is obtained from Eqs. (1).

3. Results and discussion

All the results discussed in the following were obtained on IM7/997-2 carbon-epoxy specimens. The test specimen should be constituted of a flexible and a stiff part, thus a stacking sequence including a non symmetric pre-delamination should be used. The specimen in-plane dimensions were 230×20 mm and the chosen stacking sequence is $[0_2 / + 45 / - 45 / 0]_s$, with a 50 mm long pre-delamination situated after the first two 0° plies.

3.1. Displacement controlled tests

The results reported here illustrate the use of CDP test as an alternative to the DCB test to measure the mode I critical strain energy release rate of monolithic composite specimens.

A first series of tests was carried out at room temperature using the normalized CDP setup [5]. The load-displacement plots are given in Fig. 2, where the measured displacement is the displacement of the testing head. The critical strain energy release rate is obtained via Eq. (2) and given in Tab. 1 (left).

A second series of tests was carried out on the modified CDP setup within the temperature controlled chamber, at an imposed temperature of 150° C (Fig. 3 and Tab. 1 (right)). The displacement controlled zone is kept short in order to have a longer testing zone for the oxidation-driven propagation. Microscopic observations of the delaminated surface at the end of the test

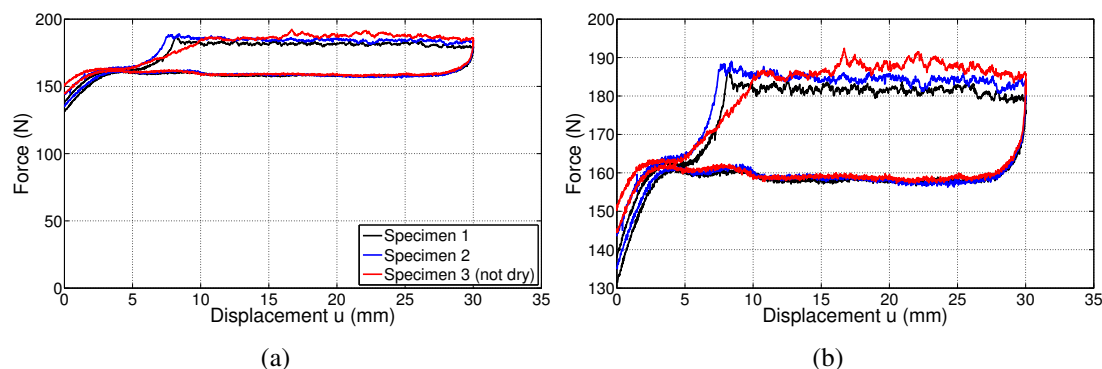


Figure 2. Load-displacement curves for the normalized CDP setup at room temperature.

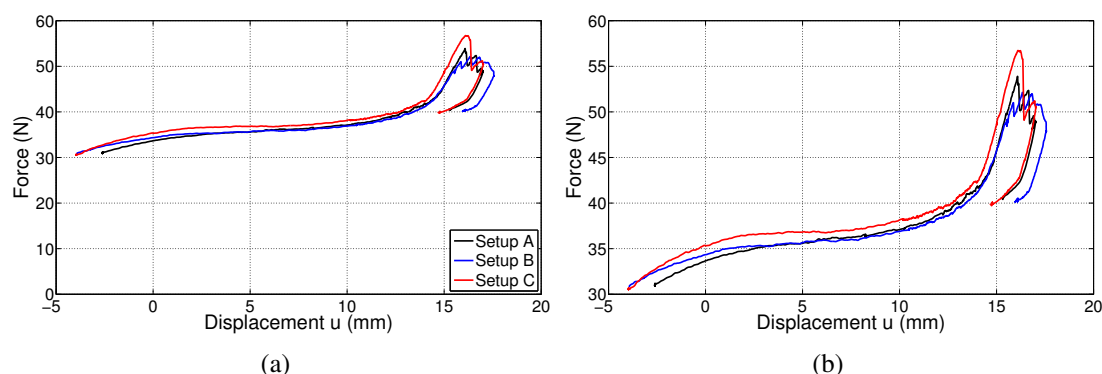


Figure 3. Load-displacement curves for the modified CDP setup at 150° C.

(Fig. 4) revealed a straight delamination front. As in [1], one can distinguish the displacement controlled propagation from the oxidation controlled propagation (discussed in Section 3.2).

In the modified test setup, the ratio between the delamination energy and the total energy is higher than in the normalized setup, due to a lighter drum; thus, the differential measure is more precise. The critical strain energy release rate values obtained for the two series of specimens are similar.

The CDP test is an excellent alternative to the classical DCB test for the determination of the mode I strain energy release rate of monolithic composite specimens. Since the standard test procedure does not require a measurement of the crack length, it is easy to carry out this type of test in a confined chamber, controlling different environmental variables during the test. Furthermore, the differential energy measure used to determine the critical strain energy release rate allows to test different stacking sequences, automatically excluding the dissipation eventually occurring because of damage and/or anelasticity within the plies.

Specimen	1	2	3	average	Specimen	A	B	C	average
G_{Ic} [J/m ²]	302	331	355	329	G_{Ic} [J/m ²]	347	367	372	362

Table 1. Critical strain energy release rates at room temperature (left) and at 150° C (right).

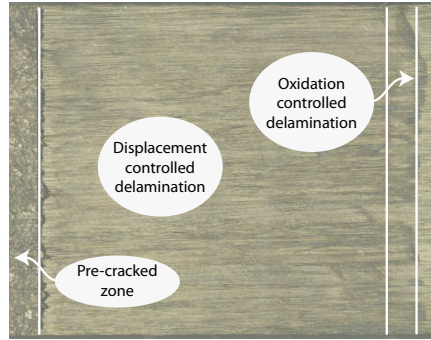


Figure 4. Microscopic observations of the cracked surfaces after the CDP test.

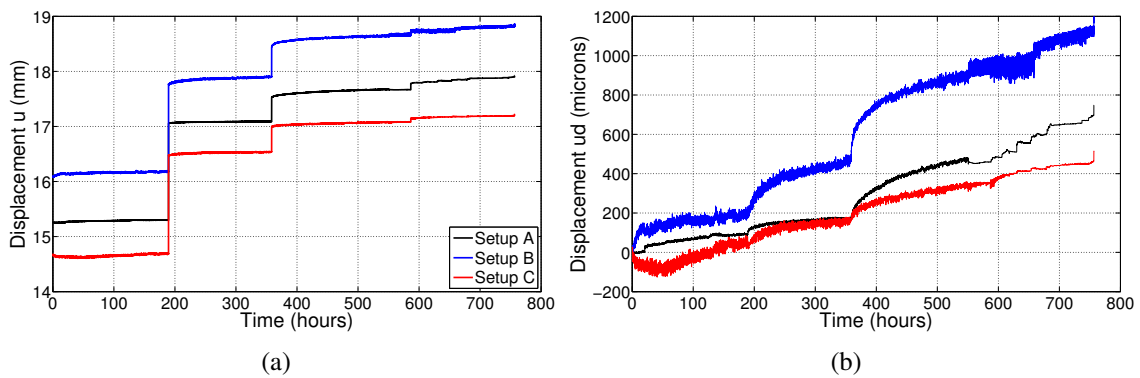


Figure 5. First series of oxi-delamination tests for increasing applied forces (40, 44, 48 and 49 N).

3.2. Load controlled tests under oxidative environment

The results reported here concern the use of the modified CDP test to measure the oxidation-driven propagation of delamination.

A first series of tests was carried out within the temperature chamber at 150° C. Different load levels were imposed via dead weights and the displacement u was measured as described in Section 2.3. Each load level was kept constant for about one week before increasing it.

The displacement-time curves are given in Fig. 5. The raw measures of u are given in Fig. 5(a): the displacement gaps correspond to the load increases and include adjustment of the setup (elastic deformation and variation of the angle α), as well as ‘accidents’ due to the manual positioning of the dead weights. In Fig. 5(b) the measured displacements u are converted to u_d via Eq. (1) and joined by eliminating the gaps corresponding to the load increases.

The displacements measured during the constant load portions are very small with respect to the overall displacements involved, which makes it difficult to obtain accurate crack propagation velocities. Furthermore, a significant noise is observed in Fig. 5(b). The oscillations in the displacement measures are in phase with the temperature oscillations within the chamber ($\pm 1^\circ$ C) thus, if the average temperature chamber is kept constant, the average values should not be affected.

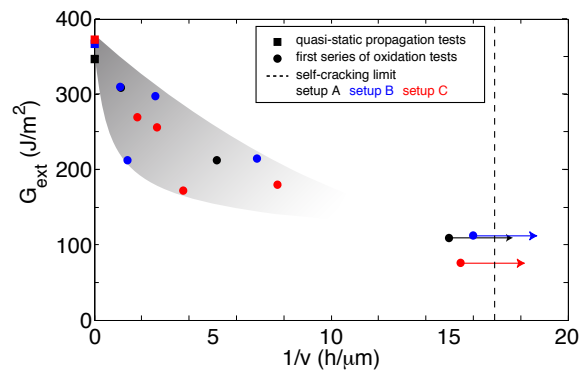


Figure 6. Imposed strain energy release rate versus inverse of the stable velocity.

A transient phase is observed in the initial few hours after the load increase. Then, the velocity appears to become stable. These stable velocities were calculated and their inverse was plotted against the imposed strain energy release rate in Fig. 6, together with the values obtained in the displacement controlled tests (infinite velocity) and with the self-cracking limit (estimated from [2]). Notwithstanding a significant dispersion, the velocities appear to decrease with decreasing imposed strain energy release rate, until no appreciable propagation can be observed within the observation time frame. The inverse of the velocity can be related to an oxidation level via a diffusion/reaction model [7]: as the available energy release rate G_{ext} decreases, more oxidation induced degradation is necessary to allow crack propagation.

The transient phase is harder to interpret. Two possibilities were considered: on one hand, a perturbation of the measurement associated to global viscosity and friction; on the other hand, the presence of local loading history effects enhanced by the high temperature. In order to evaluate the effect of the global phenomena on the measurements, a second series of tests was carried out, blocking the delamination in one of the setups by fixing the whole specimen (and not only the delaminating part) to the drum. Although a non negligible displacement was observed for the blocked specimen, global viscosity and friction do not seem sufficient to explain the transient behavior of the delaminating specimens.

A third series of tests, in which the dead load was decreased at each step, was carried out in order to study the loading history effect. Results for the same load level during the second and third series of tests (increasing and decreasing load steps) are given in Fig. 7. The measured responses are different: in the case of decreasing load steps, crack propagation does not appear to have started again after 300 hours. A possible interpretation is that, at high temperatures, local viscous effects at the crack tip may come into play in addition to the oxidation and brittle crack propagation mechanisms. Once again, the CDP test will be a useful tool to test this hypothesis, since it allows to modify independently the different loading parameters (mechanical load, temperature and partial pressure of oxygen).

4. Conclusions

A new test setup, based on the CDP test, was presented to study the oxidation/cracking coupling in polymer matrix composites. The CDP test was shown to be a promising tool for the char-

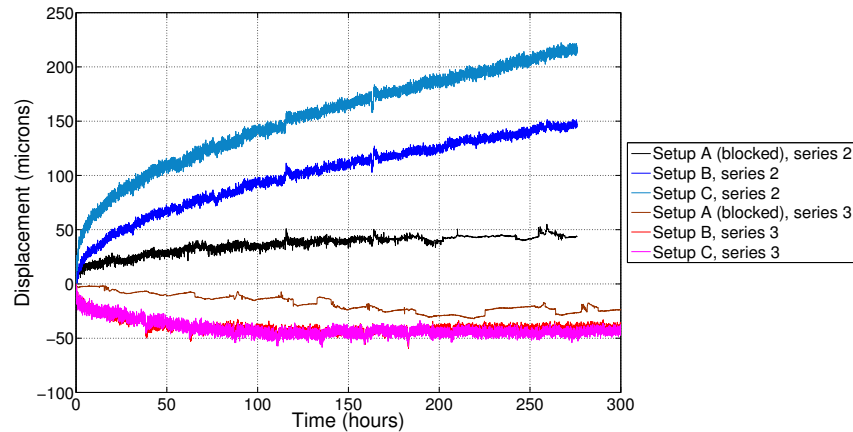


Figure 7. Displacement-time plots for increasing to 44 N (series 2) and decreasing to 44 N (series 3) load steps.

acterization of the mode I critical strain energy release rate of monolithic composites. Some preliminary insight could be gained on the coupling between oxidation and crack propagation under different imposed mechanical loads.

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