

Multifunctional Thin Film Lithium Ion – Graphite Composite Structural Laminates

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A dissertation
submitted in partial fulfillment of the
requirements for the degree of

Doctor of Philosophy

University of Washington

2013

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Program Authorized to Offer Degree:

Aeronautics and Astronautics

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Abstract

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The consolidation of batteries and airframe into a multifunctional structure can theoretically reduce the aircraft weight by exploiting the battery components as load bearing elements and by enabling a distributed power supply. To realize this vision, a composite structure for load bearing and electrical energy storage comprised of thin film Li-ion batteries and carbon fiber/epoxy laminae was proposed. In this framework, the experimental characterization and finite element analysis of delamination growth and delamination buckling collapse response of the highly inhomogeneous laminates was conducted to support the design of the next generation of airborne multifunctional structures for load bearing and electric energy storage. The study assessed the technology readiness and showed that the low fracture toughness and the inhomogeneity of the material properties of a baseline battery packaging design led to critical unstable delaminations. Moreover, it demonstrated that the packaging design is a determining factor for the integrity of the multifunctional laminates. The study provided a design space for improving the limits of utilization of the next generation of structural thin film batteries by optimum materials selection.

Based on these results, the use of thin film lithium ion technology seems to be possible for low stress applications and secondary structures with short lifecycle. This research also provided a general and novel automatic incremental solution algorithm for nonlinear static finite element equations, designed to compute local/global structural collapse by stable and unstable delamination propagation.

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1. Manufacturability of composite laminates with integrated thin film Li-ion batteries

1.1 Summary

The limits of processability of solid state thin film lithium-ion batteries embedded into composite laminates were identified through testing under pressure, temperature and a liquid resin environment representative of composite processing cycles. Battery failures were characterized based on optical microscopy and capacity retention, and three distinct types of failures were recognized and analyzed. Failures were associated either to the battery polymeric sealant failure, or to the physiochemical degradation of the electrolyte or the anode. Results gave evidence that the cure temperature was the most influential parameter for battery survivability. Based on these experimental results, the optimum curing cycle was identified and embedding tests that retained full battery capacity were successfully performed. The minimal three-layer battery packaging proved to be an efficient air and moisture barrier within the above conditions.

1.2 Introduction

The flight endurance of an electric propeller-driven unmanned aerial vehicle (UAV) is significantly improved by reducing the weight more so than increasing the battery capacity [1]. Moreover the structure and the battery each typically contribute 20-40% to the total UAV mass [1, 2]. The consolidation of battery and structure can theoretically reduce the total weight by exploiting the battery components as load bearing elements and by eliminating battery fittings or supports. In addition, rather than bulky, centralized batteries, the integration of multiple lightweight batteries into the structure enables distributed power supply and storage, potentially reducing the amount of wiring.

However, to date, system performance improvements achieved with such integration were documented only for applications demanding low mechanical stress [3]. Furthermore the current battery technology, based on micro-structured lithium intercalation compounds, is such that high specific energy and specific power, as well as good mechanical properties cannot be achieved [3-8]. In order to meet the requirements

for the next generation of airborne, load bearing batteries, solid state thin film Li-ion batteries (TFB) were recently proposed [9-13]. The thin film structure maximizes the specific contact surface between electrodes and electrolyte, thereby increasing the electric stored energy and power per unit mass by increasing the fraction of reactants and the rate of the electrochemical reaction respectively. Downsizing the electrodes and electrolyte to a thin film dramatically reduces the path length for ionic and electronic transport, allowing to achieve the desired conductivity without utilizing conventional porous compounds comprised of electrically conductive lattices and binders, leading to higher energy and power density. Moreover scalable manufacturing processes for nanostructured materials, such as electrospinning, chemical vapor deposition (CVD), atomic layer deposition (ALD), are suitable for TFB manufacturing. The future application of nano-scale technology to the battery active components can potentially lead to several advantages. First, it allows further increase of TFB capacity and power by decreasing the path length of Li-ion and electronic transportation [14-16]. It can also enable the simultaneous implementation of electric energy storage and load bearing capabilities through engineered composite electrodes comprised of electrochemically active particles or fibers bound by a structural matrix with electrically conductive filler. Last, nano-scale technology allows better accommodation of the cycling strain induced by Li-ion insertion and removal. Currently this cyclic strain causes low durability and capacity fading during operation of conventional bulk material electrodes, in particular when silicon is used as anode material for high specific capacity [17-19].

The low TFB thickness, typically less than 300 μm , facilitates integration within thin sections, but it implies that a significant weight fraction is constituted by the packaging layers. In the proposed configuration the battery packaging consisted of a substrate, which provided mechanical support to the battery manufacturing process, and a sealant, which ensured electrical insulation and sealing of the highly reactive cell components, Figure 1-2. The high weight fraction of passive components, such as the packaging, is a deterrent for most large-scale applications of TFBs, which are instead typically employed as micro power sources for memory chips, microelectromechanical systems (MEMS) and medical implantable devices. However, if the packaging is by design an active load bearing element instead of

passive battery mass, this technology becomes an appealing multifunctional system for electric propelled air vehicles. Low-power airborne applications, such as localized power sources for structural health monitoring systems or for fly-by-feel distributed sensing materials, would also benefit from TFBs integrated in the airframe, without requiring a load bearing battery design.

The manufacturability and functionality of TFBs integrated within structural composite laminates have to be proven in terms of physiochemical compatibility of TFB materials to the composite curing environment, durability of the electrodes, laminate structural integrity and TFB packaging integrity under applied loads. Mechanical tests conducted on a commercially available TFB by [11] showed that the battery was capable to withstand uniaxial transverse (out-of-plane) pressure up to 830 kPa without any detrimental effects on the electric functionality, thereby indicating that the TFB was compatible with the autoclave curing pressure of epoxy-based composite materials. The same TFB type was successfully embedded in a carbon/epoxy composite laminate cured at 121°C. TFB failure was documented for a 177°C cure [12], giving evidence that the processing temperature is an influential parameter for battery survivability. The ability to tolerate exposure to curing temperatures, which range from 121°C to 177°C, in a quiescent status (e.g. with no electric current flow) is neither fully covered by scientific literature, nor by commercial electronics standards. On the other hand, the ability to operate from -55°C to 100°C was extensively investigated because of the relevant commercial applications. High temperature characterization of TFB with the same chemistry of the batteries tested in the present study was performed under charge/discharge cycling (e.g. not in a quiescent status) and permanent capacity reduction was detected at 80°C [20]. In [21], cycling at 100°C and 150°C caused permanent capacity reduction associated to a decrease in grain dimension of the LiCoO_2 cathode, whereas the processing in a quiescent status did not lead to battery capacity loss. The effect of the state of charge, however, was not investigated. The increasing detrimental effect of temperature exposure at an increasing state of charge [22] up to thermal runaway [23] was studied only for conventional 18650 Li-ion cells, not for TFBs.

The objective of this study was to experimentally assess the limits of processability and identify the failure types of the current solid state thin film battery materials within composite curing temperatures,

pressures and a liquid resin environment, in order to enable the manufacturing of composite laminates with embedded or externally bonded TFBs. The research covered the 121°C to 199°C processing temperature range of batteries in a quiescent status at two states of charge, corresponding to the higher and lower limit of the operating charge level.

1.3 Methodology

1.3.1 Solid state, thin film Li-ion battery

The TFBs utilized in this research were manufactured by FrontEdge Technology under license from the Oak Ridge National Laboratory (ORNL), Figure 1. The same batteries were utilized by [10-13]. The cathode material was lithium cobalt oxide (LiCoO_2), the anode was lithium metal (Li) and the solid state ceramic electrolyte was lithium phosphorus oxynitride ($\text{Li}_{2.9}\text{PO}_{3.3}\text{N}_{0.46}$), also known as LiPON [24]. This battery chemistry was electrically characterized in [25]. This electrochemical cell generates an electric current outside the cell and a Li-ion flow inside the cell through the simultaneous oxidation reaction of the lithium metal contained in the anode and the reduction reaction of the lithium ions that occurs at the cathode. This process is called battery discharge. The chemical reactions and corresponding electronic and ionic flow are reversed if an opposed external electric potential is applied to the cell. This process is called battery charge. During discharging, lithium ions are extracted by the anode and intercalated at the cathode. Upon charging, lithium ions are released by the cathode and plated at the anode.

Unlike conventional batteries, that require a lithium intercalation compound as anode material to achieve the required electronic and ionic conductivity, the improved electrochemical kinetics of the thin film structure allows to utilize a metallic lithium anode, resulting in higher specific capacity and lower redox potential. These characteristics allow to minimize the anode thickness and to maximize the cell voltage respectively. LiCoO_2 has the highest electronic conductivity and one of the highest specific capacities among the cathode materials that are technology ready. For these reasons it is one of the most popular cathode materials for conventional batteries and the most popular cathode material among commercially available TFBs. LiPON is currently the solid state electrolyte with the highest ionic conductivity that can

be manufactured into thin films. The chemistry of this cell is representative of the commercial state-of-the-art of TFBs and the electrochemical characterization reported hereinafter enabled its use in multifunctional composite structures and can be regarded as a baseline performance for the next generation of nanostructured TFB materials that are not technology ready yet.

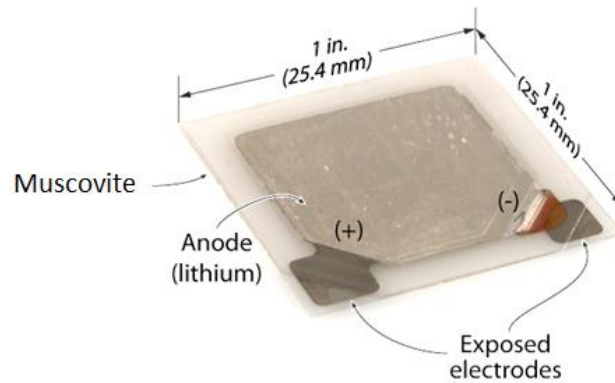


Figure 1. Perspective photo of all-solid state thin film Li-ion battery (TFB) with dimensions. Manufactured by FrontEdge Technology Inc.

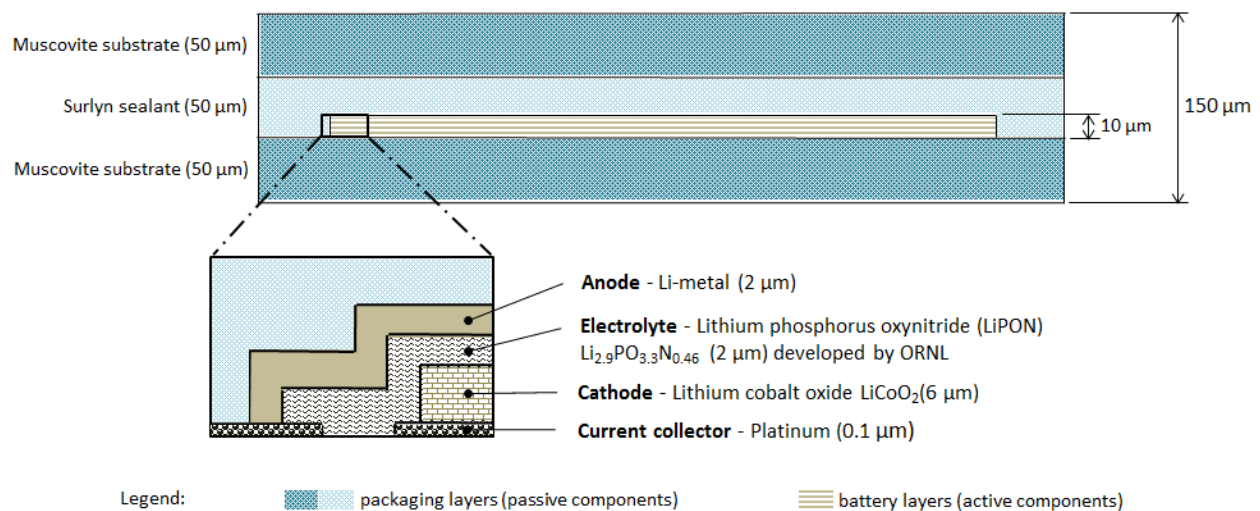


Figure 2. TFB Cross-sectional schematic

Component	Material	Temperature	Property
Sealant	Surlyn	98°C	Melting point
Anode	Lithium	181°C	Melting point
Electrolyte	$\text{Li}_{2.9}\text{PO}_{3.3}\text{N}_{0.46}$ (LiPON)	300°C	Maximum operating temperature ¹⁾
Cathode	LiCO_2	700°C	Annealing temperature ²⁾
Substrate	Muscovite	700°C	Calcination temperature
Current collector	Platinum	1768°C	Melting point

1) Yu X. et al. *J. Electrochem. Soc.* 147(2), 517 (2000)

2) Bates J. B. et al. *J. Electrochem. Soc.* 147, 59 (2000)

Table I. Relevant temperature thresholds for TFB components

The active components were encased by two muscovite substrates bound by a thermoplastic layer of Surlyn sealant, leading to a total thickness of 150 μm , Figure 2. Critical temperature thresholds for the TFB materials are summarized in Table I.

The electrochemically active components of the battery were grown by physical vapor deposition (PVD) performed in-situ on the muscovite substrate. The cathode was fabricated through RF magnetron sputtering and annealing at a high temperature (typically 700°C) to obtain a crystalline microstructure with large grains and uniform preferred crystalline orientation to maximize ionic conductivity [26]. The muscovite substrate ensured dimensional stability during the annealing process, preventing cathode cracking or dis-bonding due to thermal stresses. The patented process was described in [27]. The electrolyte was deposited by RF magnetron sputtering in N_2 atmosphere [28] according to the ORNL process [24]. The Li anode was deposited by thermal evaporation [25]. These materials are highly reactive. Li-metal reacts with N_2 [24] and O_2 [25], while all of the three active component materials react with H_2O [29]. Therefore the TFB had to be hermetically sealed. A permanent failure of the sealant during manufacturing or operation would lead to the failure of the battery by compromising the ability of

the electrochemical cell to store energy. The electrical capacity loss that would occur after the failure of the sealant could be almost instantaneous or the capacity could fade progressively, depending on the flow rate of contaminants entering the battery.

The battery was a 25.4 mm square with a nominal voltage of 4.2V, and a nominal capacity of 1 mAh. In order to calculate the actual specific energy, the energy delivered by one battery during a full discharge was measured according to the method described in the next section. The active components of the battery were separated from the substrate and sealant, and weighed with a Mettler Toledo XS64 analytical balance. The active mass, given by the sum of cathode, electrolyte, anode and current collectors, was 0.0113 g, which led to a specific energy of 353 Wh kg⁻¹. The specific energy calculated with respect to the total TFB mass was 22 Wh kg⁻¹.

1.3.2 Experimental approach

Battery survivability was monitored through discharge and charge cycling prior to and following a thermal processing test. The same survivability check was repeated two months following the treatment to assess the survivability after aging. An automated circuit board featuring a charge-discharge electronic circuit, Figure 3, was connected to a National Instruments BNC-2120 connector and controlled via a LabView program. Current and voltage readings were collected every three seconds. The discharging occurred under a constant 3.8 kΩ resistive load, which led to an average discharge current of approximately 1mA, Figure 4, corresponding to a discharge rate of 1C. The TFB was considered fully discharged when the voltage reaches 3 V. Immediately following a discharge, charging was performed at a constant voltage of 4.2 V, applied by a Hewlett-Packard 6632A System DC Power Supply. A shunt resistance of 10 Ω was utilized to measure the current. The battery was considered fully charged when the current dropped below 50 μA. For each battery, five charge-discharge cycles were performed; the first was used to condition the battery and the following four provided the average discharge capacity. In Figure 4 the current peak denotes the beginning of a charge cycle. The capacity was obtained by numerical integration of the discharge current, as a function of time, over a discharge cycle.

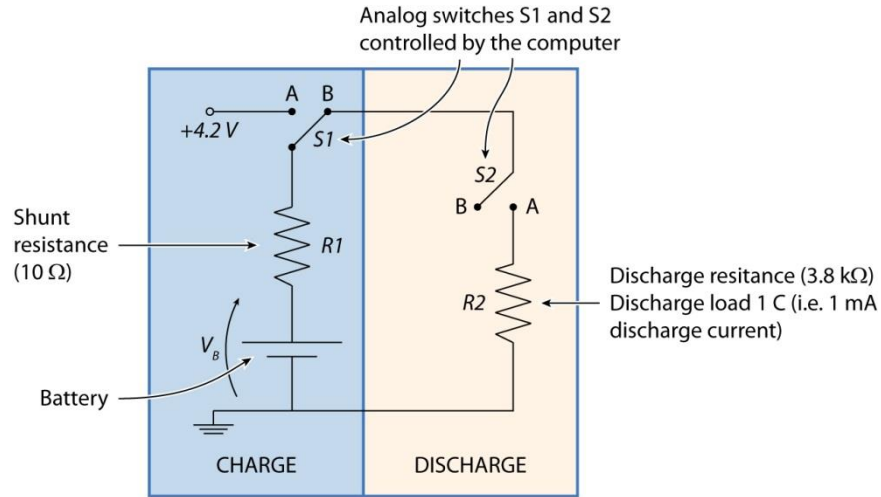


Figure 3. Schematic of the circuit used to test the battery's survivability by monitoring charge and discharge performance before and after thermal processing

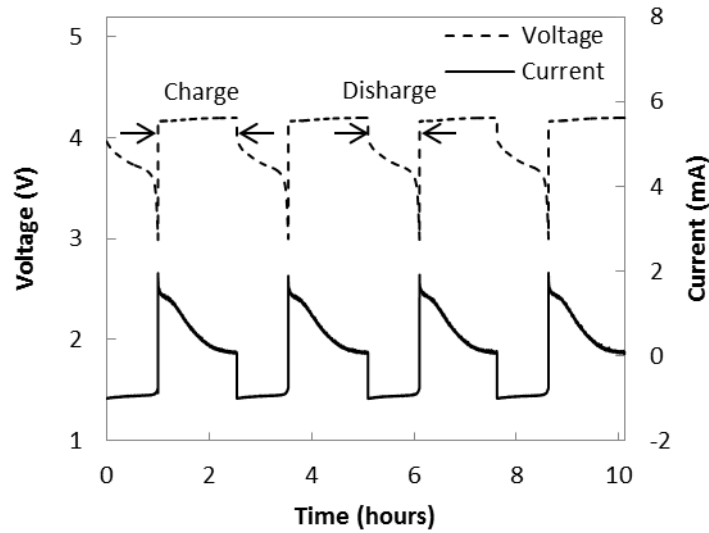


Figure 4. Description of the four discharge and charge cycles for the survivability test

In order to assess the limits of process-ability, the TFBs were subjected to pressure, temperature and liquid resin environment that were representative of composite curing cycles. The batteries were exposed to a one hour isothermal hold at a given temperature, 121°C, 149°C, 177°C or 199°C. For each

temperature, one battery was tested at ambient pressure, a second was placed under a 711 mm Hg vacuum and a third was embedded in a 50.8×50.8×3.8 mm pool of Fiberlay Pro Glas 1300 series neat epoxy resin under a 711 mm Hg vacuum. The vacuum was created within a flexible nylon bag in order to apply a hydrostatic pressure to the resin, Figure 5.

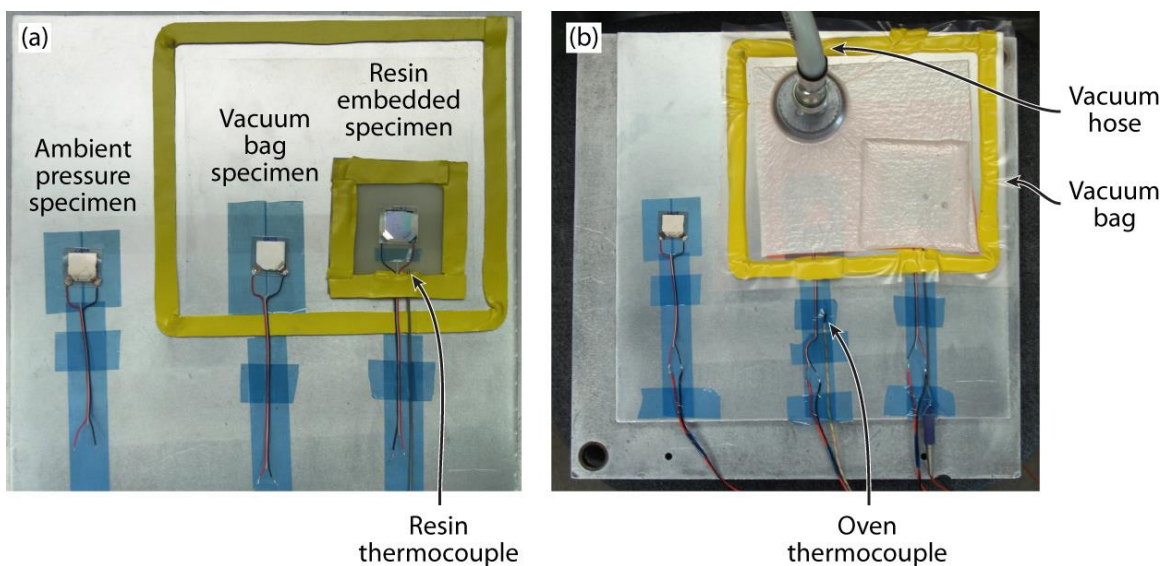


Figure 5. Simultaneous thermal treatment of batteries under ambient pressure, in a vacuum bag and embedded in a neat resin pool. (a) Location of batteries before addition of (b) the vacuum bag

A sample of batteries was thermally processed at 121°C in a fully charged state and in a partially charged state to determine if the state of charge affected battery survivability. In order to avoid over-discharging the batteries, as recommended by FrontEdge, the partial state of charge was achieved by discharging to 3.9 V_{OC} (open circuit voltage) prior to thermal processing. Based on these preliminary results, which gave evidence of the detrimental effect of the full charge status on the TFB capacity retention, only partially charged TFBs were tested at higher temperatures.

Temperatures were recorded by a LabView program, which monitored the oven temperature and the resin temperature from two independent thermocouples via a National Instrument USB-6210 multifunctional data acquisition (DAQ) module. A Despatch LAC bench-top ventilation oven heated the specimens at a rate of 5°C min⁻¹ from room temperature to the test temperature. The test was conducted under an

electrical load of 108 k Ω (R3) to facilitate battery failure detection. The current drained from the batteries was small enough to consider the state of charge constant throughout the test. Voltages and currents were recorded by the DAQ and LabView program with a frequency of one measurement every 3 seconds, Figure 6.

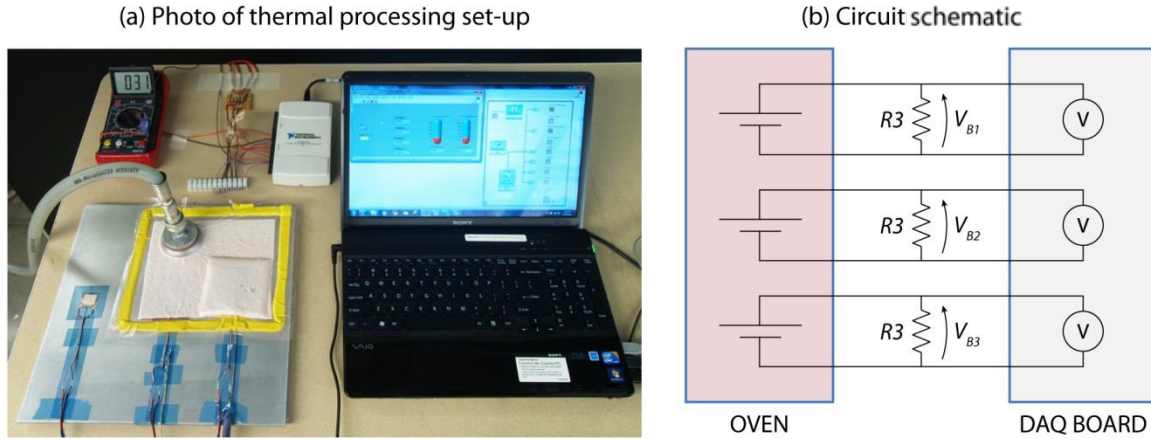


Figure 6. Battery and thermocouple voltages are recorded by a LabView program via the DAQ board during thermal testing under electrical load. (a) Set-up before the specimen tray is placed in the oven. (b) Circuit schematic including TFB inside the oven and resistive load (R3) outside the oven

A total of 18 batteries were subjected to the thermal processing test. Based on the results, a cure cycle was designed and validated through embedding tests in glass fiber/epoxy (GFRP) and carbon fiber/epoxy (CFRP) composite laminates. The TFB was embedded at the midplane of the laminate, Figure 7. Two slits in the top layers of the laminates allowed for the Flat Flexible Cables (FFC), Nicomatic 254PW01E6095 polyester coated single copper conductor, to exit the laminate. FCCs are 0.25 mm thick and 5.12 mm wide, and they were rated for operation up to 150°C for short durations and to 100°C for continuous use. They were connected to the TFB electrodes using MG Chemicals silver conductive epoxy 8331-14G. The glass/epoxy composite material utilized was Toray AGATE prepreg glass fiber FGF7781/2510, 8 harness satin weave fabric. The 4-ply laminate was cured at 132°C and 520 kPa for two hours through heated press molding. The laminate stacking sequence was [0,90]₄. The battery was located at the panel mid-span, Figure 8. A second embedding test was performed using Toray AGATE prepreg

carbon fiber tape T700/2510 with stacking sequence $[0/90]_{2s}$, Figure 9. The same fabrication process was adopted with the addition of a localized application of silicone conformal coating MG Chemicals 422-55 on the exposed battery leads to insulate the connections. Attempts of embedding the TFB without insulated connections failed due to electrical shorts cause by the CFRP.

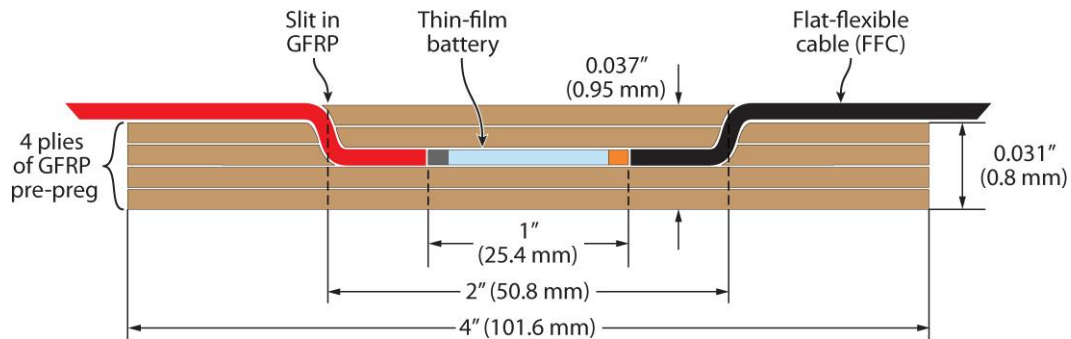


Figure 7. Cross-sectional schematic of a thin-film battery embedded at the mid-plane of a four ply GFRP laminate. Two slits in the top two layers allow for the FFC to exit the panel

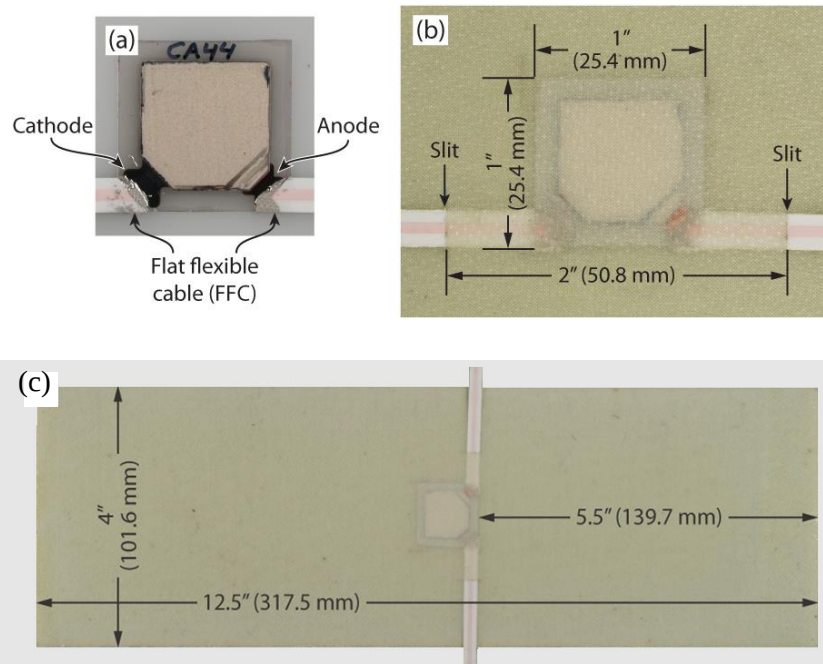


Figure 8. Photos of a battery (a) before and (b) after embedding within a GFRP laminate. (c) Panel with the embedded battery

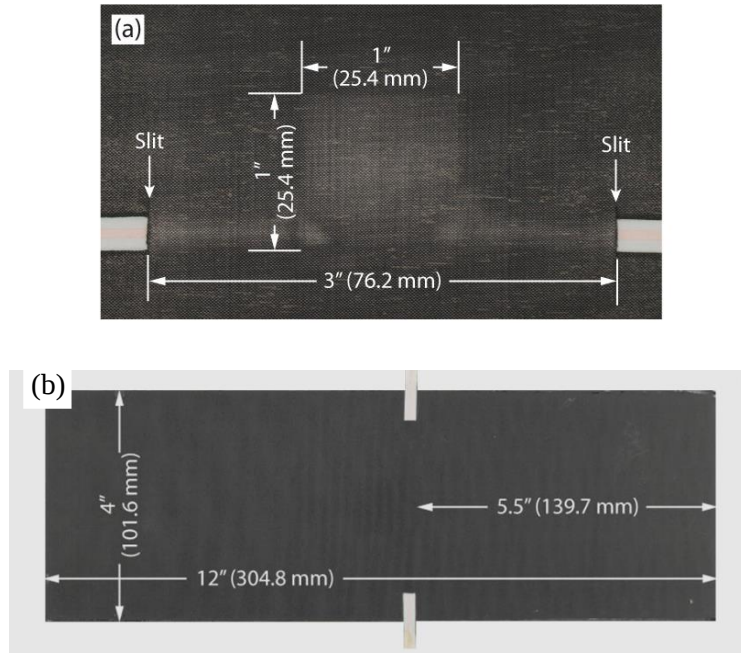


Figure 9. Photos of a CFRP panel with embedded TFB showing (a) a close-up of the panel at battery location and (b) the whole panel.

1.4 Results

Typical temperature and voltage profiles for the thermal processing test are shown in Figure 10. The battery voltage, regardless of the processing environment, decreased with increasing ambient temperature and recovered its initial value when the battery was brought back to room temperature at the end of the one hour long isothermal phase. During temperature ramp-up the resin temperature showed a spike due to the exothermic reaction associated with the resin crosslinking. The maximum difference between resin and oven temperature during crosslinking never exceeded 15°C and the peak temperature never exceeded the isothermal hold temperature. The voltage of the battery embedded in neat resin showed a negative spike which corresponded to the beginning of the exothermic event. This voltage drop, which lasted 3 to 9 seconds, was observed for all of the TFBs processed in the neat resin without having any detrimental effect on the survivability of the battery. While the reason for the voltage drop was unknown, the fact that it occurred during embedment within Fiberlay Pro Glas 1300 neat epoxy resin, but it did not occur during the embedment in the Toray 2510 and Cytec 977-3 prepreg resin systems, suggested that possible causes

should be dependent to the resin chemical composition or to the amount of resin undertaking exothermic reaction.

Thermal testing at 121°C of fully charged batteries led to an average capacity reduction of 9%, as summarized in Table II. The capacity reduction was proportional to a reduction in discharge time, whereas the cell voltage and power were unchanged, Figure 11. On the other hand, if the TFBs were partially discharged to 3.9 V before being tested, they withstood thermal processing up to 149°C without any detrimental effects on their electric performance. The detailed summary of the results, Tables III and IV, showed that the short term and long term capacity after thermal processing remained within $\pm 3\%$ of the baseline capacity measured before the test. The capacity variation was associated with a slight offset of the discharge current and voltage profiles, Figure 12. Neither the liquid resin nor the vacuum bag environment seemed to have an effect at this processing temperature. All the batteries tested at 177°C, either at ambient pressure or in the vacuum bag, were affected by partial or total capacity loss, Table III and IV, associated with a decreased discharge time and a negative translation of the current and voltage discharge profile, Figures 13 and 14. The batteries embedded in neat resin at 177°C, as well as all the ones processed at 199°C, failed during testing.

	Fully Charged		Partially Charged	
	Capacity	Survival	Capacity	Survival
	Retention	Rate	Retention	Rate
Ambient	94%	1/1	102%	1/1
Vacuum Bag	95%	1/1	102%	1/1
Neat Resin	84%	1/1	102%	1/1

Table II. Capacity retention of batteries undergoing a 1 hour isothermal hold at 121°C in either a fully charged state or partially charged to 3.9 V_{OC} (open circuit voltage)

	Isothermal Temperature			
	121°C	149°C	177°C	199°C
Ambient	1/1	1/1	1/2	0/1
Vacuum bag	1/1	1/1	(2)/2	0/1
Neat Resin	1/1	1/1	0/2	0/1

Table III. Survival rate of partially charged batteries subjected to temperatures, pressures and liquid resin environment typical of composite manufacturing. Parentheses indicate failure after two months of aging

	Isothermal Temperature			
	121°C	149°C	177°C	199°C
Ambient	102% (100%)	100% (100%)	76% (70%)	0%
Vacuum bag	102% (100%)	103% (100%)	87% (0%)	0%
Neat Resin	102% (100%)	98% (98%)	0%	0%

Table IV. Average discharge capacity retention of survived batteries from Table 3. Values in parenthesis measured two months after test

Three distinct types of failures were recognized and characterized based on optical microscopy at the anode side and capacity retention, Figure 15(a-d). Type I failure, Figure 15(b,bb), was observed as a localized grey spot on the Li-anode with a complete loss of grain boundaries. FrontEdge Technology attributed these observations to electronic failures consisting of a local breakdown of the electrolyte. The grey spot was always associated with bubbling of the overlaying Surlyn sealant layer at the failure location. This failure occurred in batteries processed at ambient pressure or within the vacuum bag. The affected batteries were operational, but with reduced capacity. Type II failure produced a neutral grey discoloration, which extended from the edges of the active components, Figure 15(c,cc). The micrographs revealed dark patches scattered across the anode. It is believed that this failure was caused by the reaction

of the Li-anode with contaminants diffusing through the Surlyn sealant and entering into the battery. The appearance of Type II failure always led to total capacity loss. This failure was observed after thermal processing of batteries embedded in neat resin, or in aged batteries previously processed at 177°C.

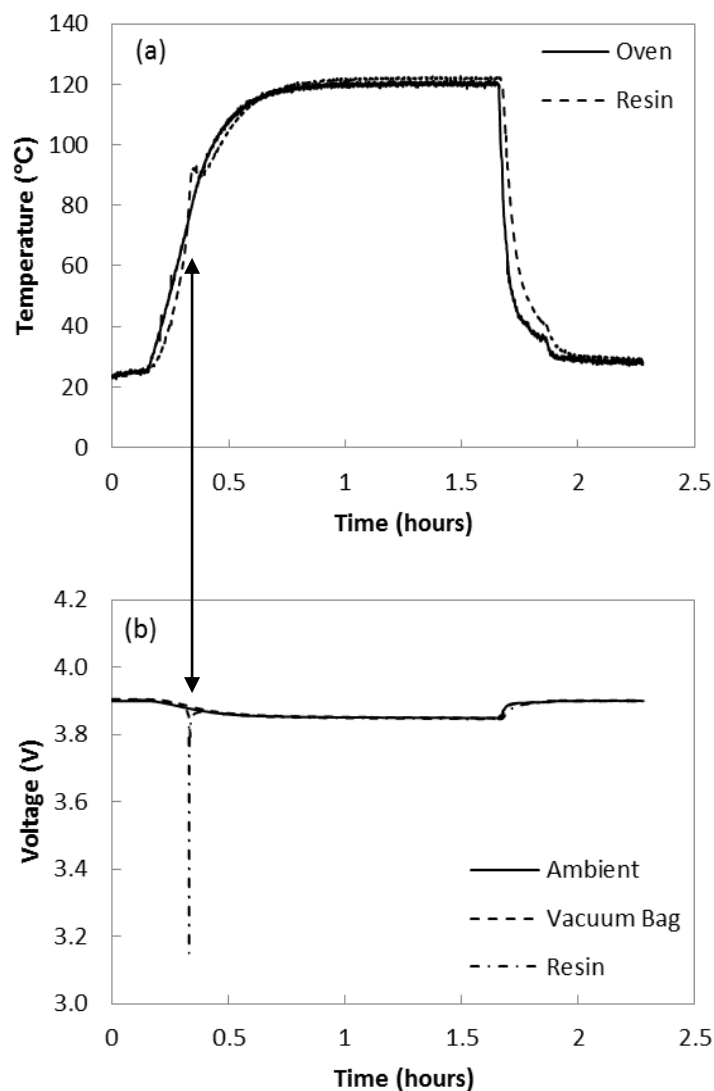


Figure 10. (a) Typical temperature cycle for TFB thermal processing test. The temperature profile for the oven and neat resin is shown for a 1 hour isotherm at 121°C. (b) Voltages of the three batteries shown in Figure 5 undertaking the thermal treatment. A voltage drop of the battery embedded within the resin coincides with heat generation by the resin due to exothermal crosslinking reaction

The Type III failure shown in Figure 15(d,dd) occurred for all of the batteries tested at 199°C. The failure occurred above 177°C during the temperature ramp-up, leading to a sudden loss of voltage and battery failure. This failure was due to lithium melting (M.P. 181°C) and the TFB became black at the anode side, occasionally showing gray spots which were probably Type I failures formed prior to the melting of the anode.

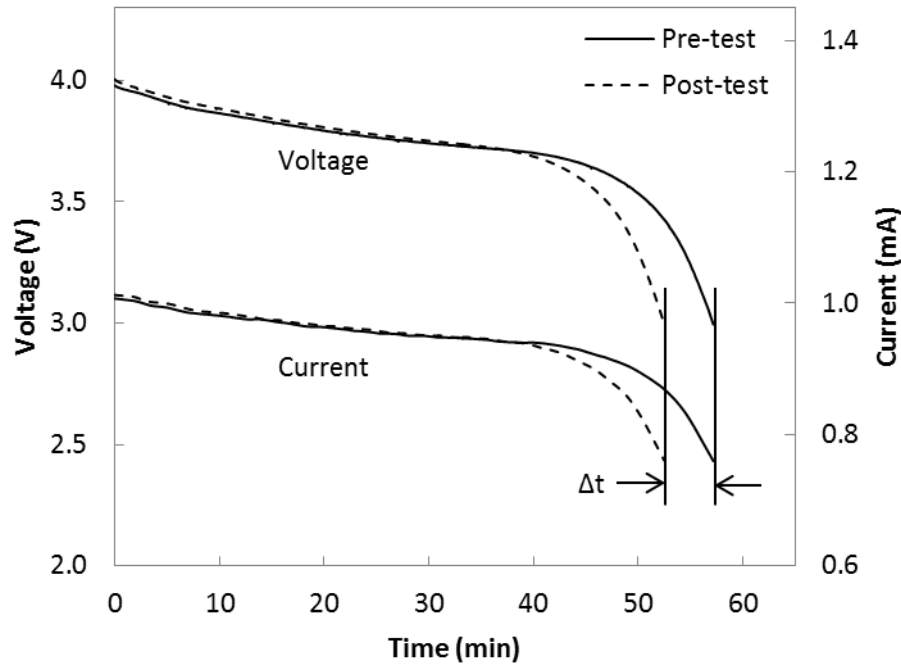


Figure 11. A discharge profile from the survivability test for a fully charged battery subjected to a 1 hour isotherm at 121°C under ambient pressure. The decrease in the discharging time is proportional to the 6% decrease in capacity retention after processing (see Table II)

	Capacity	Survival
	Retention	Rate
CFRP	103%	1/1
GFRP	102%	1/1

Table V. Survival of partially discharged batteries embedded in either a GFRP or CFRP laminate cured at 132°C and 520 kPa

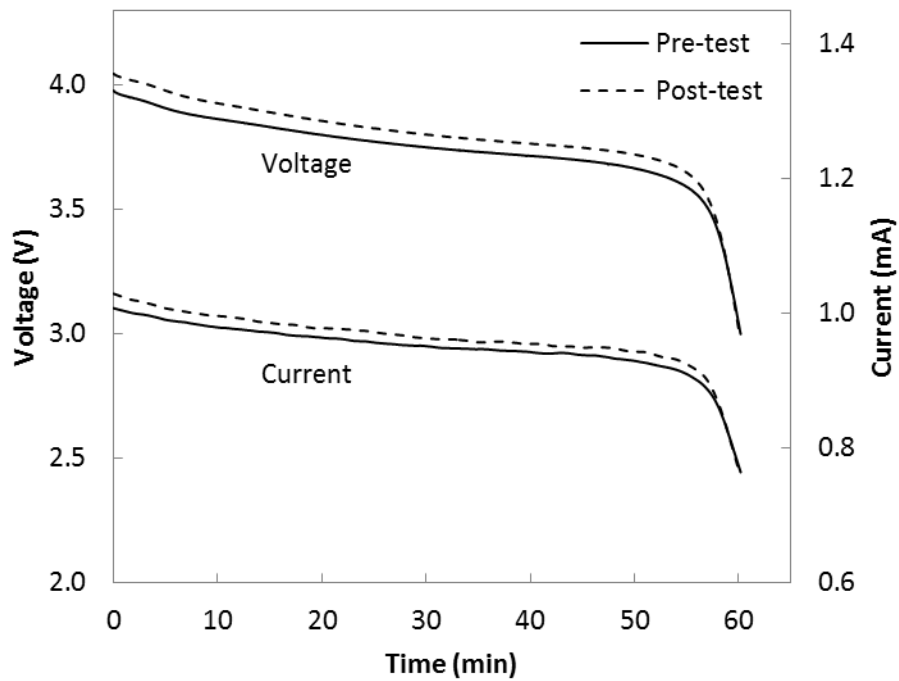


Figure 12. A discharge profile from the survivability test for a partially charged battery subjected to a 1 hour isotherm at 121°C under ambient pressure. The battery had 102% capacity retention after processing (see Table II)

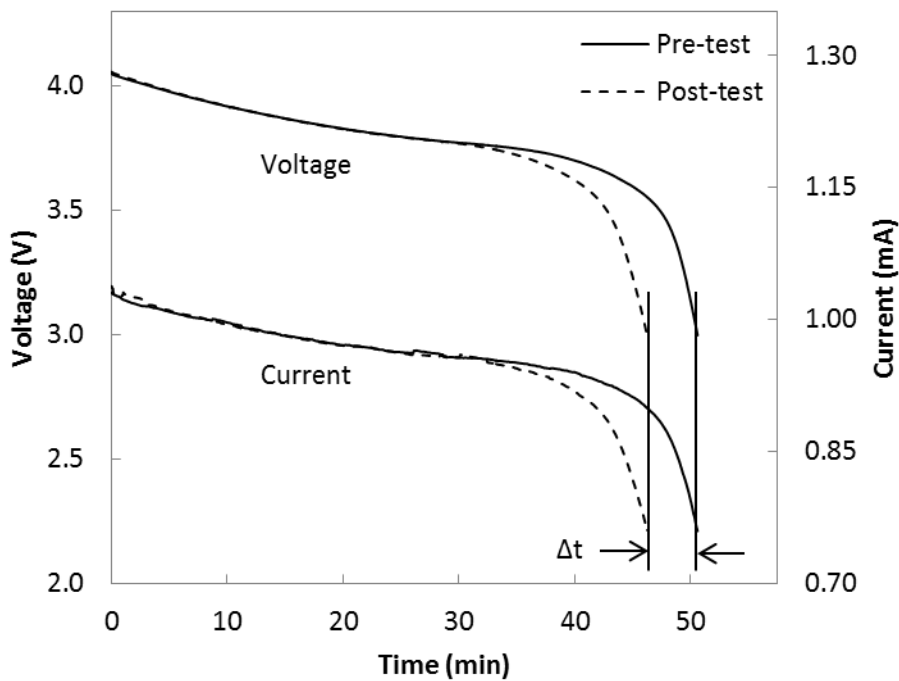


Figure 13. A discharge profile for a partially charged battery subjected to a 1 hour isotherm at 177°C under vacuum. The battery had 91% capacity retention after processing, ultimately failing with aging (see Table III)

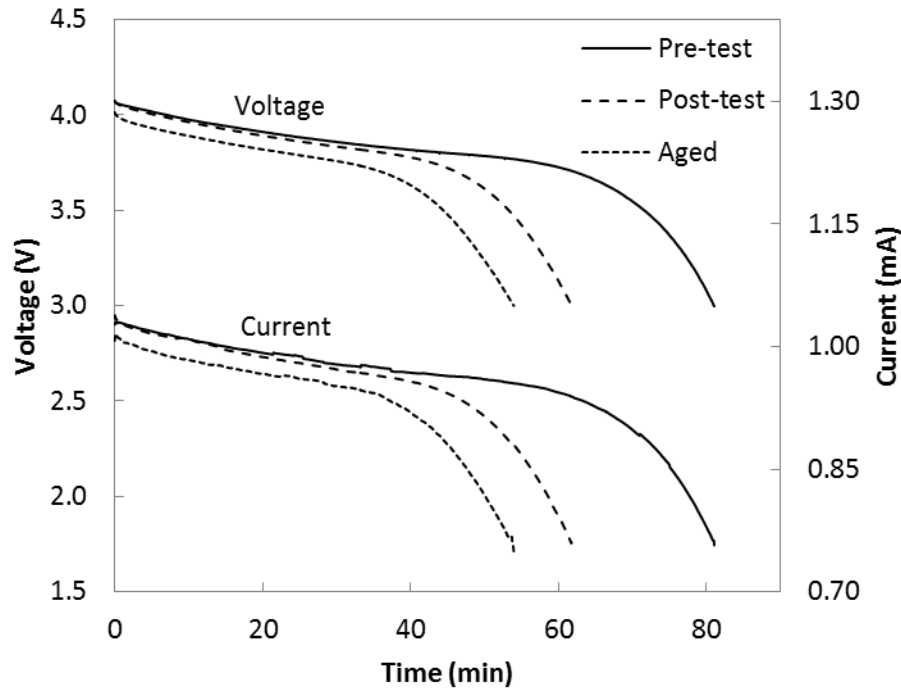


Figure 14. A discharge profile from the survivability test for a partially charged battery subjected to a 1 hour isotherm at 177°C under ambient pressure. The battery had 76% capacity retention after processing, reducing to 70% with two months of aging

The battery shown in Figure 16 was affected by Type I failure and the associated capacity reduction caused by processing at 177°C in the vacuum bag. With aging, however, this battery completely failed by Type II failure. This indicates that the Type II failure is time-dependent. A similar result was observed when the battery seal was artificially compromised under ambient conditions. Hence the low temperature thermoplastic sealant (M.P. 98°C) seemed to be the limiting factor for survivability from a manufacturing standpoint.

Further examination of treated batteries revealed that sealant bubbling and flowing affects TFBs tested at temperatures as low as 121°C. This led to the formation of a disbonded front along the edges of the active components (Figure 17(b)) and over the leads (Figure 17(c)). For the TFBs processed under vacuum bag, the sealant was squeezed out of the battery edges (Figure 17(d)). However the functionality of these

batteries, as tested for short and long term survivability, was not affected. Further screening conducted on as-received batteries revealed that some had long strands of bubbles along the edges of the active components. The bubbles were suspected of diffusing within the sealant layer when the processing temperature led to melting of the Surlyn. Depending on the concentration of bubbles and the viscosity of the sealant, access paths to the active components could have propagated through the sealant itself. Based on these findings, successful embedding tests with full capacity retention were performed by co-bonding the TFB within a composite laminate through press molding of prepreg materials at 132°C and 517 kPa, with a cure time of 2 hours, Figure 7-9.

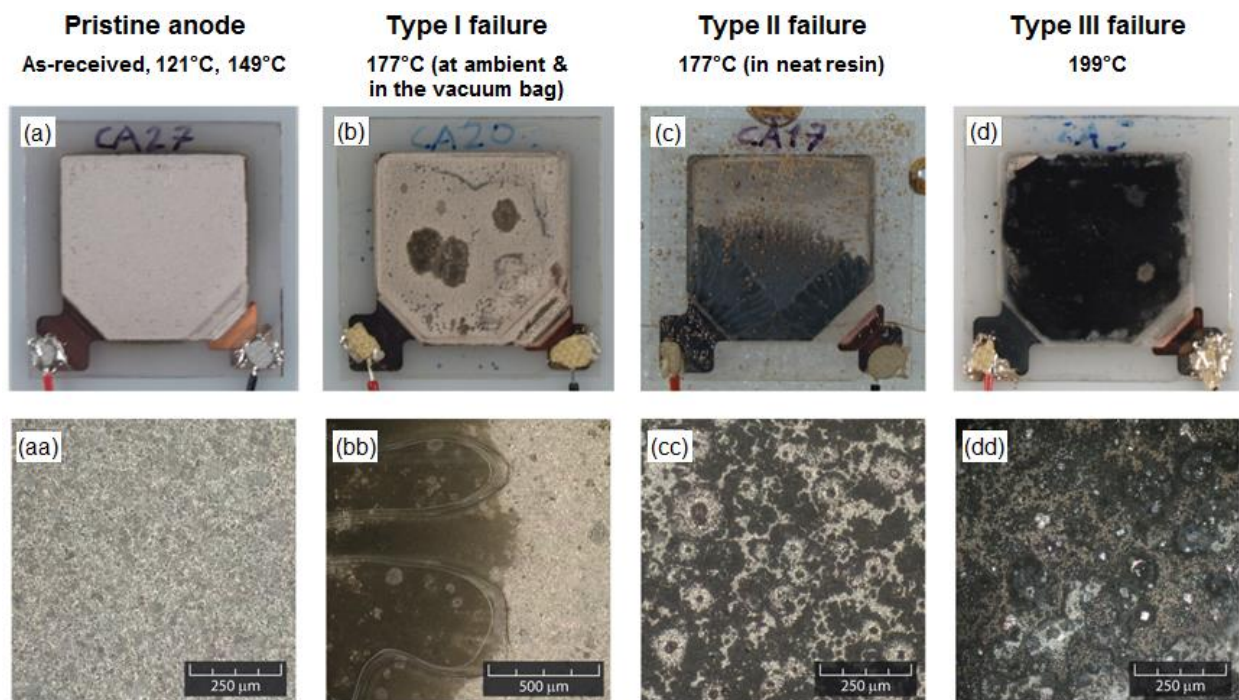


Figure 15. Anode side photos of the three observed types of failures ((b), (c) and (d)) compared to an as-received battery (a). Corresponding dark-field optical micrographs are shown in (aa) through (dd). Type I failure is associated with a local breakdown of the electrolyte and bubbling of the sealant (Surlyn M.P. of 98°C). Reaction of the anode with contaminants results in a Type II failure. Type III failure occurs above the Li melting temperature (181°C). Type I leads to partial capacity loss; Type II and III lead to total capacity loss.

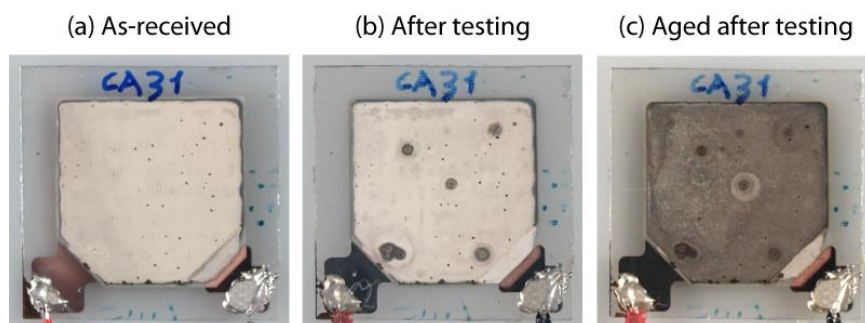


Figure 16. A battery thermally tested at 177°C in the vacuum bag for an hour showing (b) the formation of Type I failures immediately after testing, followed by (c) Type II formations with two months of aging. Immediately after testing the cell exhibited 87% capacity retention; however, it completely failed with aging

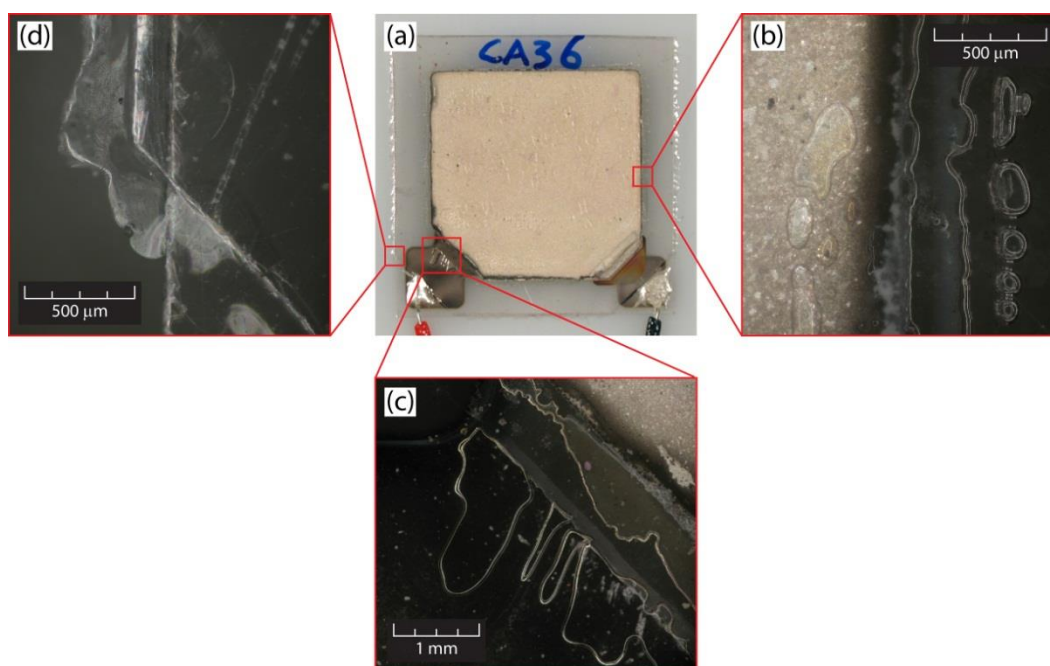


Figure 17. The appearance of bubbles and a disbonded front (b) along the edge of the active components and (c) over the leads after 121°C processing. In some as-received batteries, bubbles are apparent along the edge of the active components. (d) shows the Surlyn which has been squeezed out of between the muscovite layers (exhibiting a slight offset); this is typical of the specimens tested under vacuum bag.

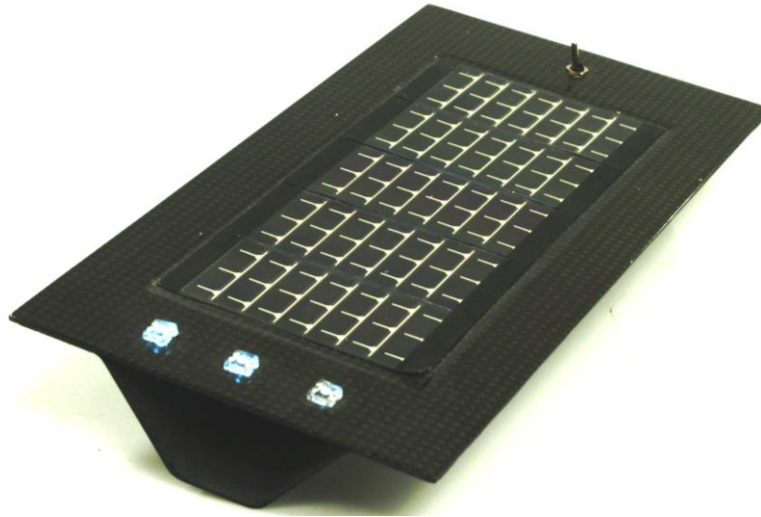


Figure 18. Working prototype of energy self-sufficient CFRP stiffened panel with integrated TFBs and flexible solar array

1.5 Conclusions

This study identified the limits of processability of solid state thin film lithium batteries embedded into composite laminates. Cure temperature was the most influential parameter for battery survivability during composites manufacturing. Successful embedding tests with full capacity retention were performed with glass fiber/epoxy and carbon fiber/epoxy cured at 132°C and 517 kPa. With the proper procedure it was possible to successfully cure the battery inside the laminate up to 149°C. If higher processing temperatures were reached, either locally due to resin exothermic reaction or by design, the battery's electrical performance deteriorated. Battery failures were caused either by the TFB polymeric sealant failure, or by the physical-chemical degradation of the electrolyte or the Li-anode. The mechanical and electrical characterization of laminates with embedded batteries under applied strain and curvature is reported in Chapter 2. The integration of all-solid-state thin-film batteries within composite laminates could find application in commercial and military lightweight multifunctional structures, Figure 18.

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2. Experimental characterization

2.1 Summary

The concept of a multifunctional laminated composite airframe material for load bearing and electrical energy storage is proposed. The laminated structure is comprised of integrated solid state thin film lithium-ion batteries and carbon fiber/epoxy laminae. Mechanical and electromechanical tests were conducted in order to characterize the stress-strain field and determine the operational envelope of the hybrid laminate featuring the current thin film battery technology. The limits of applicability of classical analysis methods in mechanics of composite materials were assessed and several critical failure modes were determined to support the design of the next generation of airborne structural thin film batteries.

2.2 Nomenclature

su– identifies upper substrate property

sl– identifies lower substrate property

CFRP– identifies CFRP laminate apparent property

s– identifies sealant property

a– identifies adhesive film property

ac – identifies active components properties

σ_{x-n} – normal stress in layer n

τ_{xz-n} – shear stress in layer n

ε_{x-n} – normal strain in layer n

γ_{xz-n} – engineering shear strain in layer n

u_n – displacement of layer n along x-axis

P – load in CFRP layer

Q – load in lower substrate

R – load in upper substrate

E_n – Young modulus of layer n

G_s – shear modulus of sealant

t_n – thickness of layer n

w – TFB width (along y-direction)

l – TFB length (along x-direction)

2.3 Introduction

2.3.1 Literature review

The consolidation of batteries and airframe into a multifunctional structure can theoretically reduce the aircraft weight by exploiting the battery components as load bearing elements, by eliminating batteries supports and by enabling distributed power supply and storage, potentially reducing the amount of wiring. Currently sought-after for unmanned aerial vehicles (UAV), this design solution could lead to a significant improvement of the flight endurance of electrical propeller-driven aircraft [1, 2]. To realize this vision, new constituents and a novel design of the composite material system have to be developed to enable simultaneous electrical energy storage and mechanical load bearing capabilities.

Research on lithium-polymer (Li-Po) batteries showed that elastic moduli and strengths of mechanically reinforced batteries as high as 1.02 GPa and 3.9 MPa respectively can be achieved, retaining a specific electrical energy of 160 Wh/kg [3, 4]. These pioneering works proved the capability of the multifunctional airframe concept for a micro-UAV. However the lack of stiffness of the battery packaging [4], as well as the plasticized state of the relatively thick organic polymer electrolyte [5], compromises the mechanical properties of the current Li-Po technology. Furthermore today's available electrode materials, consisting of lithium intercalation compounds, are such that high specific energy and specific power, as well as good mechanical properties cannot be achieved [3,4,6,7]. In [6] a battery with an equivalent Young's modulus of 3.1 GPa was developed, whereas the specific energy dropped to 35

Wh/kg. The causes of the low electrochemical performance were the insufficient electrical capacity of the carbon nano-fiber reinforced lithium transition metal oxide cathode and the low ionic conductivity of the solid state polymer electrolyte. These multifunctional composites can lead to a system improvement for applications with either low mechanical or electrical demands. However, mechanical and electrical performances have to be simultaneously increased in order to fulfill the high specific modulus and strength, as well as the high specific electrical energy and power, required by a lightweight airborne application. The literature review gives evidence that improvements are needed not only in the multifunctional performance of the electrolyte and the electrodes, but also in the mechanical design of the composite structure.

Organic polymer electrolytes with engineered mechanical properties have been studied for three decades as solid state electrolytes [5]. Their ionic conductivity is still inadequate to deliver the power density required by most conventional applications, explaining why they are exclusively employed in a gel state obtained by plasticizing them with the addition of liquid electrolytes, as confirmed by the most recent studies, like [8]. Notably in [9 - 11] the effects of constituents concentration and glass transition temperature, polymers chemistry and architecture on the electrolyte multifunctional properties were thoroughly investigated by synthesizing and analyzing a large variety of vinyl ester-based solid electrolytes and epoxy-based gel electrolytes. The study demonstrated and characterized the inverse correlation between ionic conductivity and elastic modulus, but it also showed a promising trend of increased multifunctional performance, with conductivities approaching the values required for thin film batteries.

Cathode materials are the limiting factor in today's battery systems in terms of specific capacity and specific power, due to their low rate of ionic diffusion and electronic conductivity. For this reason they typically have to be mixed with electrically conductive diluents [12,13]. Adding a large quantity of structural binder to the mixture, as needed to accomplish the load bearing function, causes the specific

and volumetric electrochemical performances to drop even more. This is particularly undesirable for aeronautical applications.

The development of a structural anode seems to be a relatively easier task because available anode bulk materials have an intrinsically higher specific capacity than cathode materials. Above all are silicon and graphite, the latter being not only an intercalation material for lithium ions, but also an electrically conductive and structural material [7].

2.3.2 Multifunctional thin film battery (TFB)

In order to meet the requirements for the next generation of airborne load bearing batteries, solid state thin film Li-ion batteries (TFB) with nanostructured electrodes were recently proposed [8,14-25], Figure 1. The thin film structure maximizes the specific contact surface between electrodes and electrolyte, thereby increasing the stored energy and power per unit mass by increasing the fraction of reactants and the rate of the electrochemical reaction respectively. In fact, downsizing the electrodes and electrolyte to a thin film dramatically reduces the path length for ionic and electronic transport, allowing to achieve the desired conductivity without utilizing diluents or micro-porous compounds [5,14].

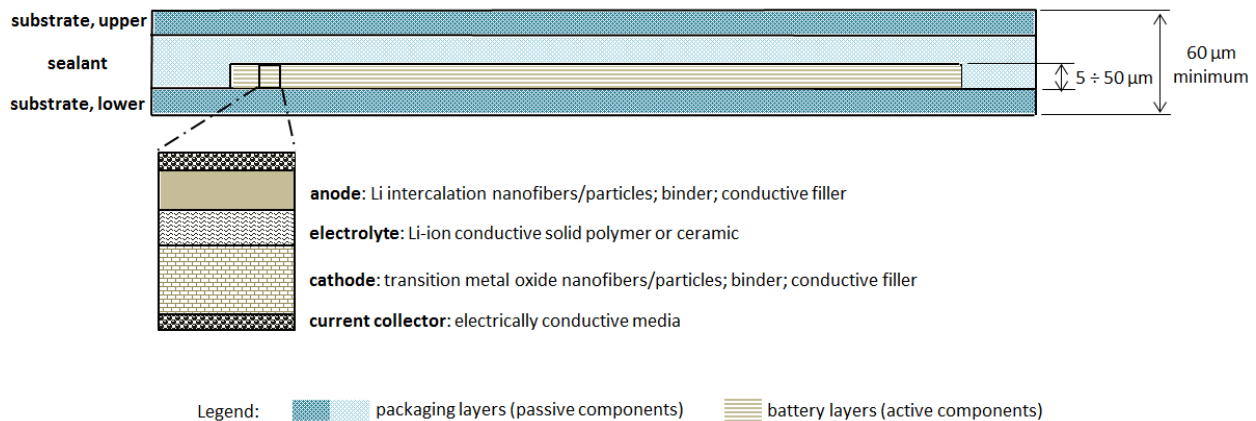


Figure 1. Cross-sectional schematic of a next generation all-solid state thin film Li-ion battery (TFB). Typical thicknesses are indicated. Envisioned design is based on current research status on nanostructured electrode materials.

Scalable manufacturing processes for nanostructured materials, such as electrospinning, chemical vapor deposition (CVD), atomic layer deposition (ALD) and others are suitable for TFB manufacturing. The future application of nano-scale technology to the electrode materials can potentially lead to several advantages. First, it enables further increase of TFB specific energy and power by decreasing the path length of Li-ion and electronic transportation, allowing full exploitation of the theoretical electrical capacity of the materials [14-18]. It can also implement energy storage and load bearing multifunctionality more efficiently than bulk materials, through engineered nanocomposites comprised of electrochemically active nanostructures bound by an electrically conductive reinforced composite. Lastly, nanostructured materials allow better accommodation of the cycling strain induced by the mechanical boundary conditions and by Li-ion insertion and removal. The latter is a known cause for low durability and capacity fading of electrode materials, in particular for silicon, which provides the highest capacity among the known anode materials [15,19-21]. However, nanostructured electrodes are not technology ready. Although several of such cathode materials were synthesized from lithium transition metal oxides, vanadium or manganese oxides and transition metal phosphates [14,17,18], as well as anode materials from silicon and graphite [8,15,16], none of them are commercially available. The reason is the complex synthesis, the low packing of particles and the occurrence of undesirable reactions with the electrolyte that undermines durability.

Finally, all the battery materials have to be solid state to ensure three-dimensional continuity of the load path at any point within the structure, as the presence of discontinuities, such as porosity, voids or delaminations, is a major threat for the mechanical integrity of a composite laminate. Solid state batteries also ensure higher safety since they do not leak hazardous chemicals if a mechanical failure of the packaging occurs. In addition, the solid state electrolyte is also not combustible and does not suffer from thermal runaway, which can cause explosion of conventional lithium-ion prismatic batteries. Besides, a liquid electrolyte would not allow the manufacturing of a thin film cell because of its liquid surface tension and because of the need of a porous separator between the electrodes to avoid electrical shorting.

The low TFB thickness, typically less than 300 μm , facilitates the integration within thin sections and laminated composite structures, but it also implies that a significant weight fraction of the battery is constituted by the packaging layers. The simplest configuration of the battery packaging is comprised of three layers, Figure 1: a lower substrate provides mechanical support for the in-situ deposition process of the electrochemical cell components; a sealant layer ensures that the highly reactive and hygroscopic cell materials are sealed; an upper substrate provides an almost symmetric stacking sequence. Such a casing allows safe processing and lasting operation of the TFB outside of the deposition chamber, while providing in-plane strength and stiffness to the battery lamina.

The integration of TFBs and structural composite laminae, such as carbon fiber/epoxy (CFRP) plies, into a hybrid thin film lithium ion-graphite composite battery (TFB-CFRP) laminate poses manufacturing and design challenges. The compatibility of the commercial state-of-the-art TFB cell to the epoxy based composite curing pressures and temperatures was previously assessed by the authors. This electrochemical cell was comprised of a nanocrystalline lithium cobalt oxide (LiCoO_2) cathode, a ceramic LiPON ($\text{Li}_{2.9}\text{PO}_{3.3}\text{N}_{0.46}$) electrolyte and a metallic lithium (Li) anode, Figure 2 and Figure 3 . The study, which is published separately, successfully determined the cure cycle and the TFB charge level that prevented the physiochemical degradation of the electrochemically active materials due to the high temperature exposure and preserved the full electrical functionality.

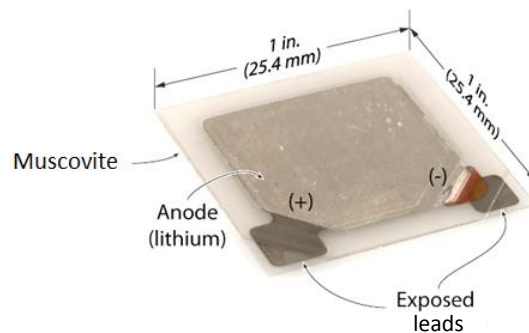


Figure 2. Perspective photo of all-solid state thin film Li-ion battery with dimensions. Manufactured by FrontEdge Technology Inc.

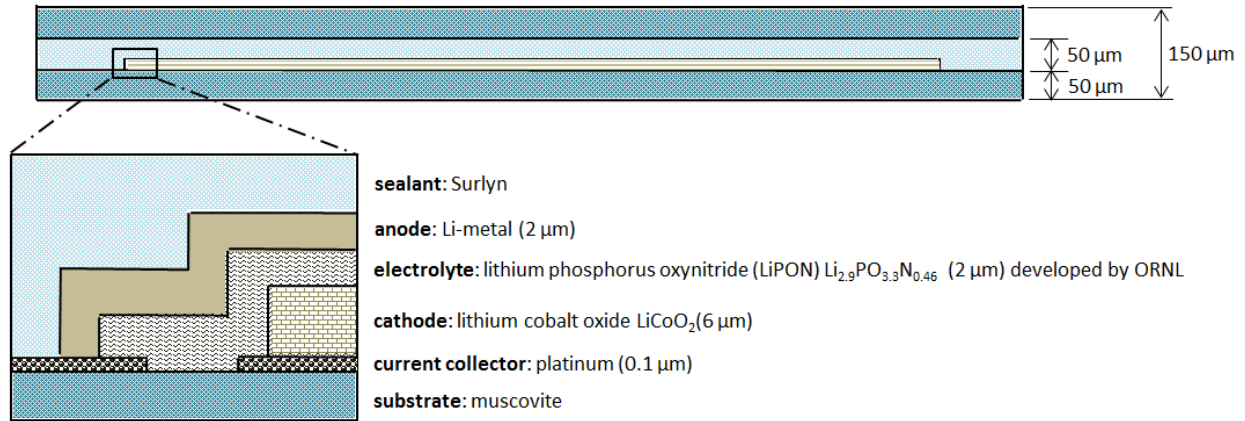


Figure 3. Cross-sectional schematic of the commercially available TFB showed in Figure 2.

The multifunctional design of the battery packaging to achieve integrity of the electrochemical cell as well as efficient structural capability has never been investigated. Such a design is an enabling technology for the structural battery concept. The preliminary step is to characterize the stress-strain field and the electromechanical failure modes of the TFB-CFRP laminate under loading. Whether the TFB is embedded within the CFRP sub-laminate, Figure 4 (a), or externally bonded onto its surface, Figure 4 (b), a three-dimensional stress-strain field has to be considered because of the lay-up transition at the battery ends, as well as of the highly inhomogeneous mechanical properties of the material layers involved. Applicable failure modes include ply failure, disbonding, mixed-mode delamination at multiple bi-material interfaces and delamination buckling. Any of these mechanical failures can lead to the electrical failure by introducing an electrical discontinuity, an electrical short or a path for air and moisture through the battery packaging, leading to contamination of the electrochemically active materials.

Research conducted in [22,23] showed that the aforementioned commercial TFB was capable to operate up to a radius of curvature of 181 mm [22] and under out-of-plane pressures as high as 830 kPa [23] without any detrimental effects on the electrochemical performance. The same TFB type was successfully embedded in a CFRP laminate and electrochemically characterized under uniaxial mechanical tension.

The study focused on the laminate residual mechanical strength and stiffness, which was not penalized by the presence of the TFB embedded at the laminate mid-plane. On the other hand the TFB electrical failure, which consisted of an irreversible and complete loss of capacity, occurred prematurely at about 50% of the laminate mechanical failure strain [24,25]. These electromechanical tests proved that the mechanical boundary conditions have no effect on the electrochemical performance up to a sudden electrical failure. This was confirmed by the multi-physics finite element analysis developed by [21]. The finding implies that the sudden electrical failure is caused by a mechanical failure, such as the failure of the packaging layers, with subsequent cell contamination or tearing, or by cracking of the electrodes or the electrolyte.

To date, neither the stress-strain field nor the mechanical failure modes have been characterized for basic loading conditions. This fundamental knowledge is required in order to prove the functionality and assess the benefits of an optimized TFB-CFRP system, which includes a battery packaging specifically designed for this multifunctional application. Unlike standard flexible electronics, the packaging has to be physically and chemically compatible with the TFB deposition and annealing process, as well as highly performing in terms of specific modulus and specific strength, and capable of ensuring sealing integrity when subjected to severe stress.

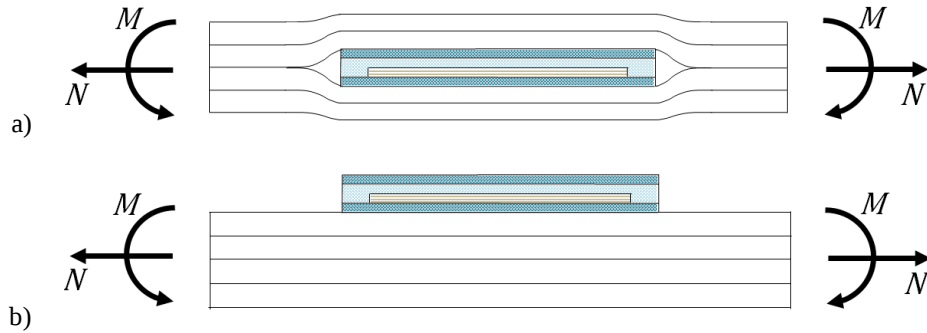


Figure 4. Hybrid thin film lithium ion-graphite composite battery (TFB-CFRP) laminate configurations: thin film battery (a) embedded within the carbon fiber /epoxy laminate, or (b) bonded onto the laminate surface.

Taking advantage of the highest specific energy on the market, currently available TFBs used as structural elements could theoretically reduce the weight of certain aircraft types even without relying on load bearing electrodes, provided that the packaging possesses mechanical properties comparable to airframe structural materials. Hence the structural efficiency of substrate and sealant becomes a determining factor of success.

2.3.3 Semi-empirical performance assessment of structurally integrated TFB

In order to clarify the concept we analyzed the weight reduction obtained by replacing the energy storage system of a propeller aircraft with a structural TFB system. The weight of the original propulsion system, based on an electric engine or an internal combustion engine, is assumed to be the same as an equivalent electric propulsion system. It is important to note that this assumption is applicable to reciprocating and small turbine engines, but it is not valid for turbofan cores, which have a higher specific energy than any currently available electric motor [26]. The new aircraft weight W^* is given by

$$W^* = W - W_{F/B} + W_{TFB} \quad (1)$$

where W is the original gross weight at takeoff, $W_{F/B}$ is the weight of the fuel (W_F) or of the original batteries (W_B) and W_{TFB} is the weight of the TFB active components required to power the aircraft. For preliminary calculation purposes, it is assumed that the packaging has the same specific strength and specific modulus as the airframe materials being replaced. This permits omitting its weight from the above equation.

The new battery weight W_{TFB} is equal to the total propulsion energy required for the new aircraft configuration to complete a mission, E_S^* , divided by the product of the TFB specific energy, e , and the efficiency of the new electric motor, η_M

$$W_{TFB} = \frac{E_S^*}{e\eta_M} \quad (2)$$

The new mission energy, E_S^* is therefore defined as the energy delivered by the engine and is equivalent to the total drag of the new configuration multiplied by the flight range and divided by the propeller efficiency.

$$E_S^* = \frac{D^* d}{\eta_P} \quad (3)$$

Imposing that the range of the aircraft remains the same and assuming the propeller efficiency is the same for the two configurations, the original mission energy is

$$E_S = \frac{D d}{\eta_P} \quad (4)$$

which gives

$$E_S^* = E_S \frac{D^*}{D} \quad (5)$$

In a steady level flight the drag is defined as

$$D = \frac{1}{2} \rho V^2 C_D A \quad (6)$$

By writing the above equation also for the new configuration, and assuming that the cruise speed remains the same we obtain that the ratio of the drag is equal to the ratio of the drag coefficients

$$\frac{D^*}{D} = \frac{C_D^*}{C_D} \quad (7)$$

Imposing that the aerodynamic efficiency of the new configuration is the same as the original configuration, the ratio of the drag coefficients is also equal to the ratio of the lift coefficients

$$\frac{C_D^*}{C_D} = \frac{C_L^*}{C_L} \quad (8)$$

which in turn equals the weight ratio

$$\frac{C_L^*}{C_L} = \frac{W^*}{W} \quad (9)$$

From equations (9), (8), (7) and (5), we can finally obtain that for an electric aircraft, E_S^* is related to the total mission energy of the original configuration E_S as follows

$$E_S^* = E_S \frac{W^*}{W} \quad (10)$$

By algebraic substitution of equation (10) into equation (2) and (1) we can obtain the following relation

$$\frac{W^*}{W} = \frac{1 - \frac{W_B}{W}}{1 - \frac{E_S}{e\eta_M}} \quad (11)$$

For an aircraft equipped with an internal combustion engine a similar equation can be derived with the additional assumptions that the rate of fuel consumption and the aerodynamic efficiency remain constant throughout the flight, which means that

$$\frac{C_L^*}{C_L} = \frac{W^*}{W - W_F/2} \quad (12)$$

where C_L is an average lift coefficient to accounts for the aircraft weight change during flight. The combination of equations (9), (10) and (12) provides

$$E_S^* = E_S \frac{W^*}{W - W_F/2} \quad (13)$$

By plugging the above mission energy into equations (1) and (2) we finally obtain

$$\frac{W^*}{W} = \frac{1 - \frac{W_F}{W}}{1 - \frac{E_S}{e\eta_M} \frac{W}{W - W_F/2}} \quad (14)$$

Empirical data from different aircraft categories were averaged in order to calculate the nondimensional groups of aircraft weights in equation 11 and 14. The data are summarized in Table 1. The electric motor efficiency, η_M , was assumed equal to 0.95. To calculate the mission energy, E_S , the original flat rated engine power was multiplied by the maximum range and divided by the cruise speed, thereby neglecting takeoff and landing. The resulting semi-empirical curves for the weight change ratio are plotted in Figure 5 at increasing TFB energy densities, starting from the today's available specific energy of 353 Wh/kg. The white plot area identifies the aircraft configurations that benefit from weight saving. The grey plot area indicates weight increase. The model shows that weight saving could be currently achieved for electric aircraft such as High Altitude Long Endurance (HALE), mini-UAV and motor gliders. For aircraft characterized by higher mission energy per unit mass, such as UAVs with internal combustion engines and general aviation, further improvement in specific energy is required. However, a battery

packaging with the assumed structural efficiency is not technology ready and its mechanical performance requirements have to be assessed.

Aircraft	E_s [kWh]	W [kg]	$W_{F/B}$ [kg]	E_s/W [kWh/kg]
Pipistrel Taurus Electro G2 ¹⁾	7.17	472.50	101.00	0.015
NASA Helios ^{2,3)}	21.00	929.00	148.75	0.023
AeroVironment RQ-11 Raven ⁴⁾	0.11	1.90	0.46	0.056
Solar Impulse ⁵⁾	120.00	2000.00	450.00	0.060
AAI RQ-7A Shadow ⁴⁾	34.25	154.00	23.10	0.222
AAI Shadow 400 ⁴⁾	73.47	201.00	68.40	0.366
Cessna 172R ⁵⁾	565.51	1111.00	144.00	0.509
General Atomics RQ-1 Predator ⁴⁾	768.04	1020.00	295.00	0.753
Pilatus PC-12 ⁵⁾	4187.09	4740.00	1226.00	0.883
Piaggio P-180 ⁵⁾	5291.84	5488.00	1271.00	0.964
¹⁾ http://www.pipistrel.si/plane/taurus-electro/technical-data . ²⁾ http://www.nasa.gov/centers/dryden/news/FactSheets/FS-068-DFRC.html . ³⁾ Saft LO 26 SHX battery data sheet. ⁴⁾ Jane's Unmanned Aerial Vehicles and Targets, Issue thirty, May 2008. ⁵⁾ Jane's All the World's Aircraft 2010-2011.				

Table 1. Aircraft weight and power plant energy characteristics utilized to feed the semi-empirical weight prediction model shown in Figure 5.

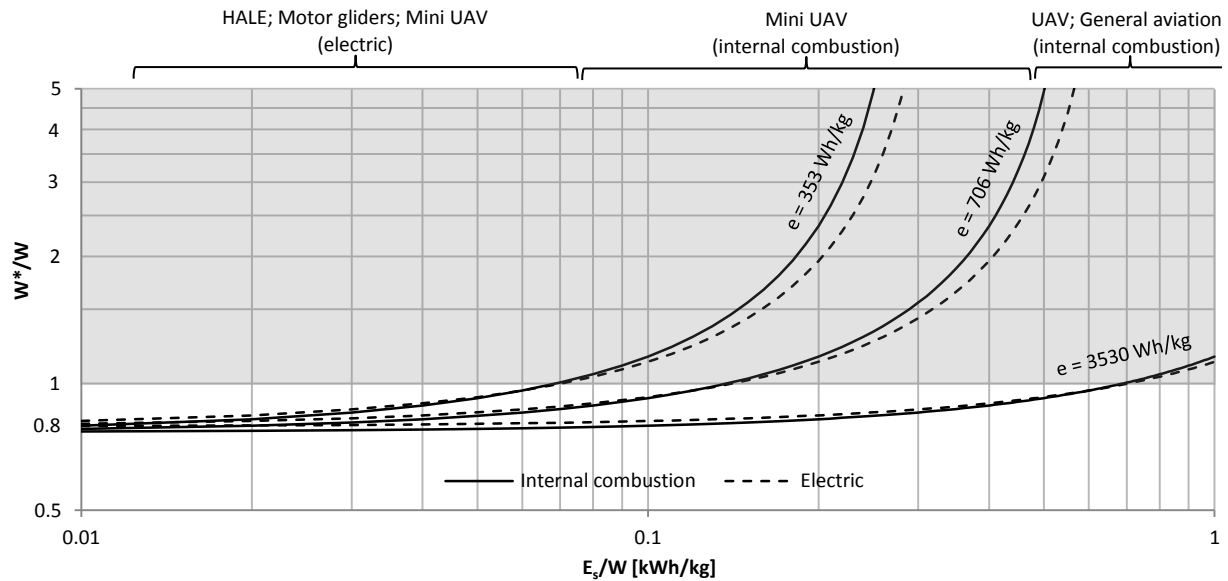


Figure 5. Predicted gross weight of aircraft equipped with TFB energy storage system (W^*) normalized by the original gross weight at takeoff (W), plotted as a function of the original specific mission energy. Semi-empirical curves (as per eq. 3 and 4 and averaged data from Table 1 plotted at increasing TFB specific energies (e)).

2.3.4 Objective of the experimental characterization

The objective of this chapter is to introduce the concept of hybrid TFB-CFRP laminate and to experimentally characterize the stress-strain field and failure mechanism of the multifunctional laminate subjected to uniaxial strain, single curvature and a combination of the two. This work is intended to support the future development of damage initiation and propagation analyses finalized to the optimization of the TFB packaging design. In this perspective, experimental data was required for determining the critical failure modes to be considered and for validating such analysis methods. The research also aims at providing a performance envelope for the current TFB technology that could be used as a baseline for the next generation of airborne thin film structural batteries.

2.4 Methodology

A commercially available TFB was utilized to manufacture TFB-CFRP laminates. The chemistry of the selected battery, as well as its manufacturing process, is representative of the commercial state-of-the-art that is currently adopted by all the manufacturers of solid state thin film lithium-ion batteries. The TFB selected for this study, Figure 2, was deemed the most suitable for this application because of the simplicity of the three-layer design and high stiffness of the packaging. Two laminate configurations were considered. The first was comprised of a TFB embedded at the laminate mid-plane and co-bonded within the CFRP sub-laminate, Figure 4 (a). The second was the secondary bonding of the TFB onto the surface of a pre-cured CFRP sub-laminate, Figure 4 (b).

Mechanical tests were conducted in order to measure the stress-strain field under uniaxial loading and assess the applicability of classical analysis methods in mechanics of composite materials. Disbonding, delamination characteristics and laminate mechanical failure modes were also assessed in order to determine the weakest link and critical properties that led to mechanical failure. The cyclic TFB thickness variation due to the migration of the lithium ions during battery charge and discharge, which could generate interlaminar normal stress in the laminate, was also experimentally measured. Finally, an electrochemical

characterization under mechanical loading was performed in order to determine the strain and curvature at electrical failure.

2.4.1 Materials

The TFBs utilized in this research are manufactured by FrontEdge Technology under license from the Oak Ridge National Laboratory (ORNL), Figure 2. Chemistry and thicknesses of the cell components are summarized in Figure 3. The active components are encased by two 50 μm thick muscovite substrates bound by a thermoplastic layer of Surlyn sealant, leading to a total thickness of 150 μm . The battery is a 25.4 mm square with a nominal voltage of 4.2V, and a nominal capacity of 1 mAh. The specific energy measured by the authors with respect to the mass of active components is 353 Wh/kg, which drops to 22Wh/kg with packaging weight included. Experimental characterizations of the same battery under mechanical loading were performed by [22-25].

The electrochemically active components of the battery are grown by a sequence of different physical vapor deposition (PVD) processes performed in-situ on the substrate. The need of annealing the cathode at temperatures of 300°C or higher to increase the ionic conductivity requires dimensional stability of the substrate material at those temperatures in order to avoid cracking, disbonding or undesired crystalline orientation of the cathode due to thermal stresses. Chemical stability is also important in order to avoid releasing contaminants in the controlled atmosphere of the deposition chamber. All these requirements have to be considered for the selection of a substrate material. The manufacturing process was discussed in detail in [27-31].

The active components are highly reactive with N_2 [27], O_2 [28] and H_2O [32], therefore the TFB has to be hermetically sealed. A failure of the sealant or substrate during manufacturing or operation of the TFB-CFRP leads to the failure of the battery by compromising its ability to store energy. The electrical capacity loss can be almost instantaneous or can fade progressively, depending on the flow rate of

contaminants entering the battery. Since the CFRP material is hygroscopic, this failure mode applies to the embedded TFB configuration as well.

Mechanical properties of muscovite and Surlyn are listed in Table 2. The first is a crystalline mineral whose crystallographic structure is comprised of 1 nm thick layers separated by perfect basal cleavages. This characteristic determines low fracture toughness against cracks that are planar with the battery. On the other hand it can be considered quasi-isotropic in the battery plane [33] with an elastic modulus of 178 GPa, which is in the range of intermediate modulus CFRP materials. Surlyn is a high toughness thermoplastic polymer with a melting temperature of 98°C. A compatibility study to the composite curing process revealed that in some batteries the Surlyn layer is affected by long strands of bubbles along the edges of the active components, and that during curing of the TFB-CFRP laminate bubbles diffused within the sealant layer and coalesced into a disbonded front, Figure 6. These defects could potentially cause delamination onset within the battery packaging, Figure 7.

Property	Symbol	Muscovite ¹⁾	Surlyn	IM7/977-3 ²⁾	AF 163-2 ³⁾
Tensile modulus of elasticity [GPa]	E_1	178 ⁴⁾	0.28 ⁵⁾	162	1.10
	E_2	178 ⁴⁾	0.28 ⁵⁾	8.34	1.10
Shear modulus of elasticity [GPa]	G_{12}	70.7 ⁴⁾	0.11 ⁶⁾	4.96	0.41
Poisson's ratio	ν_{12}	0.26 ⁷⁾	0.3 ⁸⁾	0.34	0.34
Strain to tension failure [%]	ε_1^{TU}	unkn.	8.00 ⁵⁾	1.46	unkn.
	ε_2^{TU}	unkn.	8.00 ⁵⁾	0.77	unkn.
Mode I fracture toughness [J/m ²]	G_{IC}	1.30 ⁹⁾	1200 ¹⁰⁾	316	3682
¹⁾ Average in-plane properties in the plane parallel to the basal cleavage, which coincides with the battery plane. ²⁾ Cytec IM7/977-3 data sheet. ³⁾ 3M Scotch-Weld Structural Adhesive Film AF 163-2 data sheet. ⁴⁾ McNeil et al., <i>J Phys.: Condens. Matter</i> 5, 1681 (1993). ⁵⁾ Yield strain, ref. NCAR Report FRB-4 (1965). ⁶⁾ Calculated assuming isotropy and $\nu = 0.3$. ⁷⁾ Calculated assuming isotropy in the battery plane. ⁸⁾ Assumed. ⁹⁾ Hill et al., <i>Int. J. Fracture</i> 119/120, 365 (2003). ¹⁰⁾ Compston et al., <i>J Mater. Sci. lett.</i> 20, 509 (2001).					

Table 2. Relevant mechanical properties of materials.

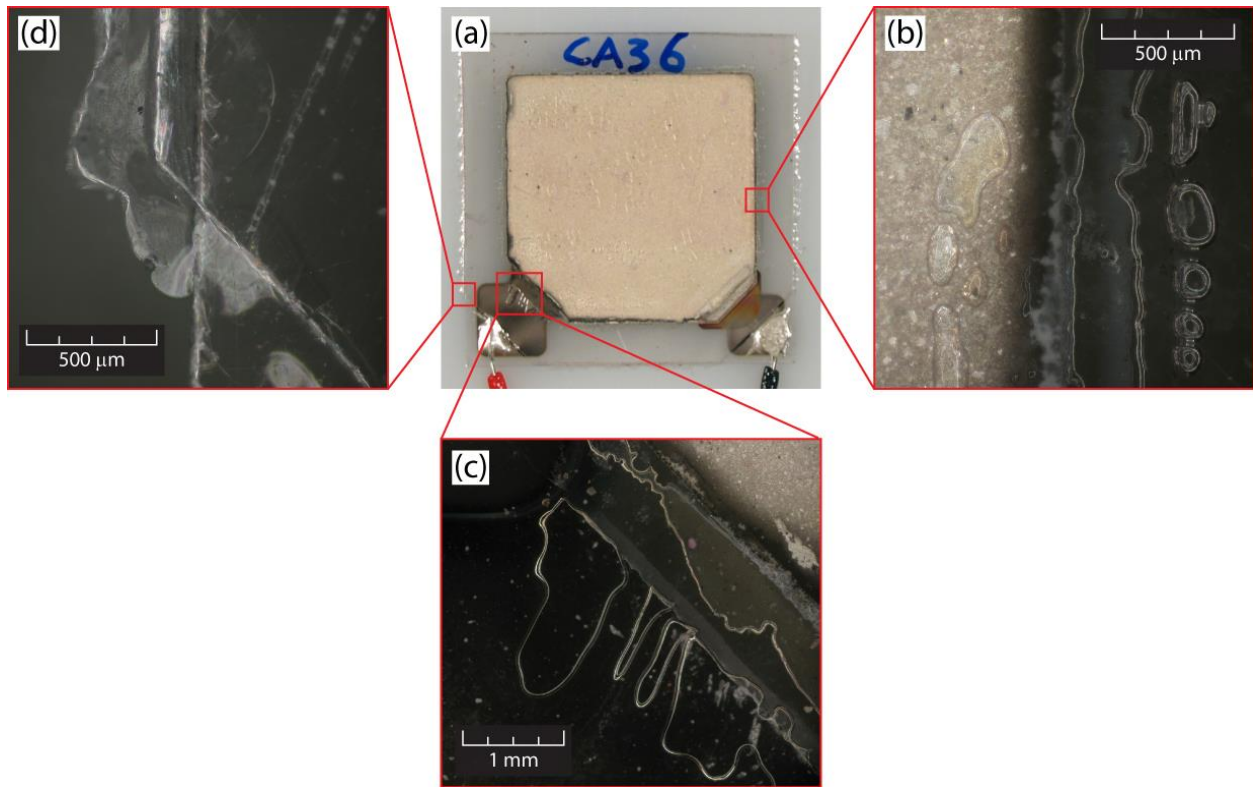


Figure 6. Micrographs of a TFB after processing at 121°C for one hour under vacuum bag (-711 mmHg) showing sealant bubbling and a disbonded front along the edge of the active components.



Figure 7. Cross-sectional schematic of the TFB with locations of disbonded areas, as shown in Figure 6, highlighted with dotted lines.

The TFB-CFRP laminates were cured by heated press molding of IM7/977-3 prepreg tape materials at 132°C and 590 kPa, with a cure time of two hours. In the case of embedded TFB, the battery was laminated within the prepreg material prior to curing and without the addition of adhesive film (AF). For the externally bonded TFB configuration, the CFRP laminate was pre-cured with the same process and the battery was subsequently bonded using one layer of epoxy adhesive film 3M AF163-2 cured for 1.5 hours at 121°C in the vacuum bag. These cure cycles were validated by the authors in order to retain the

full functionality of the battery. The mechanical properties for the carbon/epoxy material and the adhesive film are listed in Table 2. For the electromechanical characterization of the TFB-CFRP laminate, the battery was connected to the testing circuit through flat flexible cables (FFC), Nicomatic 254PW01E6095 polyester coated single copper conductor. The FFCs were 0.25 mm thick and 5.12 mm wide. They were connected to the TFB leads using MG Chemicals silver conductive epoxy 8331-14G. For the embedded battery configuration they were laminated within the CFRP at the laminate mid-plane.

Unlike previous studies [24,25], the battery was not encased within a pre-cured polymeric case, but silicone conformal coating MG Chemicals 422-55 was applied on the exposed leads only, Figure 2, to insulate the connections and prevent electrical shorting with carbon fibers.

2.4.2 Experimental characterization

The experimental campaign was comprised of five test types: double cantilever beam (DCB); uniaxial mechanical tension; battery thickness variation occurring during charge/discharge cycling; uniaxial mechanical tension with battery capacity monitoring and four point bending with battery capacity monitoring, Table 3. The battery thickness variation test setup and procedure were conceived ad hoc since a test standard is not available. The remaining tests adopted ASTM standard methods for reinforced plastics modified to cope with the multifunctional system.

The specimens for DCB test were 25.4 mm wide, which is equal to the TFB width, and 304.8 mm long. Tests were conducted according to the ASTM standard [34]. Three baseline specimens, comprised of thirty plies oriented at 0° for a laminate thickness of 3.8 mm, were tested to determine the mode I fracture toughness G_{IC} of the CFRP material. Three additional specimens with a TFB embedded at the laminate mid-plane were then tested. These specimens featured a battery embedded at specimen mid-span, leading to the stacking sequence (0_{30}) away from the battery and $(0_{15}/TFB/0_{15})$ at battery location. The crack was started from the loaded end of the specimen, at the laminate mid-plane, and propagated towards the TFB. A precrack was induced so that a delamination was visually observed on the edge of the specimen before

starting the test. The objective was to determine the G_{IC} and the crack propagation path. The G_{IC} was calculated by dividing the total strain energy released during the test by the final crack surface measured from the tip of the precrack.

Test description	Lay-ups	Repetitions
Double cantilever beam (DCB)	(0 ₁₅ /TFB/0 ₁₅)	3
Uniaxial mechanical tension with full-field strain monitoring	(0/90/TFB/90/0)	1
	(0 ₂ /TFB/0 ₂)	1
	(0/45/90/-45/TFB/-45/90/45/0)	1
	[(0/45/90/-45) _s /AF/TFB]	1
	(0 ₈ /AF/TFB)	1
TFB thickness variation during charge/discharge cycling	TFB alone	1
Uniaxial mechanical tension with TFB capacity monitoring	(0/45/90/-45/TFB/-45/90/45/0)	2
Four point bending with TFB capacity monitoring	[(0/45/90/-45) ₃ /TFB/(-45/90/45/0) ₃]	2
	[(0/45/90/-45) _{3s} /AF/TFB]	2
	[TFB/AF/(-45/90/45/0) _{3s}]	2
	(0/90/TFB/90/0)	2

Table 3. Summary of tests.

Uniaxial mechanical tension tests with full-field strain monitoring were performed in order to characterize the stress-strain field for different laminate lay-ups. The procedure complies with the standard ASTM test [35], except for an increased specimen width of 76.2 mm, Figure 8. This modification was introduced to minimize the interaction between the edge effect and the stress-strain gradient caused by the TFB, which was located at the center of the specimen. A total of three lay-ups with embedded battery were investigated: a 4-ply cross-ply laminate, a 4-ply unidirectional laminate tested along the 0° direction and an 8-ply quasi-isotropic laminate. The embedding method adopted throughout this study is referred to as interlaminar embedding, since it consisted of enclosing the battery in the space between two plies, as opposed to enclosing it into a ply cut-out. This design avoided sharp stress-strain gradients to arise at the battery edges, although it led to resin pockets at the TFB ends, Figure 9. The applied strain was increased only up to a far-field strain of approximately 3000 μ strain in order to observe the elastic behavior. The failure behavior for this laminate configuration was already reported in [24,25].

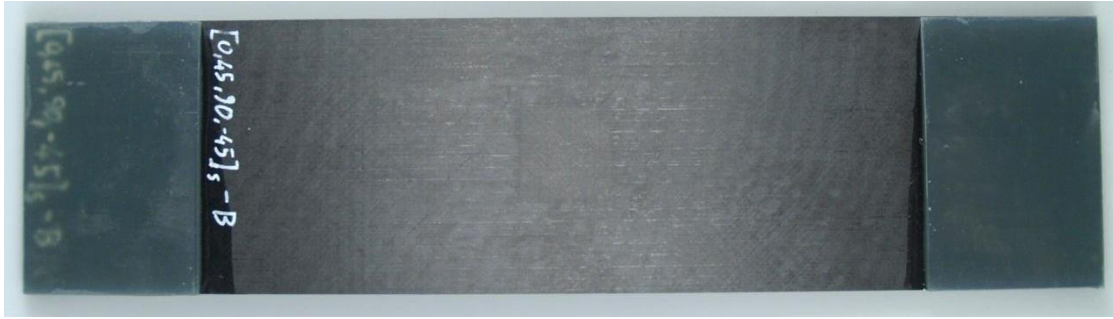


Figure 8. Uniaxial mechanical tension test specimen with embedded TFB at laminate mid-plane. Laminate stacking sequence is (0/45/90/-45/TFB/-45/90/45/0).

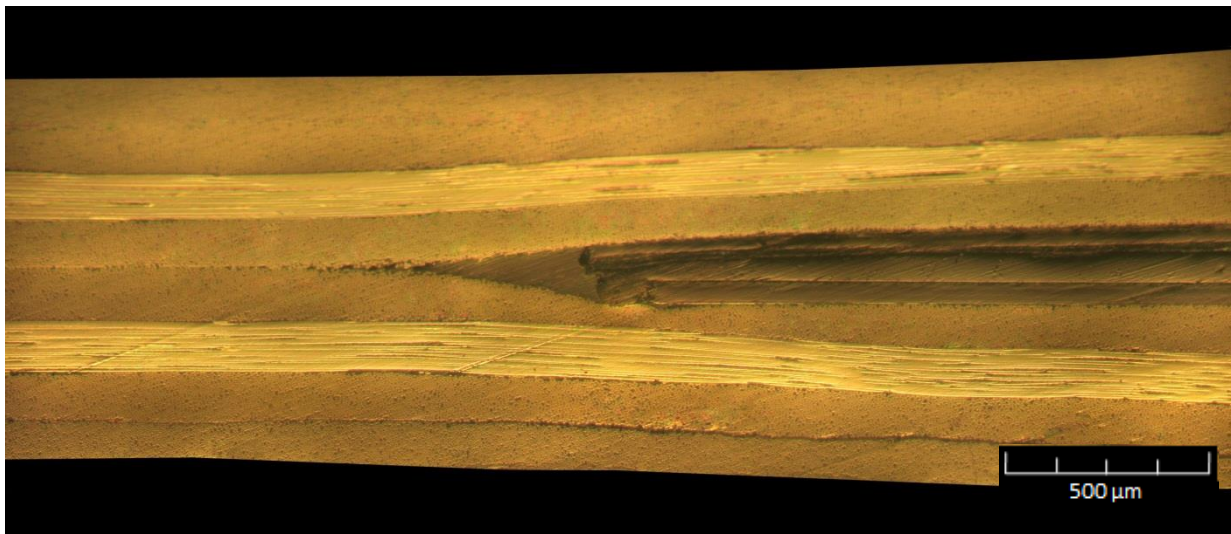


Figure 9. Micrograph of quasi-isotropic laminate with TFB embedded at laminate mid-plane. Laminate stacking sequence is (0/45/90/-45/TFB/-45/90/45/0).

Two additional 8-ply lay-ups with an externally bonded TFB were tested in uniaxial tension: a unidirectional laminate tested along the 0° direction and a quasi-isotropic laminate. Their stacking sequences at battery location were (0₈/AF/TFB) and [(0/45/90/-45)_s/AF/TFB] respectively. A specimen with an externally bonded battery is shown in Figure 10. The test procedure was the same as for the embedded TFB configuration, but the applied load was increased up to failure in order to investigate the elastic stress-strain field as well as the failure mode.

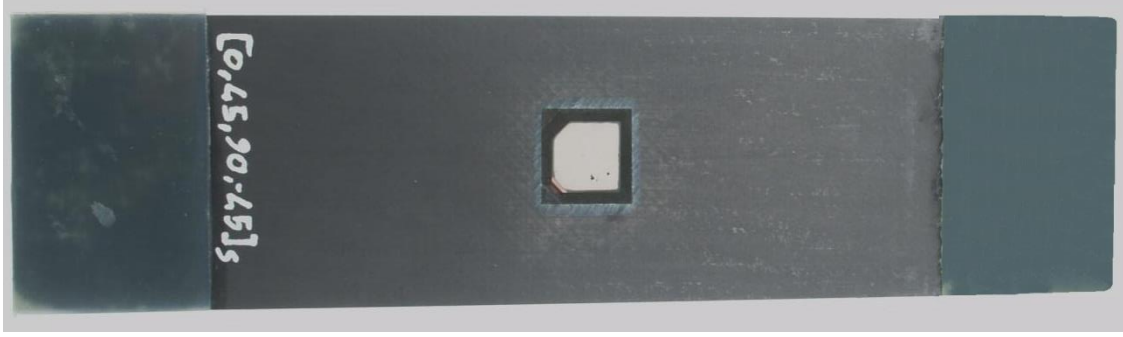


Figure 10. Uniaxial mechanical tension specimen with externally bonded TFB. Laminate stacking sequence at battery is [(0/45/90/-45)_s/AF/TFB].

For all the uniaxial mechanical tension tests a digital image correlation (DIC) technique was adopted to monitor the surface strain of the specimen. To avoid using FFCs that would alter the strain field or interfere with the optical measurement technique, the electrical response of the TFB-CFRP laminate under loading was investigated separately.

Contrary to the common perception that their thickness remains constant, the lithium ion batteries expand during charge and contract during discharge. This thickness, thus volume, change is caused by lithium ion intercalation into host electrode materials. Since the TFB has a metallic lithium anode instead of an intercalation compound, the thickness increase is attributed to the plating of lithium at the anode during the charging process. The DIC was employed for monitoring the thickness increase of one TFB alone during a full electric charge process. A full charge/discharge cycle was performed before the test in order to condition the battery. Discharge was operated under a constant resistive load of 3.8 kΩ, which led to an average discharge rate of 1C (i.e. 1 ma discharge current). Charge took place at a constant voltage of 4.2 V. The battery was considered fully discharged when the voltage reached 3 V, the lower limit to avoid overdischarge, while full charge was reached when the current dropped below 50 μA. The DIC measured the out-of-plane displacement of the TFB outer surface at anode side with time intervals of 2 minutes, starting from the fully discharged state, which was taken as the reference state. Therefore the displacement field corresponding to the fully charged state was equal to the maximum thickness increase.

The TFB temperature was monitored for the entire duration of the test with an infrared (IR) camera with $\pm 1^\circ\text{C}$ accuracy.

Lastly, an electrochemical characterization of the TFB-CFRP laminate subjected to uniaxial tension and flexure was conducted. The purpose was to assess the operational envelope and to determine the interaction between electrical and mechanical failure modes, where electrical failure was the partial or total capacity loss. Two specimens with a stacking sequence (0/45/90/-45/TFB/-45/90/45/0) were tested up to failure through uniaxial mechanical tension. The selected laminate lay-up showed the best correlation between experimentally measured and calculated strain field in the mechanical tests, thereby increasing the degree of confidence for the calculation of the strain at failure. The dimensions of the specimens matched those of the previously described mechanical tension tests, but in this case FFCs were connected to the battery and laminated within the CFRP plies. Figure 11 shows the cables exiting from the laminate and connecting to the charge/discharge circuit (not shown) to allow for capacity monitoring. The discharge or charge capacity was calculated by numerically integrating the discharge or charge current over time. The procedure and characteristics of the charge and discharge circuit were as previously described.

For the mechanical tension tests with capacity monitoring, constant strain intervals of increasing magnitude were applied until failure, where the duration of an interval corresponded to the time required to perform a full discharge followed by a full charge for capacity measurement under constant strain. Load was released after each strain interval and capacity was measured again at zero strain. The far-field strain was measured by an extensometer, Figure 11, for control and data acquisition, while the strain at failure location was calculated using the analysis methods described in the following section.

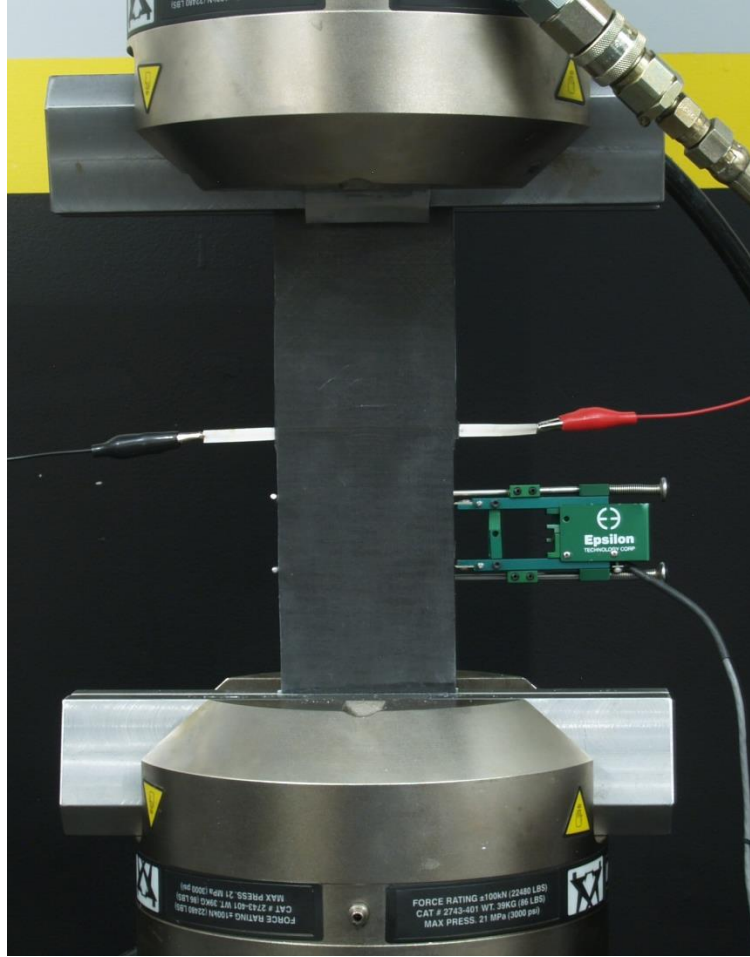


Figure 11. Electromechanical uniaxial tension test setup showing the hybrid TFB/graphite laminate specimen clamped by the grips of test frame. Laminate stacking sequence is (0/45/90/-45/TFB/-45/90/45/0).

Similarly, four point bending tests were conducted on laminates with embedded or externally bonded battery configurations, Figure 12. While in the first configuration the TFB was subjected to pure curvature, the second led to curvature and compressive or tensile in-plane strain depending on which side of the specimen the TFB was bonded on. The width of the specimen was always 76.2 mm, while the support span to thickness ratio was initially set to 60:1 with a support span of 180 mm and a loading span of 90 mm. The laminate stacking sequences were [(0/45/90/-45)₃/TFB/(-45/90/45/0)₃], [(0/45/90/-45)_{3S}/AF/TFB] and [TFB/AF/(-45/90/45/0)_{3S}]. For the embedded battery configuration, a stacking sequence (0/90/TFB/90/0) was also tested with an increased span-to-thickness ratio of 145:1. Support and

loading span were 72.5 mm and 36.5 mm respectively. This modified setup, which does not comply with the ASTM standard [36], was designed for applying higher curvatures to the battery without experiencing mechanical failure of the laminate. The same load-hold load-unload procedure with increasing curvature intervals was adopted, but in this case the test was conducted under displacement control. The curvature applied by a given displacement of the test frame was calculated through integration of the elastic line equation. Small deformations, normality condition of the sections and uniform bending stiffness were assumed. The uniformity of the bending stiffness is applicable only if the presence of the battery does not significantly change the section inertia of the CFRP laminate. The far-field strain applied to the battery was then calculated by multiplying the curvature by half the CFRP laminate thickness. The curvature is reported in the results section as the radius of curvature, which was assumed equal to the inverse of the curvature.

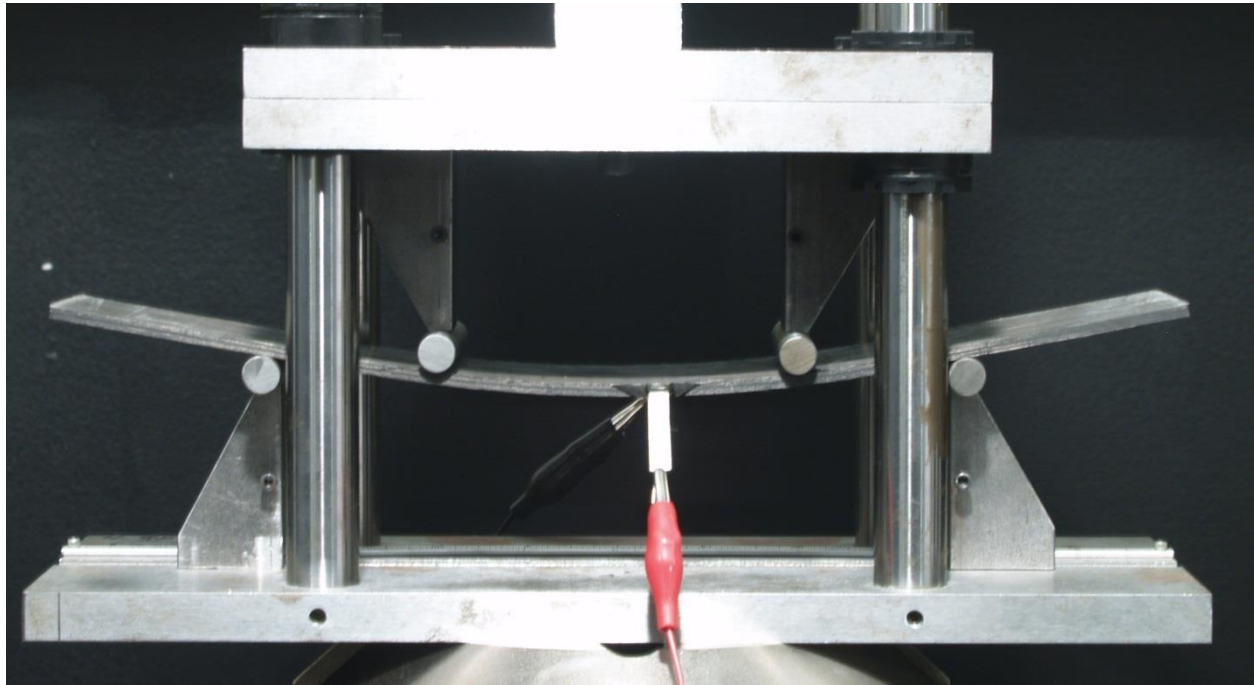


Figure 12. Electromechanical flexure test setup with standard 60:1 span to thickness ratio, showing the hybrid TFB/graphite laminate specimen loaded by the four point bending fixture. Laminate stacking sequence is $[(0/45/90/-45)_3/\text{TFB}/(-45/90/45/0)_3]$.

2.5 Analysis methods

2.5.1 Classical laminate theory

Classical laminate theory (CLT), finite element analysis (FEA) and a closed form elasticity solution of the TFB-CFRP laminate were utilized to interpret the experimental results and calculate the strain at failure. These methods were validated by comparing their results with the experimentally measured elastic strain field from the uniaxial mechanical tension tests. Given the applied far-field strain ε_0 , the strain field at the center of the specimen, which was affected by the presence of the TFB, was calculated and compared to the surface strain field measured by the DIC.

The first analysis method used CLT to calculate the apparent modulus of elasticity in the x -direction at a point away from the battery, where the laminate was comprised of the CFRP layers only, and at the battery location. The model accounted for the stiffness of the battery substrate, sealant and the adhesive film. The x -direction was defined as the loading direction. The laminate strain at the battery was then calculated by multiplying the far-field strain by the ratio of the two moduli multiplied by their respective thicknesses. The results obtained with this method are identified in the results section with the acronym CLT.

At specimen mid-span, the section was not constant across the width due to the presence of the battery. In order to account for the resulting stress redistribution, which was ignored by the first analysis method, a second analysis method that employs a linear finite element solution with NX Nastran SOL101 was developed. FEA also accounted for the extension-bending coupling caused by the laminate asymmetry. The entire specimen was modeled with linear shell elements (CQUAD4) and a mesh size of 1.524 mm that matched the DIC resolution. The element formulation used the CLT for computing element stiffness and outputs, therefore plane stress and uniform strain through-the-thickness were assumed. The desired far-field strain was applied by an enforced nodal displacement at the loaded end. The transverse strain was unconstrained.

2.5.2 Elasticity solution

The third analysis method was a linear elastic shear lag model that was developed in order to capture the extensive three-dimensional stress-strain field of the externally bonded battery configuration. The sealant, identified by the letter s , and the adhesive film, identified by a , were assumed to carry only shear, while the bending and shearing deformation of the upper and lower substrates, su and sl respectively, were neglected. The carbon/epoxy composite material was modeled as an equivalent isotropic material with the apparent laminate Young's modulus in the x -direction calculated according to the CLT. Bending and shearing deformation of the CFRP were also neglected. Perfect bonding was assumed at all interfaces and the presence of the active components was neglected due to their relatively small thickness. Based on these assumptions, when a far-field stress σ_0 is applied to the CFRP laminate, a normal stress σ_x is generated in the CFRP, su and sl layers, and a shear stress τ_{xz} is carried by the a and s layers, Figure 13. No other stress components arise, therefore the equilibrium equations are reduced to three, one for each of the layers that carries axial loading. The addition of the strain-displacement and stress-strain equations leads to a system of thirteen equations with thirteen unknowns.

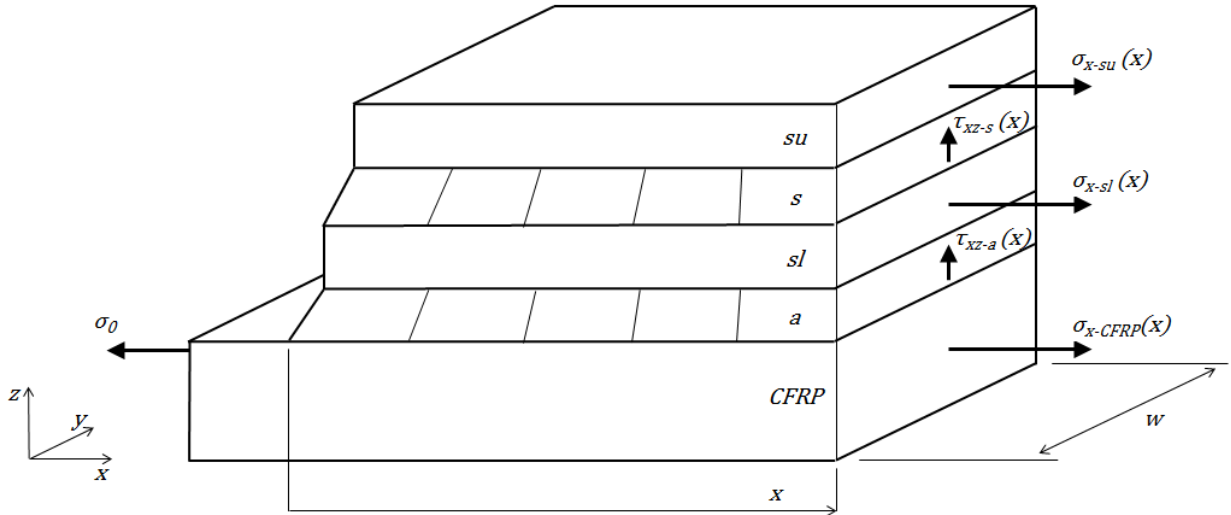


Figure 13. Shear lag elasticity model of the externally bonded TFB, stress representation. Laminate layers are: carbon/epoxy sub-laminate ($CFRP$); adhesive film (a); lower substrate (sl); sealant (s); upper substrate (su).

The equilibrium equations are

$$\frac{d\sigma_{x-su}}{dx} = \frac{\tau_{xz-s}}{t_{su}} \quad (15)$$

$$\frac{d\sigma_{x-sl}}{dx} = \frac{\tau_{xz-a} - \tau_{xz-s}}{t_{sl}} \quad (16)$$

$$\frac{d\sigma_{x-CFRP}}{dx} = -\frac{\tau_{xz-a}}{t_{CFRP}} \quad (17)$$

While the overall equilibrium equation is

$$\sigma_0 t_{CFRP} = \sigma_{x-su} t_{su} + \sigma_{x-sl} t_{sl} + \sigma_{x-CFRP} t_{CFRP} \quad (18)$$

The strain-displacement equations are as follows

$$\gamma_{xz-s} = \frac{u_{su} - u_{sl}}{t_s} \quad (19)$$

$$\gamma_{xz-a} = \frac{u_{sl} - u_{CFRP}}{t_a} \quad (20)$$

$$\varepsilon_{x-su} = \frac{du_{su}}{dx} \quad (21)$$

$$\varepsilon_{x-sl} = \frac{du_{sl}}{dx} \quad (22)$$

$$\varepsilon_{x-CFRP} = \frac{du_{CFRP}}{dx} \quad (23)$$

The stress-strain equations for the five layers are

$$\sigma_{x-su} = E_{su} \varepsilon_{x-su} \quad (24)$$

$$\sigma_{x-sl} = E_{sl} \varepsilon_{x-sl} \quad (25)$$

$$\sigma_{CFRP} = E_{CFRP} \varepsilon_{x-CFRP} \quad (26)$$

$$\tau_{xz-s} = G_s \gamma_{xz-s} \quad (27)$$

$$\tau_{xz-a} = G_a \gamma_{xz-a} \quad (28)$$

By plugging equation (19) and (20) into (27) and (28) respectively and subsequently deriving with respect to x and substituting the normal strains with the corresponding stresses according equations (24) to (26)

$$\frac{d\tau_{xz-s}}{dx} = \frac{G_s}{t_s} \left(\frac{\sigma_{x-su}}{E_{su}} - \frac{\sigma_{x-sl}}{E_{sl}} \right) \quad (29)$$

$$\frac{d\tau_{xz-a}}{dx} = \frac{G_a}{t_a} \left(\frac{\sigma_{x-sl}}{E_{sl}} - \frac{\sigma_{x-CFRP}}{E_{CFRP}} \right) \quad (30)$$

By substituting τ_{xz-s} , τ_{xz-a} and σ_{x-sl} from equations (15), (17) and (18) respectively we obtain

$$\frac{d^2\sigma_{x-su}}{dx^2} = \sigma_{x-su} \frac{G_s}{t_s} \left(\frac{1}{t_{su}E_{su}} + \frac{1}{t_{sl}E_{sl}} \right) + \sigma_{x-CFRP} \frac{t_{CFRP}G_s}{t_{su}t_{sl}t_sE_{sl}} - \sigma_0 \frac{t_{CFRP}G_s}{t_{su}t_{sl}t_sE_{sl}} \quad (31)$$

$$\frac{d^2\sigma_{x-CFRP}}{dx^2} = \sigma_{x-CFRP} \frac{G_a}{t_a} \left(\frac{1}{t_{sl}E_{sl}} + \frac{1}{t_{CFRP}E_{CFRP}} \right) + \sigma_{x-su} \frac{t_{su}G_a}{t_{sl}t_{CFRP}t_aE_{sl}} - \sigma_0 \frac{G_a}{t_{sl}t_aE_{sl}} \quad (32)$$

The known parameters are the grouped into the following six constants

$$\lambda_1^2 = \frac{G_s}{t_s} \left(\frac{1}{t_{su}E_{su}} + \frac{1}{t_{sl}E_{sl}} \right) \quad (33)$$

$$\beta_1 = \frac{t_{CFRP}G_s}{t_{su}t_{sl}t_sE_{sl}} \quad (34)$$

$$C_1 = \sigma_0 \frac{t_{CFRP}G_s}{t_{su}t_{sl}t_sE_{sl}} \quad (35)$$

$$\lambda_2^2 = \frac{G_a}{t_a} \left(\frac{1}{t_{sl}E_{sl}} + \frac{1}{t_{CFRP}E_{CFRP}} \right) \quad (36)$$

$$\beta_2 = \frac{t_{su}G_a}{t_{sl}t_{CFRP}t_aE_{sl}} \quad (37)$$

$$C_2 = \sigma_0 \frac{G_a}{t_{sl}t_aE_{sl}} \quad (38)$$

The resulting governing equations are

$$\begin{cases} \frac{d^2\sigma_{x-su}}{dx^2} - \lambda_1^2\sigma_{x-su} - \beta_1\sigma_{x-CFRP} + C_1 = 0 \\ \frac{d^2\sigma_{x-CFRP}}{dx^2} - \lambda_2^2\sigma_{x-CFRP} - \beta_2\sigma_{x-su} + C_2 = 0 \end{cases} \quad (39)$$

Equations (39) are a system of coupled second-order linear ordinary differential equations, whose general solutions are

$$\sigma_{x-su}(x) = B_1 \cosh(A_1x) + B_2 \sinh(A_1x) + B_3 \cosh(A_2x) + B_4 \sinh(A_2x) + C \quad (40)$$

$$\begin{aligned} \sigma_{x-CFRP}(x) = \frac{1}{\beta_1} [(A_1^2 - \lambda_1^2)B_1 \cosh(A_1x) + (A_1^2 - \lambda_1^2)B_2 \sinh(A_1x) + (A_2^2 - \lambda_1^2)B_3 \cosh(A_2x) + \\ (A_2^2 - \lambda_1^2)B_4 \sinh(A_2x) - \lambda_1^2C + C_1] \end{aligned} \quad (41)$$

The known parameters A_1 and A_2 are defined as

$$A_1 = \sqrt{\frac{1}{2} [\lambda_1^2 + \lambda_2^2 + \sqrt{(\lambda_1^2 - \lambda_2^2)^2 + 4\beta_1\beta_2}]} \quad (42)$$

$$A_2 = \sqrt{\frac{1}{2} [\lambda_1^2 + \lambda_2^2 - \sqrt{(\lambda_1^2 - \lambda_2^2)^2 + 4\beta_1\beta_2}]} \quad (43)$$

$$C = \frac{\lambda_2^2 C_1 - \beta_1 C_2}{\lambda_2^2 \lambda_1^2 - \beta_1 \beta_2} \quad (44)$$

The terms B_1 to B_4 were calculated for the following boundary conditions

$$\begin{cases} x = 0 \rightarrow \sigma_{x-su} = 0; \sigma_{x-CFRP} = \sigma_0 \\ x = l \rightarrow \sigma_{x-su} = 0; \sigma_{x-CFRP} = \sigma_0 \end{cases} \quad (45)$$

leading to

$$\begin{cases} B_1 = \frac{-A_2^2 C + C_1 - \sigma_0 \beta_1}{A_2^2 - A_1^2} \\ B_2 = \left(\frac{A_2^2 C - C_1 + \sigma_0 \beta_1}{A_2^2 - A_1^2} \right) \frac{\cosh(A_1 l) - 1}{\sinh(A_1 l)} \\ B_3 = \frac{A_1^2 C - C_1 + \sigma_0 \beta_1}{A_2^2 - A_1^2} \\ B_4 = \left(\frac{A_1^2 C - C_1 + \sigma_0 \beta_1}{A_2^2 - A_1^2} \right) \frac{1 - \cosh(A_2 l)}{\sinh(A_2 l)} \end{cases} \quad (46)$$

The shear stresses in the sealant and adhesive can be calculated using equations (15) and (17)

$$\tau_{xz-s}(x) = t_{su} [B_1 A_1 \sinh(A_1 x) + B_2 A_1 \cosh(A_1 x) + B_3 A_2 \sinh(A_2 x) + B_4 A_2 \cosh(A_2 x)] \quad (47)$$

$$\tau_{xz-a}(x) =$$

$$\begin{aligned} & \frac{-t_{CFRP}}{\beta_1} [(A_1^2 - \lambda_1^2) B_1 A_1 \sinh(A_1 x) + (A_1^2 - \lambda_1^2) B_2 A_1 \cosh(A_1 x) + (A_2^2 - \lambda_1^2) B_3 A_2 \sinh(A_2 x) + \\ & (A_2^2 - \lambda_1^2) B_4 A_2 \cosh(A_2 x)] \end{aligned} \quad (48)$$

The displacement u_I is calculated by integration of ε_I as follows

$$\begin{aligned} u_{su}(x) = & \frac{1}{E_{su}} \int_0^x \sigma_{x-su} dx = \frac{1}{E_{su}} \left\{ \frac{B_1}{A_1} \sinh(A_1 x) + \frac{B_2}{A_1} [\cosh(A_1 x) - 1] + \frac{B_3}{A_2} \sinh(A_2 x) + \frac{B_4}{A_2} [\cosh(A_2 x) - \right. \\ & \left. 1] + Cx \right\} + \frac{\tau_{xz-a}(0)}{G_a} t_a + \frac{\tau_{xz-s}(0)}{G_s} t_s \end{aligned} \quad (49)$$

The other two displacements are calculated by using equations (19) and (20)

$$u_{sl} = t_s \left(\frac{u_{su}}{t_s} - \gamma_{xz-s} \right) \quad (50)$$

$$u_{CFRP} = t_a \left(\frac{u_{sl}}{t_a} - \gamma_{xz-a} \right) \quad (51)$$

The results provided in the next section are based on the actual thicknesses taken from microscopies of polished specimen sections, similar to the micrograph shown in Figure 9. The following thicknesses were

used: 0.125 mm for the CFRP plies; 0.06 mm for the TFB substrates; 0.08 mm for the TFB sealant and 0.09 mm for the adhesive film.

2.6 Results

Typical load displacement plots for the DCB tests are shown in Figure 14. The crack propagated through the carbon/epoxy laminate in a ‘stick-slip’ manner, the crack advancing at finite increments, and crossed the battery with an unstable propagation along the whole battery length. The initial slope of the curves obtained from the laminates with embedded TFB was slightly lower than for those without, because the specimens with the embedded battery were 1.9 mm narrower than the nominal width. As expected from its low fracture toughness (Table 2), the crack propagated through the muscovite substrate for all the three test repetitions, splitting one of the two substrates along a cleavage plane, whereas the bond between the substrate and the carbon/epoxy composite remained intact, Figure 15. The average G_{IC} was reduced 15% by the presence of the battery, which was equal to the amount of crack surface occupied by the TFB, Table 4. This proved that the crack propagated through the battery without releasing a significant amount of strain energy.

The uniaxial mechanical tests gave evidence that the strain measured at the surface of a TFB-CFRP laminate with an embedded battery decreased sharply at the center of the specimen, where the TFB was located. An example of a full-field strain plot measured experimentally using DIC on the cross-ply laminate is shown in Figure 16. The corresponding strain field calculated by FEA is plotted in Figure 17 using consistent contour levels for comparison. The correlation between experiments and analysis was assessed based on the strain distribution along the longitudinal center-line of the specimen, identified by line a-a in Figure 16. The experimental and calculated strain values along line a-a are plotted in Figure 18. The FEA values are averaged corner outputs. Furthermore, the strain level predicted by the simplified CLT method is plotted as a constant strain throughout the nominal battery length and as a constant far-field strain elsewhere. A strain concentration factor, defined as the ratio between the average strain over

the TFB ($\bar{\epsilon}_{TFB}$) and the far-field strain (ϵ_0), was calculated for each of these data sets. The summary of the strain concentration factors for the tested lay-ups is reported in Table 5, showing a good agreement between the experimental and calculated strain field for all the stacking sequences. The results seem to confirm that the TFB shared load with the laminate as desired and that its adhesion to the CFRP material remained intact within the 3000 μ strain range. Moreover, the assumptions of plane stress and uniform strain through-the-thickness appear valid to predict the overall elastic behavior of the laminate with an embedded battery.

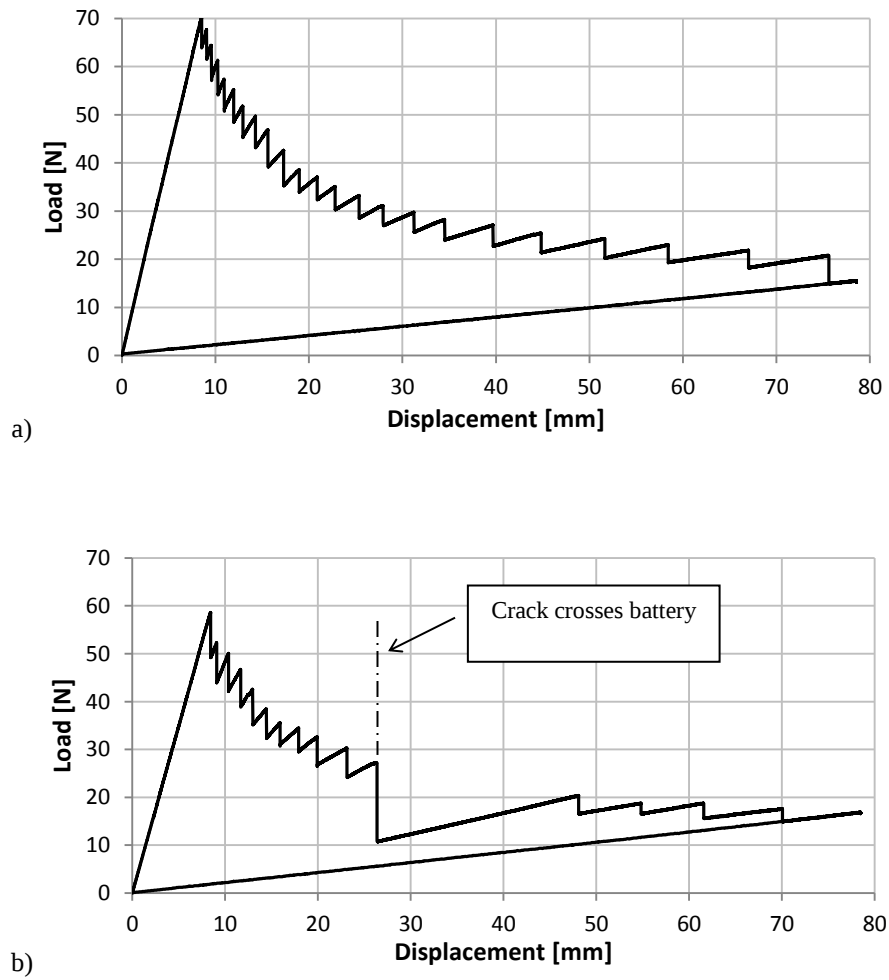


Figure 14. Typical load-displacement profiles for double cantilever beam test (a) without embedded TFB and (b) with embedded TFB.

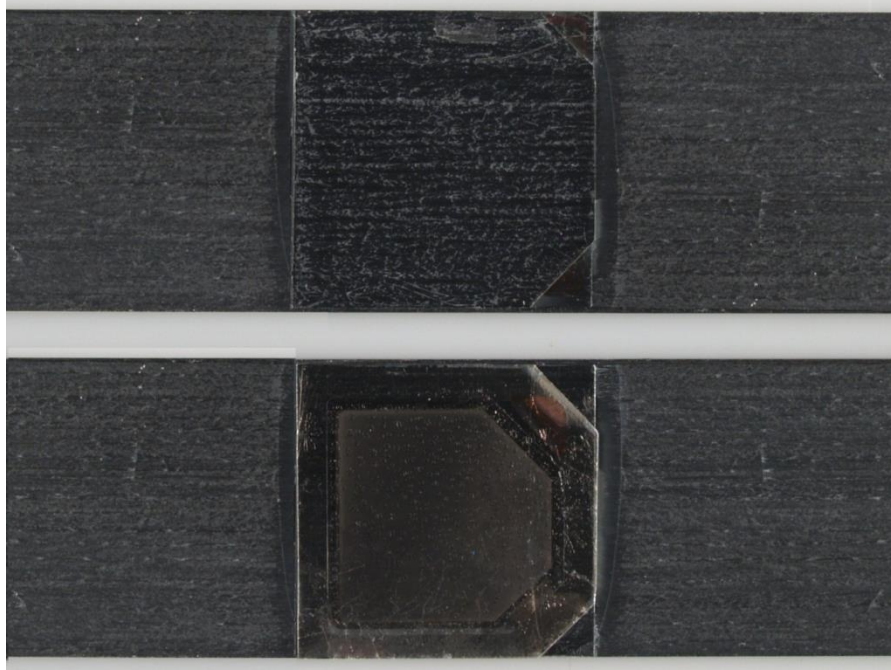


Figure 15. Close-up image of crack surfaces after double cantilever beam.

	Average G_{IC} [J/m ²]	σ [%]
Specimens without TFB	315.5	2.27
Specimens with TFB	267.0	1.55
Specimens with TFB - net crack surface ¹⁾	315.7	1.77
¹⁾ Crack surface of thin film battery (TFB) is excluded from G_{IC} calculation		

Table 4. Average mode I fracture toughness (G_{IC}) and standard deviation (σ) resulting from double cantilever beam (DCB) testing.

Within the limits of applicability of these assumptions, that is neglecting the three-dimensional stresses arising at lay-up transition, the interlaminar embedding did not lead to significant stress concentrations. In fact, the maximum stress concentration factor calculated with FEA was 1.07 and it occurred in the (0/90)_s laminate in proximity of the battery corners. This factor was calculated by dividing the maximum nodal strain in the x -direction by the far-field strain.

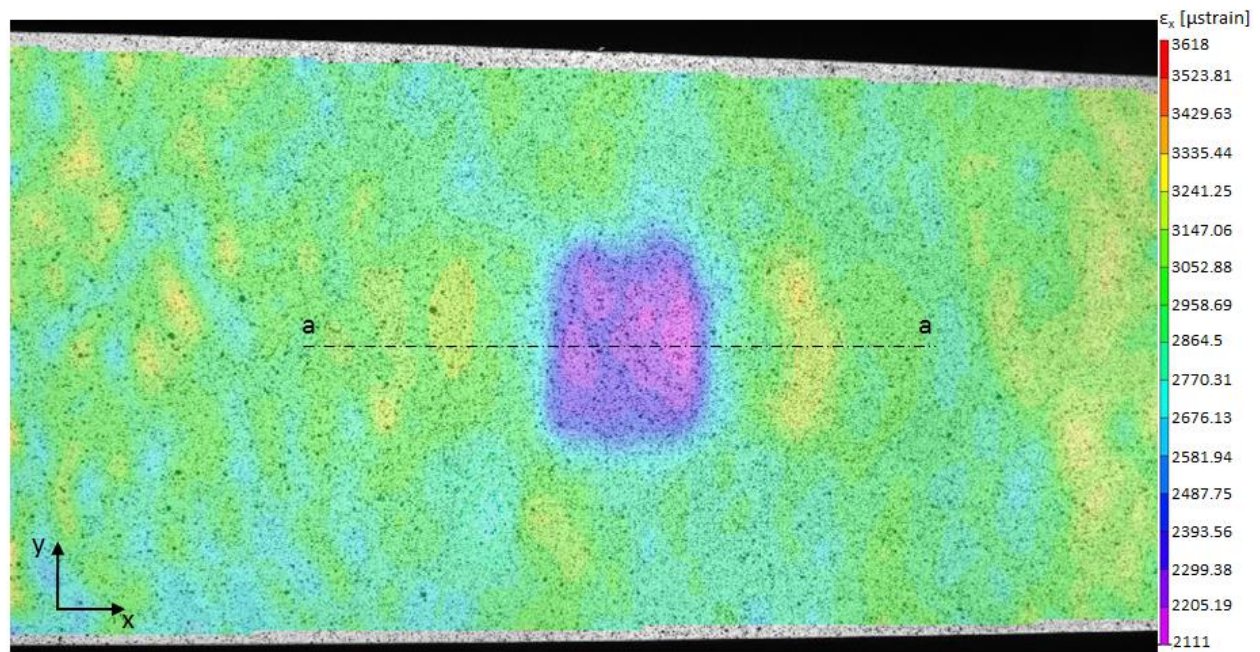


Figure 16. Uniaxial mechanical tension test. Normal strain distribution in the loading direction (ϵ_x). Laminate stacking sequence at battery is (0/90/TFB/90/0). Applied far-field strain of 3107 μstrain .

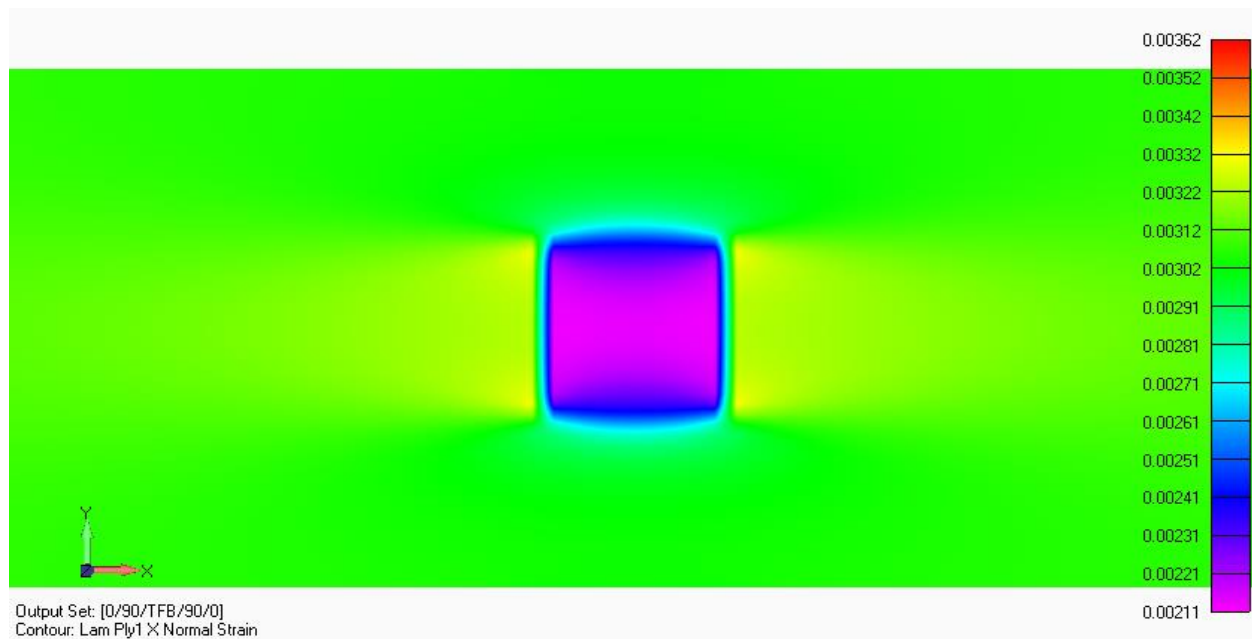


Figure 17. Uniaxial mechanical tension test simulation. Strain distribution in the loading direction (x) calculated with finite element analysis (FEA). Laminate stacking sequence at battery is (0/90/TFB/90/0). Applied far-field strain of 3107 μstrain .

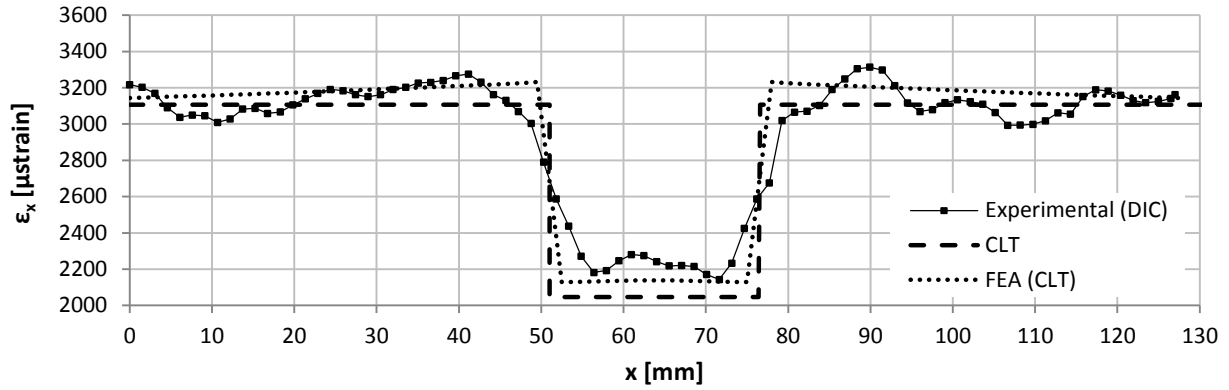


Figure 18. Measured and calculated ε_x strain distribution along line a-a shown in Figure 16. Laminate stacking sequence at battery is (0/90/TFB/90/0).

Lay-up	$\bar{\varepsilon}_{TFB}/\varepsilon_0$		
	Experimental (DIC)	CLT	FEA (CLT)
(0/90/TFB/90/0)	0.75	0.66	0.71
(0 ₂ /TFB/0 ₂)	0.84	0.79	0.81
(0/45/90/-45/TFB/-45/90/45/0)	0.80	0.74	0.80

Table 5. Uniaxial mechanical tension of interlaminar TFB embedding with full-field strain monitoring. Measured and calculated normal x-strain concentration factor along line a-a shown in Figure 16. Factor is defined as the ratio between average TFB strain $\bar{\varepsilon}_{TFB}$ and far-field strain ε_0 .

The same test procedure was adopted for the configuration with externally bonded TFB. The measured strain fields for the two tested lay-ups were characterized by higher strain gradients at the battery, in Figure 19. The highest gradients were located at the battery edges oriented transverse to the loading direction. These strain peaks were caused by a relative displacement between the upper substrate and the CFRP surface, which was detected as an apparent strain by the optical measurement instrumentation. Therefore, although this relative displacement was associated with a shear strain concentration in the sealant and in the adhesive, the normal strain peak had to be disregarded. The strain distribution over the battery, which coincides with the strain in the upper substrate, was in good agreement with the output of

the shear lag model, Figure 20. As predicted by the analysis, the normal strain of the upper TFB substrate, which was zero at the battery edges, increased progressively up to a maximum at the center of the battery, without reaching the strain level predicted by the CLT. The high modulus mismatch between the adherents (substrates and CFRP sub-laminate) and the adhesives (sealant and adhesive film), as well as the relatively high thickness of the adhesives, were the causes of this extensive shear lag behavior. The calculated normal strain distributions in the adherents clearly show that the TFB length was not enough to allow for a uniform strain through-the-thickness, Figure 21. As a result, the upper substrate was affected by shear lag for the entire battery length, while the lower substrate and the CFRP sub-laminate reached a uniform strain at about one-third of the battery length.

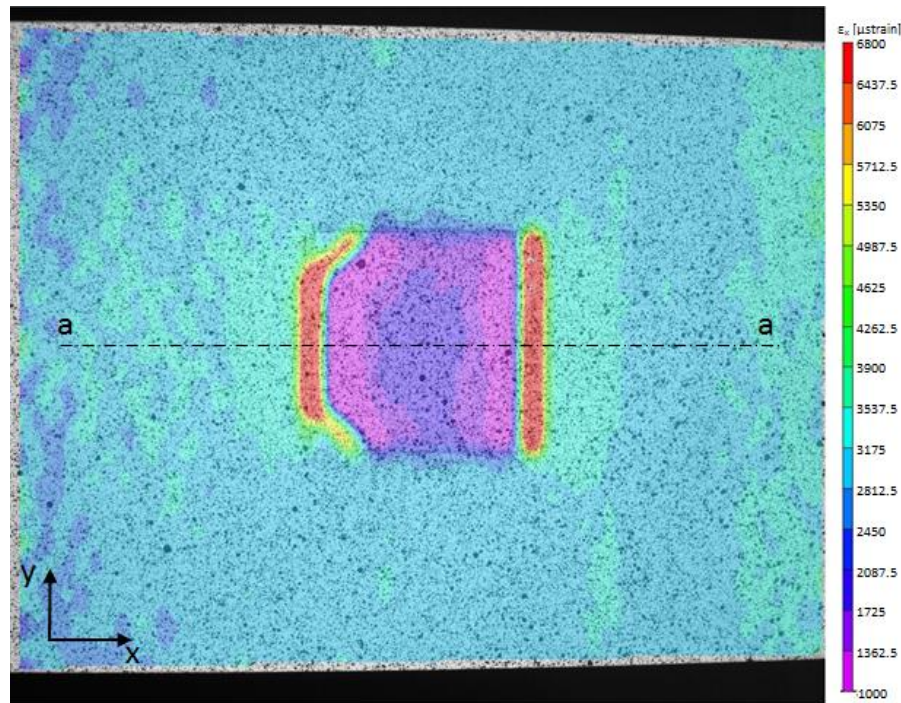


Figure 19. Uniaxial mechanical tension test. Normal strain distribution in the loading direction (ϵ_x). Laminate stacking sequence at battery is [(0/45/90/-45)_s/AF/TFB]. Applied far-field strain of 3123 μstrain .

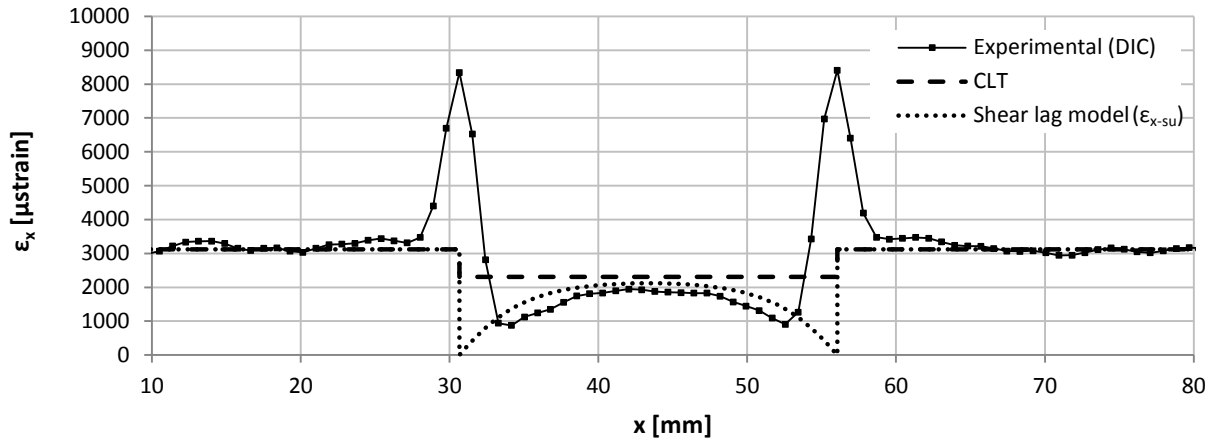


Figure 20. Measured and calculated ϵ_x strain distribution along line a-a shown in Figure 19. Laminate stacking sequence at battery is [(0/45/90/-45)_s/AF/TFB]. The x-coordinate defined in Figure 13 is offset in order to match the DIC coordinate system.

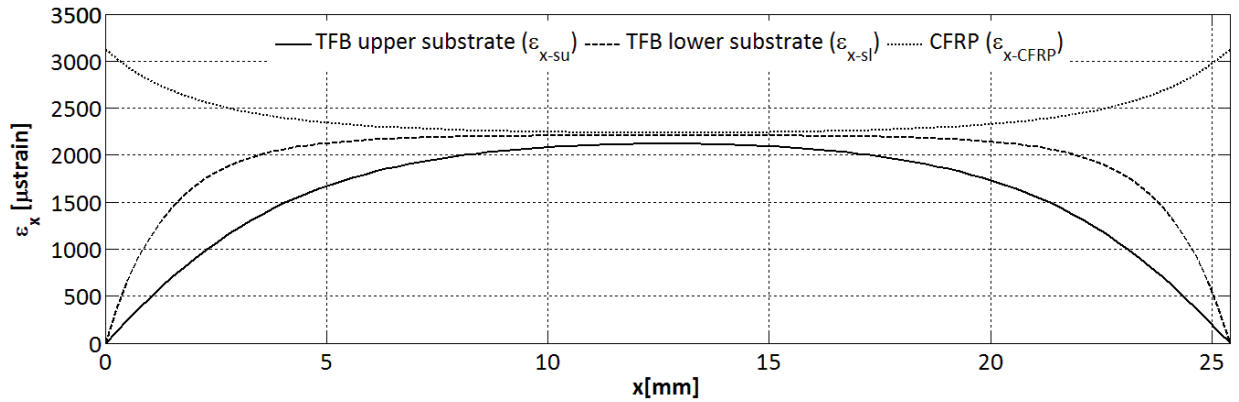


Figure 21. Normal strain distribution in CFRP laminate and TFB substrates calculated with the shear lag model. Laminate stacking sequence at battery is [(0/45/90/-45)_s/AF/TFB]. Applied far-field strain of 3123 μ strain.

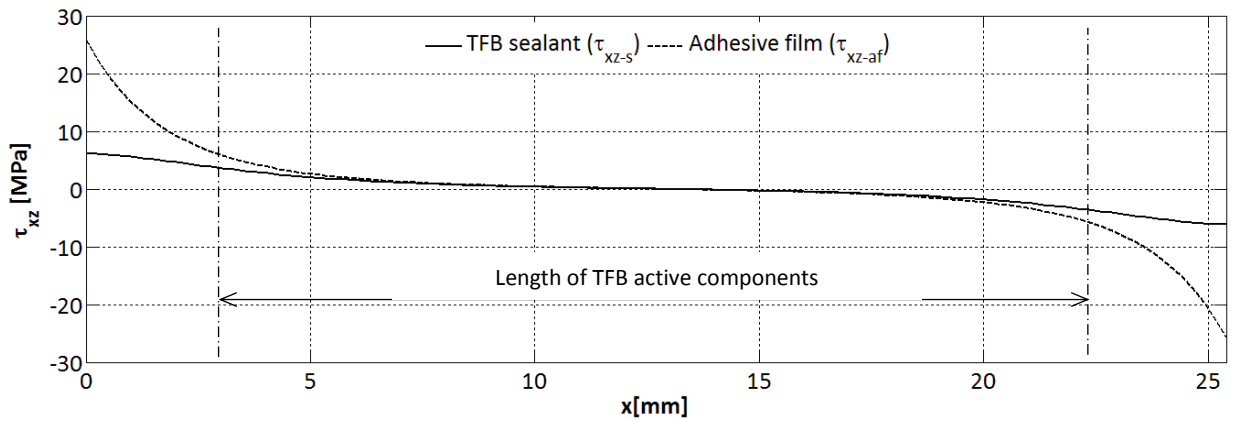


Figure 22. Shear stress distribution in adhesive film and sealant calculated with the shear lag model. Laminate stacking sequence at battery is [(0/45/90/-45)_s/AF/TFB]. Applied far-field strain of 3123 μ strain. The length occupied by the active components is indicated in the plot area.

According to the shear lag model, the uniform strain region, defined as the area where the strain in the upper substrate reached at least 95% of the strain in the CFRP sub-laminate, began at a distance of 13.3 mm and 12.8 mm from the TFB edges for the $[(0/45/90/-45)_S/AF/TFB]$ and $(0_8/AF/TFB)$ laminates respectively. Thus, in the shear lag region, which was as wide as 85 times the thickness of the battery and located next to the battery edge, the assumption of plane stress does not apply. For a sufficiently long TFB this edge effect would negligibly reduce the load sharing efficiency of the externally bonded battery, however the three-dimensional stress-strain field poses challenges for the mechanical integrity and it is a fundamental knowledge required for the design of the TFB-CFRP laminate. The shear stress peak in the sealant occurred at the battery edges, but the shear lag region extended well over the active components, Figure 22, and could trigger a mode II delamination onset at the edge of the active components, which is a known weak spot (Figure 6 and Figure 7).

Although the low sealant modulus was responsible for the extensive shear lag region, it had the advantage of leading to a low shear stress concentration factor, defined as the ratio between the peak stress and the average absolute value of the shear stress in the sealant layer. Shear stress concentration factors of 2.9 and 2.7 were calculated in the quasi-isotropic and in the unidirectional laminates, the latter being lower because the higher stiffness of the CFRP sub-laminate allowed a more gradual load transfer to the TFB. The shear stress concentration factors in the adhesive film, which had a higher modulus than the sealant, were 5.7 and 5.2 respectively.

The failure of the externally bonded battery configuration under uniaxial mechanical tension occurred at an average applied far-field strain of 4898 μ strain, Table 6. This value was less than 50% of the CFRP sub-laminate critical strain. The same failure mode occurred in both the lay-ups and is attributable to the failure of the lower substrate. Micrographs of the failed specimens demonstrated that the interface between the adhesive film and the substrate remained intact, while the failed substrate showed multiple delaminations along the cleavage planes in proximity of the interface with the adhesive film. The unstable growth of one of these delaminations caused the battery to suddenly detach from the CFRP sub-laminate,

leading to a brittle type of failure, Figure 23. The onset location of the failure can only be speculated. From the analysis of the calculated stress-strain fields it was noted that both specimens failed when the maximum shear stress occurring in the lower substrate reached 42.5 MPa, Table 6. Moreover, the TFB demonstrated during the electromechanical tension tests the capability to withstand higher normal strains than the ones calculated at failure for the externally bonded battery configuration and reported in Table 6. For these reasons a shear dominated failure starting from the substrate ends is deemed more likely than a failure caused by the peak normal stress occurring at substrate mid-span.

Lay-up	Strain at failure		Stress at failure	
	Far-field	Max. TFB	Max. TFB normal	Max. TFB shear
	ϵ_0 [μstrain]	$\epsilon_{x-sl\ max}$ [μstrain]	$\sigma_{x-sl\ max}$ [MPa]	$\tau_{xz-a\ max}$ [MPa]
[(0/45/90/-45) _s /AF/TFB]	5072	3588	639	42
(0 _g /AF/TFB)	4724	4050	721	43
Average	4898	3819	680	42.5

Table 6. Uniaxial mechanical tension of externally bonded TFB. Summary of critical stress-strain values at TFB mechanical failure. The far-field strain is experimentally measured, the other values are calculated.

The results for the battery thickness increase during charging are reported in Figure 24. The small displacements involved were comparable to the sensitivity of the measuring instrumentation, therefore the surface data was affected by a fairly high amount of noise, Figure 24-a. The imprint of the anode shape in the contoured data shows that the thickness increase was higher over the active components. In order to reduce the scatter, the average displacement of the area over the anode was calculated for every sampled displacement fields and plotted in Figure 24-b as a function of the charge level. The charge level was defined as a linear function of the battery voltage during charging: 0% corresponded to the initial charging voltage and 100% to the voltage at the end of the charging process (i.e. when the charge current dropped below 50 μA). The thickness increase at full charge resulting from regression analysis of the averaged displacement data was about 4 μm. An influence from temperature variability is excluded since no significant environmental changes or battery self-heating was detected by the IR camera during the

test. The perturbation introduced by a 4 μm battery cyclic expansion in terms of normal interlaminar stress and its long term effects on the integrity of the TFB-CFRP laminate are unknown at this stage of the research.

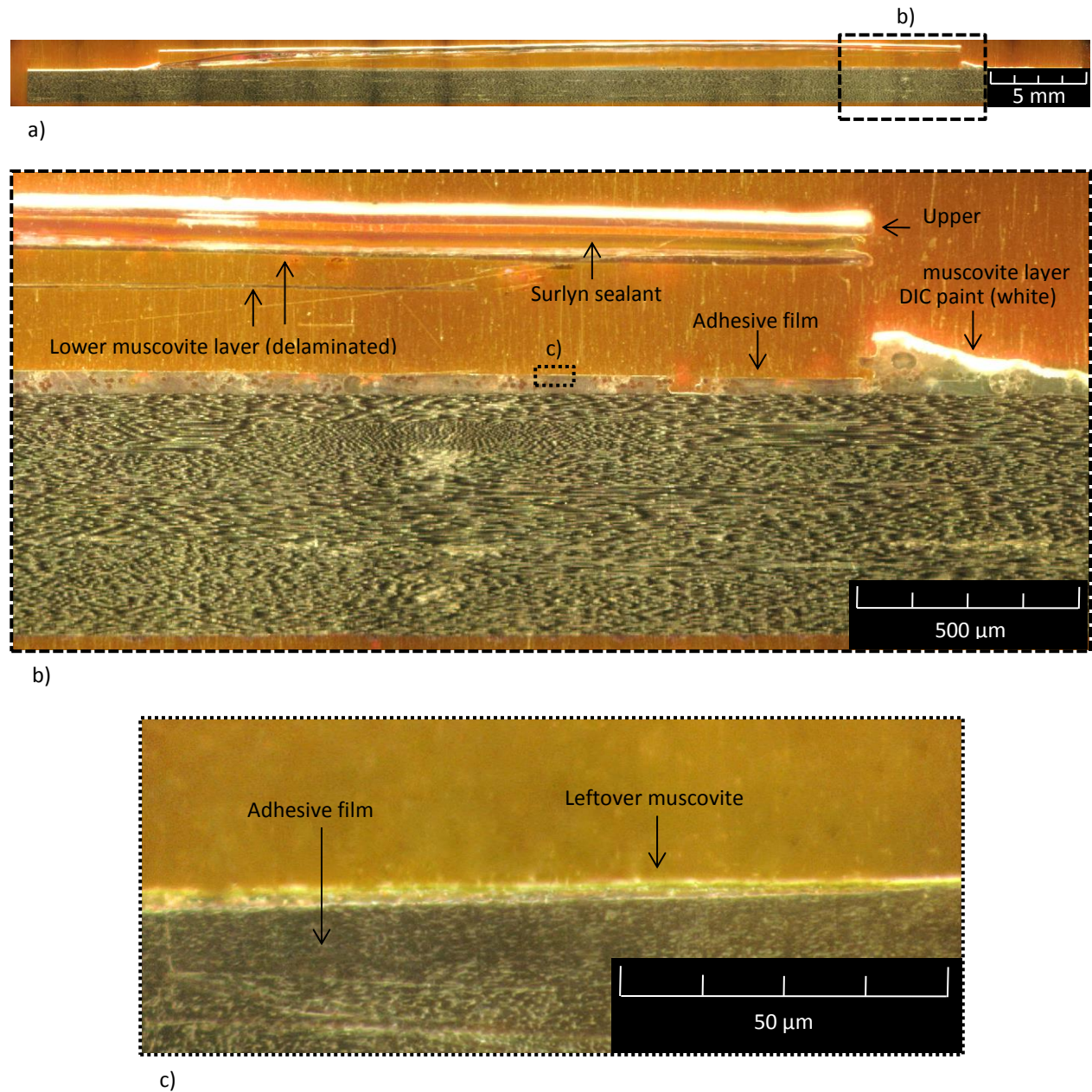


Figure 23. Micrographs of the uniaxial tension test specimen with externally bonded TFB after failure. Laminate stacking sequence is $(0_8/\text{AF}/\text{TFB})$. a) Full TFB section showing the battery lifted apart from the graphite laminate throughout the entire battery length. b) Multiple delaminations propagate through the TFB substrate. c) Adherent failure of the TFB substrate.

However, the high coefficient of expansion demonstrated by a single TFB cell suggests that the design of future stacked multicell laminates should account for the thickness variability.

The uniaxial tension tests with capacity monitoring confirmed that the battery electrochemical performance is unaffected by the applied strain, as reported by [21, 24, 25]. The sequence of capacity measurements under increasing strain for one of the two specimens tested is shown in Figure 25 and demonstrates that the ability of the battery to store energy remained constant up to the electrical failure. Also the current and voltage profiles were unchanged until electrical failure occurred. Failures consisted of an immediate and total loss of capacity that occurred as soon as a certain strain level was exceeded. The discharge voltage measured at the beginning of the strain interval was lower than 3 V, therefore the charge/discharge circuit recorded a zero discharge capacity and switched to the charging mode. Once in charging mode, the current saturated the power supply output and remained constant at 5 mA, similar to a short circuit. The charge capacity relevant to this type of failure is reported as infinite in Figure 25. Bringing the strain back to zero did not restore the battery functioning and failure was confirmed.

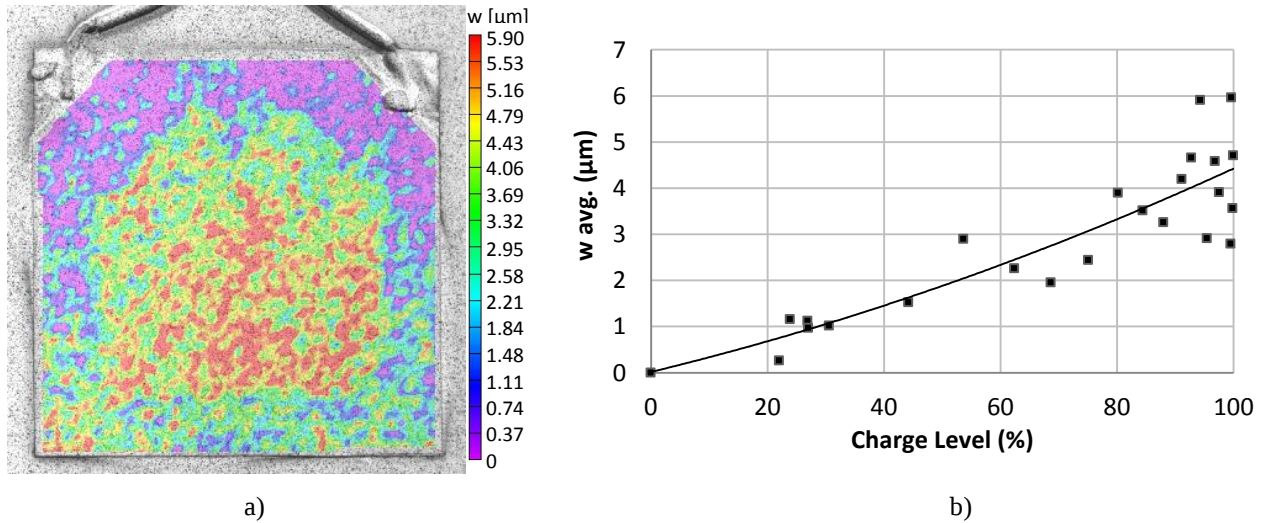


Figure 24. a) Anode side surface distribution of out-of-plane displacement w measured with digital image correlation (DIC) at 80% charge level. b) Displacement w averaged over the anode area and second order polynomial regression of the data points.

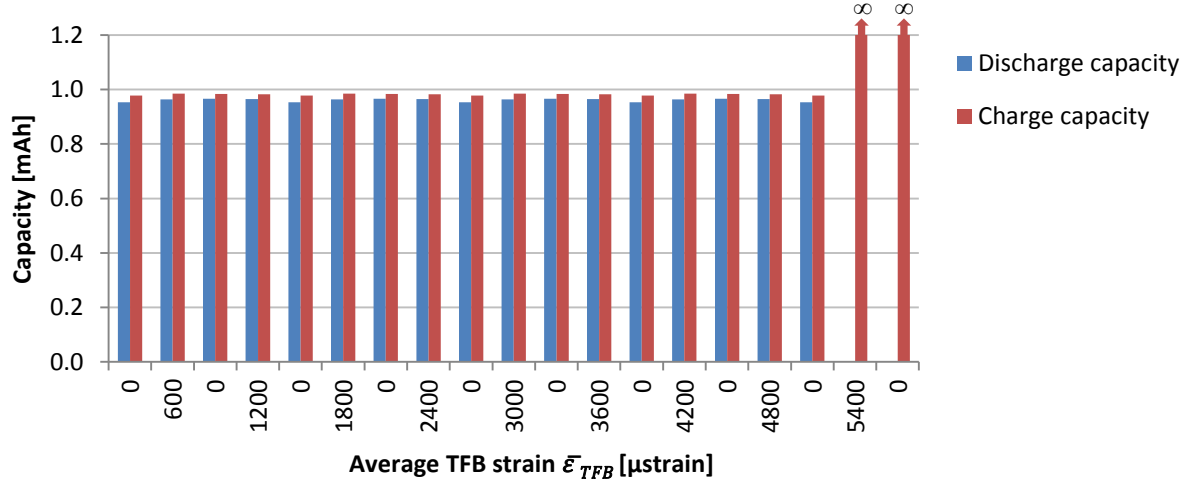


Figure 25. Electrochemical characterization of the TFB-CFRP laminate under uniaxial mechanical tension. Capacity measured at increasing strain cycles up to failure. Laminate stacking sequence is (0/45/90/-45/TFB/-45/90/45/0).

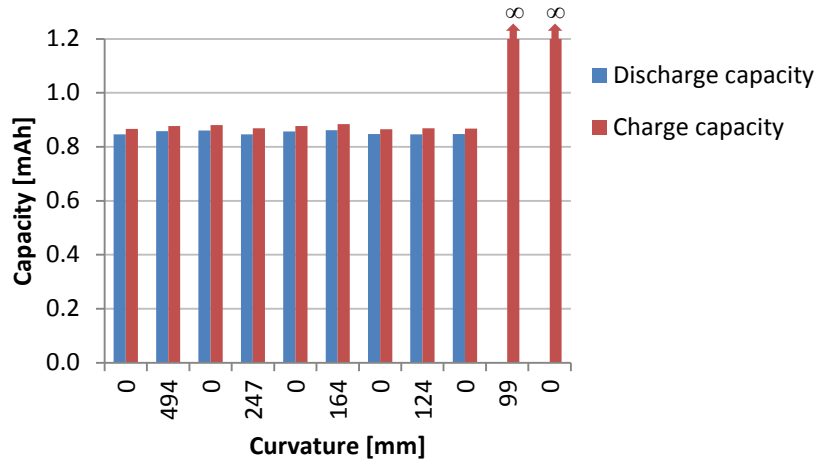


Figure 26. Electrochemical characterization of the TFB-CFRP laminate under single curvature. Capacity measured at increasing curvature cycles up to failure. Laminate stacking sequence is [0/90/TFB/90/0].

	Strain at electrical failure	
	ϵ_0 [μstrain]	$\bar{\epsilon}_{TFB}$ [μstrain]
Repetition 1	6000	4800
Repetition 2	6750	5400
Average	6375	5100

Table 7. Critical strain values for uniaxial mechanical tension with TFB electrical capacity monitoring of laminate [0/45/90/-45/TFB/-45/90/45/0]. Measured far-field strain ϵ_0 and corresponding calculated average TFB strain $\bar{\epsilon}_{TFB}$ at electromechanical failure.

The electrical failure occurred when the strain at the battery exceeded 5100 μ strain. The critical strains reported in Table 7 are relevant to the last strain cycle preceding failure, thus they are maximum operating strain values. The strain at the battery was calculated by multiplying the far-field strain by the 0.8 strain concentration factor determined through the mechanical tests and FEA described in the preceding section, Table 5. The causes of the failure are unknown since the battery was not accessible for inspection and ultrasonic inspections had not proven to be effective for this particular structure. The in-plane stiffness of the specimens along the testing direction did not change significantly after the electrical failure occurred. The results of the flexure tests showed that the applied curvature, similarly to the in-plane strain, does not affect the electrochemical performances up to failure. Results also gave evidence that the different combinations of strain and curvature lead to different failure modes. The specimens with an embedded battery at the laminate mid-plane and with a span-to-thickness ratio of 60:1 mechanically failed at a radius of curvature of 159 mm. The electrochemical performance remained constant up to the structural collapse of the laminate, which occurred by compressive failure of the upper plies and was not affected by the presence of the battery. This was the only loading condition for which the laminate structural strength was more critical than the battery integrity, thereby showing that pure curvature of the TFB is not critical for most structural applications. In order to measure the critical radius of curvature of the battery, the span-to-thickness ratio was increased to a non-standard ratio of 145:1. This test set-up allowed achievement of the electrical failures, which occurred at an average radius of curvature of 112 mm, without being anticipated by performance fading and with the same sudden loss of discharge capacity experienced during the uniaxial tension test, Figure 26.

For the combined curvature and in-plane strain loading conditions, electrical failures were obtained at the 60:1 span-to-thickness ratios. The critical radii of curvature and strains are summarized in Table 8. The compressive loading condition, with a low far-field strain at failure of 2832 μ strain, was particularly critical. Failure occurred by delamination buckling of the TFB packaging, Figure 27, and subsequent battery contamination. The failure initiation is uncertain. It can be attributed to an adhesive failure at the

sealant-substrate interface, or, given its low fracture toughness, to a delamination of the substrate. A cohesive failure of the sealant is deemed unlikely because of the relatively high fracture toughness of Surlyn. The buckling of the upper substrate, shown in a close-up view in Figure 28, opened a path for air and moisture ingress through the battery packaging and it was followed by a neutral grey discoloration of the anode occurring below the wrinkle of the substrate. This chromatic change was attributed to the reaction of lithium with contaminants. Like for the failure of the embedded battery, the discharge capacity measured after failure was considered zero because the discharge voltage was lower than 3 V. On the other hand, the charge current did not saturate the power supply as mentioned for the embedded battery, but it remained constant at about 0.45 mA and it showed a fairly high amount of random high frequency oscillations with approximately 50 μ A of amplitude. The charging process was declared failed after one hour.

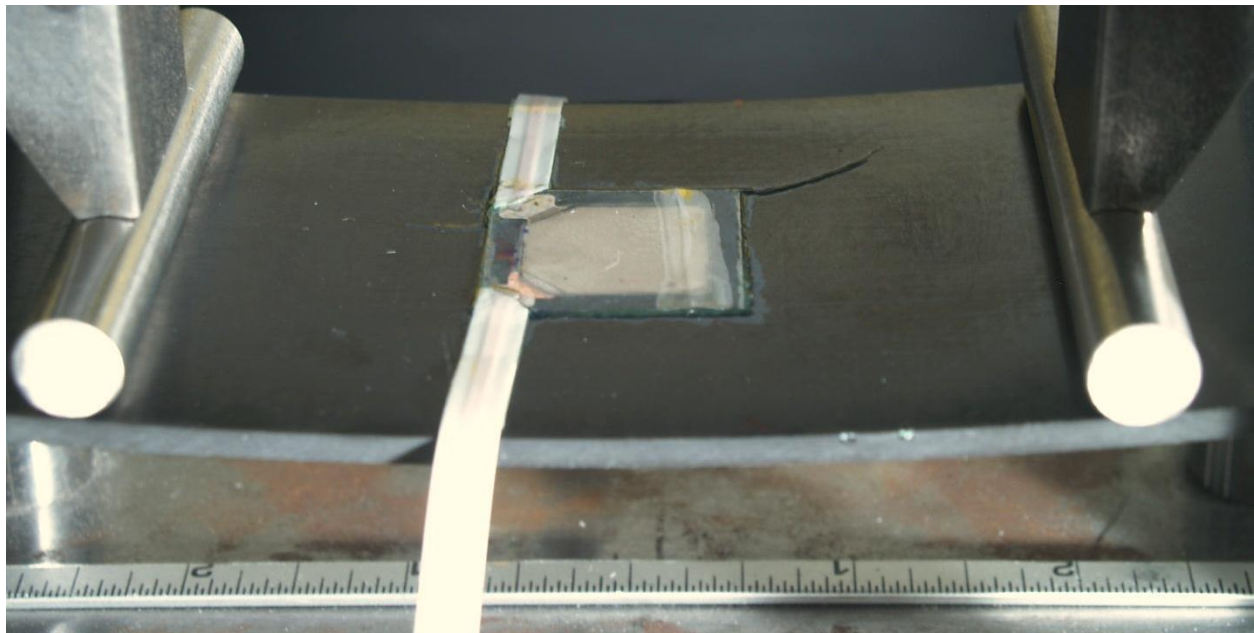


Figure 27. Failure of TFB undertaking electromechanical flexure testing. Delamination buckling of the battery at a radius of curvature of 504 mm and compressive strain of 2971 μ strain.

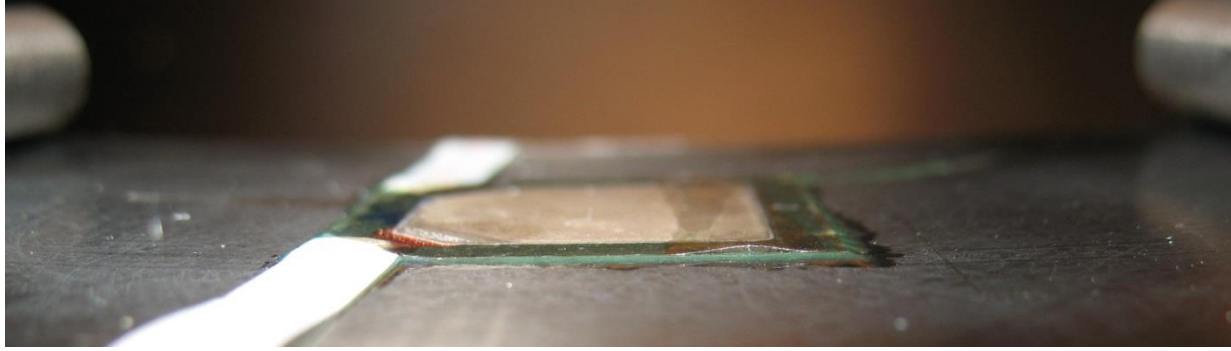


Figure 28. Failed battery shown in Figure 27 reveals air and moisture path through the buckled substrate.

		Radius of curvature [mm]	Far-field strain [μstrain]
Curvature	Repetition 1	124	0
	Repetition 2	99	0
	Average	112	0
Simultaneous curvature and negative in-plane strain	Repetition 1	504	-2971
	Repetition 2	557	-2692
	Average	531	-2832
Simultaneous curvature and positive in-plane strain	Repetition 1	204	7354
	Repetition 2	224	6683
	Average	214	7019

Table 8. Four point bending with TFB electrical capacity monitoring. Summary of critical radius of curvature and strain at electromechanical failure.

The specimens tested in curvature and positive strain failed at an average radius of curvature of 214 mm and an average far-field strain of 7019 μstrain. The failure mode consisted of brittle fracturing of the lower substrate and battery detachment from the CFRP sub-laminate, as described for the externally bonded battery configuration subjected to uniaxial tension. Although the failure modes for these two loading conditions appeared to be the same, the critical far-field surface strain of the laminates tested in flexure was 40% higher. The cause for the mismatch is partially attributed to an error associated with the calculation of the strain at failure for the curvature and positive in-plane strain loading condition. In fact, the deflections of the specimens at failure were in excess of 10% of the support span, leading to

geometrical nonlinearities. Therefore the small deformations assumption utilized for computing the strain provided overestimated values for this particular case.

2.7 Conclusions

The assumptions of plane stress and uniform normal strain through-the-thickness allowed the prediction of the overall elastic behavior of the hybrid thin film lithium ion – graphite battery laminates subjected to in-plane strain, for either the embedded battery or the externally bonded battery configuration. However, besides not being applicable at lay-up transition, these assumptions did not capture the extensive shear-lag that develops in proximity of the battery edges of the externally bonded battery configuration. Therefore three-dimensional FEA or higher order laminate, zig-zag, or layerwise theories are required for an optimized design.

The operational envelope for the hybrid laminate featuring the current thin film battery technology is reduced with respect to a conventional CFRP structural laminate. The failure of the TFB-CFRP laminate, defined as the mechanical failure of any laminate component or the electrical failure of the battery, occurred at an applied far-field strain of 4898 μ strain for uniaxial tension and 2832 μ strain for uniaxial compression in the worst case scenarios. The ability to tolerate curvature was equivalent to that of a conventional laminate as long as the battery was located at the laminate mid-plane and provided that the critical radius of curvature of 112 mm was not exceeded. Within these operational limits the electrochemical performance was not affected by the mechanical boundary conditions. Differences between the embedded battery and the externally bonded battery configuration were discussed in detail.

The critical failure modes of the externally bonded battery configuration included mode II dominated delamination or disbonding of the battery packaging when the laminate was subjected to mechanical tension, with failure onset at the battery edges or possibly at the edge of the active components. Delamination buckling of the battery upper substrate was critical under compressive loading. The packaging buckling failure triggered the electrical failure by cell contamination with air and moisture.

The delamination or cracking of the active components were not critical failure modes with the current packaging materials. Hence the battery packaging was the weakest link for the structural and electrical integrity.

The critical failure modes for the embedded battery configuration were not identified. The test campaign gave evidence that electrical failure is more critical than mechanical failure of the laminate for the in-plane strain loading condition. Battery substrate splitting with subsequent tearing of the active components, delamination of the packaging with cell contamination, or mechanical failure of the active components are all possible failure modes. The failure of the electrical connections is excluded because the battery discharge voltage remained detectable after failure. The same uncertainties apply to the electrical failure under curvature, which is, however, less critical than the mechanical failure of the laminate.

The low fracture toughness of the muscovite substrate is the most important limiting factor for the mechanical integrity of the current battery packaging. The high modulus mismatch between the Surlyn sealant and the substrate is the second most important limiting factor. The delamination of the substrate was responsible for the failure of the externally bonded battery under in-plane tension and is suspected to be the main cause for the delamination buckling that occurred under compression. The low fracture toughness of the substrate might have concealed other shortcomings such as poor mechanical strength of the electrodes and electrolyte and their bonding interfaces with the packaging, which were expected to be the critical for electromechanical survivability. A higher modulus sealant would increase the load sharing efficiency of the externally bonded battery and prevent compression buckling of the packaging. On the other hand, it would also increase the shear stress concentration factor at the battery edges, which promotes disbonding onset.

The interlaminar embedding of the battery did not have direct detrimental effects on the CFRP laminate elastic properties and mechanical strength. Perfect bonding and predictable load sharing with the composite structure up to failure were observed. Nevertheless the possible delamination onset and

propagation at lay-up transition requires further investigation. Moreover, accounting for the effects of the cyclic battery thickness variation upon the interlaminar normal stress and strain energy release rate is recommended for stacked multicell architectures. The stress induced by the battery thickness increase could be reduced by curing the laminates with fully charged batteries; however the study on manufacturability conducted by the authors revealed that thermal processing at full state of charge reduces the maximum curing temperature that the batteries can tolerate without permanent capacity loss. The design of TFB-CFRP laminates would benefit from the development of appropriate electrode materials that minimize volume variation during lithiation-delithiation.

The results demonstrate the importance of the packaging for the mechanical integration of batteries and structure. The laminate stress-strain field is highly influenced by the design of the substrate and sealant layer, as shown by the analysis of the uniaxial mechanical tension tests of the embedded and externally bonded battery configurations. Moreover, all the critical electromechanical failure modes that have been identified are initiated by the failure of a packaging component. For these reasons conventional packaging materials currently adopted by standard flexible electronics are inadequate for the multifunctional application. Lightweight aeronautical materials and high temperature structural composites developed for space application should be considered for their high elastic modulus and the ability to withstand severe stress and the high temperature associate with the manufacturing of the battery.

Based on the current performance, this multifunctional technology could be applied to a limited range of micro- and mini-UAV platforms, where the mechanical demands are minimal and the ratio of batteries to structural composites can be large enough to provide the required electrical energy and maximize the multifunctional efficiency. More demanding applications require a technological transition to improve the mechanical properties of the electrochemically active components and the packaging. In order to support the design of the next generation of airborne structural thin film batteries, modeling of the three-dimensional stress-strain field, delamination initiation and propagation are required to increase the mechanical strength and the electric survivability of the multifunctional system. The design should also

account for the constraints given by the physiochemical requirements of the TFB manufacturing process and by the composite laminate curing process. Finally, the density of the packaging materials should be minimized for lightweight design, whereas the current substrate material has a too high density of 2.6 g/cm³.

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3. Finite element analysis method

3.1 Summary

A novel automatic incremental solution algorithm for implicit nonlinear static finite element equations in the analysis of stable and unstable delamination propagation problems is presented. This load-displacement-constraint method iterates in the load displacement space utilizing the Newton-Raphson method, allowing to trace out the complete equilibrium path of the laminated structure. The procedure can calculate pre- and post-critical responses, ranging from stable delamination growth to structural collapse by unstable growth or delamination buckling with sharp snap-back instabilities. A direct application of the linear elastic fracture mechanics through the virtual crack closure technique is utilized to model failure. The method can be applied to bi-dimensional problems that exhibit geometric and material nonlinearities as well as large strains. The algorithm was implemented in Abaqus and examples illustrating the effectiveness of the proposed method were given.

3.2 Introduction

It is common practice to characterize the resistance to delamination of laminated fiber reinforced polymer matrix composites using fracture mechanics. A number of test methods have been developed for measuring the interlaminar fracture toughness (IFT) in terms of the critical value G_C of the total strain energy release rate G_T associated with delamination onset and growth. The peculiarity of laminated structures is that the delamination propagation occurs at bi-material interfaces, therefore G_C is a function of the fracture mode mixity.

The delamination propagation analysis requires the simultaneous solution of the fracture mechanics, the compliance change upon delamination propagation and typically should account for large displacements. For these reasons an incremental numerical method, such as nonlinear finite element analysis (FEA), is

often the only method of choice. Few exceptions are represented by simple and linear structures that can be solved with analytical methods.

Therefore the most general method currently available to approach the problem includes a nonlinear finite element solver coupled with a progressive failure model that simulates the damage propagation. Two classes of FEA methods have been proposed [1]. The first consists in the direct application of the linear elastic fracture mechanics (LEFM) and is applicable if the material nonlinearities occurring in the so called process zone around the crack tip can be neglected with respect to the global structural response. The delamination is propagated when the total strain energy release rate (ERR) equals a critical value, without modeling the process zone and therefore assuming that the fracture is perfectly brittle. The automated propagation occurs by removing a nodal constraint and allowing two coincident crack tip nodes to separate, causing to a sudden loss of adhesion and a finite delamination length propagation. The specific strain energy is therefore dissipated instantaneously in one single point. The computation of the ERR (or the stress intensity factor) can be performed with the virtual crack closure technique (VCCT) [2], the J-integral [3], the virtual crack extension method [4] or the stiffness derivative [5]. However, the automated delamination propagation analysis became possible thanks to the superior numerical and modeling efficiency of the VCCT over the other methods [6]. Before the use of the VCCT became well established, propagation analyses were performed by repeated analyses with constant delamination lengths. The second class of methods consists of modeling the process zone using damage mechanics and/or softening plasticity. This class of methods, initially proposed by [7], is called cohesive zone modeling (CZM) and it is usually applied in conjunction with interface elements [8]. The fracture mechanics is applied indirectly by dissipating the critical ERR over a finite delamination area. Unlike the first class of methods, G_C is not the only parameter that governs the damage progression, but other three parameters, namely the cohesive strength, cohesive stiffness and cohesive length, have to be defined. Since the relation between these parameters and the material properties is currently unknown, a cohesive law has to be calibrated on experimental results for a specific material [9]. In addition, in order to properly model the failure mechanism, a minimum number of elements in the process zone are required,

leading to a very fine mesh [10]. A mesh sensitivity analysis of the results is also required. On the other hand the VCCT cannot predict delamination initiation, but for cases where an initial delamination and a perfectly brittle failure can be assumed, it is the logical choice. In the present work we focused on the VCCT because the goal was to perform a parametric study on the elastic modulus and fracture toughness of a variety of possible TFB materials for a preliminary design. Furthermore, the experimental characterization of the CFRP-TFB laminate gave evidence of a brittle failure. Hence the characterization of the CFRP-TFB resistance to delamination, presented in the next chapter of the thesis, was based on LEFM.

In the finite element method the deformation of a given structure is described by a set of N degrees of freedom. If we assume that the loading is dependent on a single intensity parameter λ that controls the magnitude of all the applied loads, which are therefore proportionally varying, the applied load history is described by the scalar λ . In this context the load-deformation history of a structure presents itself as a curve in the $N+1$ dimensional space spanned by the degrees of freedom and the magnitude of the applied loads. Such a curve is usually referred to as the equilibrium path, Figure 1.

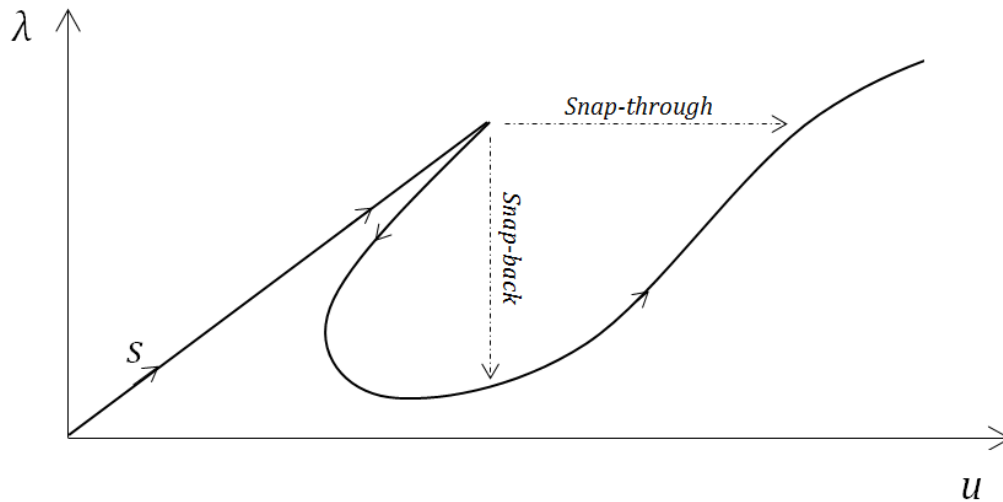


Figure 1. Equilibrium path for a bi-dimensional case with snap-back or snap-through instability.

The ductile or brittle nature of the failure depends on material properties, structure geometry, loading condition and external constraints. Using the π -theorem of dimensional analysis it was demonstrated that the transition from brittle to ductile behavior is governed by the brittleness number [11]

$$s_E = \frac{G_C}{\sigma_u b} \quad (1)$$

where σ_u is the material strength and b is a characteristic dimension of the structure. For a given structural problem, as s_E decreases the structural response upon failure becomes more brittle. As the brittleness increases, as shown in Figure 2 for a cracked beam loaded on a three point bending [12], the softening branch of the load-displacement assumes a positive slope. Both load and deflection must decrease to obtain a slow and controlled crack propagation, whereas in normal softening only the load decreases. Such a catastrophic event tends to reproduce the failure predicted by the LEFM because either the process zone or the slow crack growth are negligible before unstable crack propagation occurs and all the energy can be assumed to be dissipated on the crack surfaces. Hence initially uncracked specimens, large and/or slender structures with low fracture toughness and high tensile strength are associated with extremely brittle failure. Composite laminates are slender structures with large strength but relatively low fracture toughness; therefore they are prone to brittle behavior upon delamination. The TFB-CFRP laminate has even lower fracture toughness than conventional structural composites and the goal of parametric study was to assess its mechanical response against a wide range of fracture toughness values. Furthermore we were interested in the propagation characteristics of very short delaminations, because even if they were not a structural concern, they could potentially lead to a TFB contamination or electronic failure. The critical point at which the fracture starts to propagate tends to become a singular point of the equilibrium path where two branches having distinct tangents intersect. In [13] it was demonstrated that for an initially uncracked structure with the brittleness number tending to zero, this node becomes a cusp. In this limit case, the critical point is the intersection of a softening branch and an elastic branch that have the same tangent at intersection.

Therefore the problem of delamination propagation in structural composites is intimately connected with singularities that occur somewhere along the path under consideration. Depending on how the loading is applied, passing the critical point can result in snapping of the structure to a new equilibrium configuration that is not adjacent to the critical equilibrium. This is the case of unstable delamination growth. For example, if the softening branch has positive tangent and the structure is displacement controlled, the loading capacity presents a discontinuity with a negative jump, where the representative point drops on the lower branch. This type of instability is called snap-back. The complementary instability that occurs under load control is called snap-through and both of them are represented in Figure 1. When unstable delamination occurs, a portion of the equilibrium path becomes virtual.

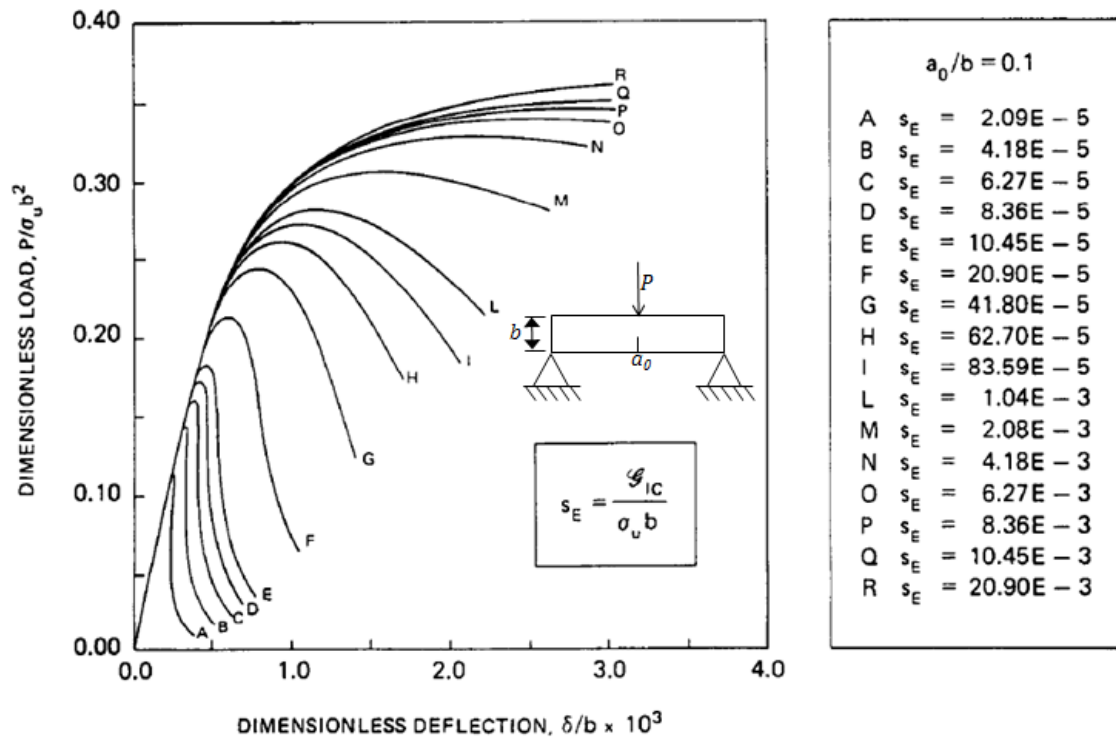


Figure 2. Load-deflection curve for a cracked beam loaded in three point bending. The ratio of initial crack length a_0 to the beam depth b is kept constant [12].

Hence, even if the composite is a linear elastic material and no elastic instabilities arise, snap-through and snap-back instabilities can occur because of the global softening associated to delamination propagation.

Snap-back instability is particularly common, even in simple structures. In fact it is one of the main reasons for the difficult standardization of the test methods for fracture toughness measurement [14], such as the end notch flexure (ENF) and the single leg bending (SLB). At a structural level, snap-back due to unstable delamination growth was documented in the analysis of a stiffened panel [15], and it should be expected as a possible failure mode in any aeronautical structure. Although the structural response described by the full equilibrium path, i.e. including the virtual parts, is somewhat artificial, the knowledge of such curve can be very important. In fact, once it is traced out, any dynamic “snap” associated with different initial crack lengths can be inferred without additional analysis.

Conventional FEA methods require prescribed load levels which are automatically changed a posteriori based on the convergence rate. Prescribing the load can be difficult without an approximate knowledge of the load carrying capacity of the structure. Moreover, these methods tend to crash in the proximity of a snap-back instability unless the structure is artificially stabilized. Such difficulties imply that a trial and error process is required to simulate the post collapse response, which is however not the equilibrium response, but rather the dynamic snapping. This leaves the analyst without information on the structural response associated with different initial delamination lengths. Because of the numerical challenge of simulating an unstable mechanical system, collapse loads are often associated with failure to achieve convergence with the iterative solution procedure. If the structural response is unknown a priori, it is uncertain whether the failure to converge is caused by a structural or a numerical collapse. This is an additional reason for which tracing the equilibrium path can be a useful preliminary analysis for a delamination problem.

3.3 The arc-length method

The same types of instabilities defined in the previous section occur in elastic buckling analysis, where critical points at which the Jacobian of the equilibrium path is undefined are also present. These singular points present themselves in the form of saddle nodes, horizontal or vertical tangent limit point, and bifurcation points. The latter, however, turns into a limit point when initial defects are introduced in the

structure. Therefore the structural response is, in general, smoother if compared to the sharp snap-back instabilities of the delamination failure.

The equilibrium path of elastic instability problems has been successfully traced with load-displacement-constraint procedures [16]. The basic idea behind these methods consists in iterating in the load-displacement space using Newton-Raphson (NR) methods, as it was proposed by a number of investigators, see for example[17][18][19]. The general method consisted in adding a control parameter to the NR method. The control parameter is the coordinate s that follows the path, Figure 1. This control parameter has the fundamental characteristic of being monotonically increasing with time. An auxiliary scalar equation constrains the norm of the incremental displacements and the load factor λ to follow a spherical or cylindrical path with radius Δl in the $N+1$ nondimensional space. The radius Δl approximates the arc-length as the increment size tends to zero. This constraint equation, which is enforced at every iteration and is not solved simultaneously to the N equilibrium equations, allows to treat the applied load as an additional variable, instead of being under user control as in conventional nonlinear FEA. The control parameter controls the progress of the computations along the path, thereby avoiding the high number of iterations and load cutbacks that automated incremental algorithms require while approaching a limit point. It also allows passing the limit point and tracing the equilibrium path in the post-critical region, for both snap-back and snap-through problems. The implementation of the arc-length method does not alter the symmetric banded nature of the set of equilibrium equations because the constraint equation is solved separately at the beginning of the iteration.

Let us describe in detail the implementation of the method. Assuming that the equilibrium state at time t is known, the constrained incremental procedure employed by an arc-length method to compute the equilibrium configuration at time $t + \Delta t$ is graphically represented in Figure 3 for a bi-dimensional case. Although in the figure a modified Newton-Raphson (MNR) solution method was arbitrarily selected, the full Newton-Raphson (FNR) or a quasi Newton-Raphson (QNR) solution method could be utilized.

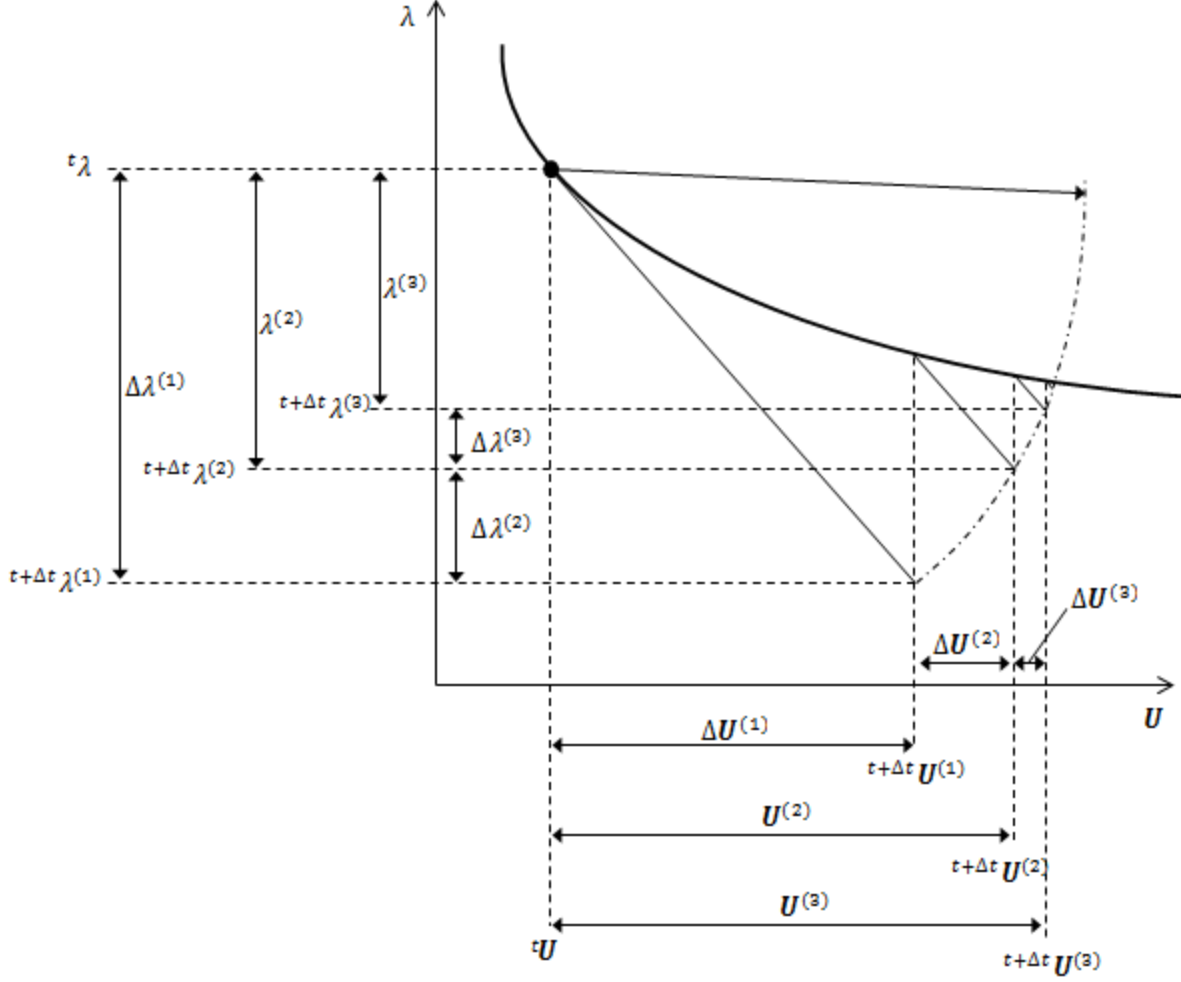


Figure 3. Schematics of the spherical arc-length method in a bi-dimensional problem.

The load factor ${}^{t+\Delta t}\lambda^{(i)}$ and displacement vector ${}^{t+\Delta t}\mathbf{U}^{(i)}$ at increment i , and their relation to the corrections to the load factor $\Delta\lambda^{(k)}$ and to the displacement $\Delta\mathbf{U}^{(k)}$ are defined as

$${}^{t+\Delta t}\lambda^{(i)} = {}^t\lambda + \lambda^{(i)}, \quad \lambda^{(i)} = \sum_{k=1}^i \Delta\lambda^{(k)} \quad (2)$$

$${}^{t+\Delta t}\mathbf{U}^{(i)} = {}^t\mathbf{U} + \mathbf{U}^{(i)}, \quad \mathbf{U}^{(i)} = \sum_{k=1}^i \Delta\mathbf{U}^{(k)} \quad (3)$$

The relation that constrains the load factor and the displacement in the $N+1$ dimensional space for the spherical method is

$$(\lambda^{(i)})^2 + (\mathbf{U}^{(i)})^T \mathbf{U}^{(i)} / \beta = \Delta l^2 \quad (4)$$

where β is a normalizing factor (to render the term dimensionless). Equation (4) was proposed by [17] without the normalizing factor and was utilized in [18]. In [16] the normalizing factor was introduced. Better results in terms of convergence rate and robustness were obtained by [19] using the cylindrical constraint equation

$$(\mathbf{U}^{(t)})^T \mathbf{U}^{(t)} = \Delta l^2 \quad (5)$$

Let us solve step-by-step the iterative procedure that computes the equilibrium configuration at time $t + \Delta t$.

The initial out-of-balance load vector is

$$-g({}^{t+\Delta t}\lambda^{(1)}) = {}^{t+\Delta t}\lambda^{(1)} \mathbf{R} - {}^t\mathbf{F} \quad (6)$$

The difference between the externally applied nodal forces at iteration (1), ${}^{t+\Delta t}\lambda^{(1)} \mathbf{R}$, and the vector of internal nodal forces at the end of increment t , ${}^t\mathbf{F}$, can also be written as a function of the load factor correction

$$-g({}^{t+\Delta t}\lambda^{(1)}) = \Delta\lambda^{(1)} \mathbf{R} \quad (7)$$

Therefore the system of equilibrium equations to be solved becomes

$${}^\tau \mathbf{K}^{(0)} \Delta \mathbf{U}^{(1)} = \Delta\lambda^{(1)} \mathbf{R} \quad (8)$$

In the above equation an iteration superscript for the tangent stiffness matrix was specified, as the matrix could be updated according to the FNR or a QNR method. In case the MNR method is utilized, the superscript (i) at the tangent stiffness matrix should be dropped. The time superscript τ would be equal to $t + \Delta t$ for the FNR and the QNR method, whereas for the MNR it would be equal to t .

The displacement corrections for iteration (1) are then

$$\mathbf{U}^{(1)} = \Delta\lambda^{(1)} \mathbf{U}_T^{(1)} \quad (9)$$

The vector $\mathbf{U}_T^{(1)}$ is the tangential displacement and is equal to $({}^\tau \mathbf{K}^{(0)})^{-1} \mathbf{R}$.

Note that the displacement correction $\Delta \mathbf{U}^{(1)}$ equals the displacement increment $\mathbf{U}^{(1)}$.

The initial load factor $\Delta\lambda^{(1)}$ for the first increment is usually specified by the user. In that case the arc-length Δl is computed by substituting equation (9) into (5)

$$\Delta l = \Delta\lambda^{(1)} \sqrt{\left(\mathbf{U}_T^{(1)}\right)^T \mathbf{U}_T^{(1)}} \quad (10)$$

Different formulations of the method have been proposed where, for example, instead of the load increment $\Delta\lambda^{(1)}$, a nodal displacement is specified by the user [18]. The arc-length Δl can also be directly specified by the user instead of the load factor; however physical parameters such as the load factor or a nodal displacement are preferred because it is easier for the user to estimate a reasonable value without knowing a priori the structural response.

The out-of-balance force vector at the beginning of the subsequent iteration (iteration 2) is

$$-g\left({}^{t+\Delta t}\lambda^{(2)}\right) = {}^{t+\Delta t}\lambda^{(1)}\mathbf{R} - {}^{t+\Delta t}\mathbf{F}^{(1)} + \Delta\lambda^{(2)}\mathbf{R} \quad (11)$$

which leads to the following displacement correction

$$\Delta\mathbf{U}^{(2)} = \left({}^t\mathbf{K}^{(1)}\right)^{-1} \left({}^{t+\Delta t}\lambda^{(1)}\mathbf{R} - {}^{t+\Delta t}\mathbf{F}^{(1)}\right) + \Delta\lambda^{(2)} \left({}^t\mathbf{K}^{(1)}\right)^{-1} \mathbf{R} \quad (12)$$

The first member of the addition at the right hand side of the equation is a known set of equations that can be solved with conventional finite element procedures. The second member features the tangential displacement

$$\mathbf{U}_T^{(2)} = \left({}^t\mathbf{K}^{(1)}\right)^{-1} \mathbf{R} \quad (13)$$

which has already been solved at iteration (1) if the MNR method is used, or has to be solved at each iteration with conventional finite element procedures if the FNR or a QNR method is employed. The correction $\Delta\lambda^{(2)}$ is then computed by imposing the constraint relation (4) or (5). Using for example (5), by substituting equation (12) into the constrain equation, which has to be written as a function of the displacement correction as follows

$$\left(\mathbf{U}^{(1)} + \Delta\mathbf{U}^{(2)}\right)^T \left(\mathbf{U}^{(1)} + \Delta\mathbf{U}^{(2)}\right) = \Delta l^2 \quad (14)$$

gives a scalar quadratic equation in the unknown $\Delta\lambda^{(2)}$. After solving for $\Delta\lambda^{(2)}$ the displacement correction are computed through equation (12).

The subsequent iterations are conducted by following the same procedure, which consist of updating the iteration superscript of equations (11), (12), (13) and (14).

Hence, the i^{th} iteration is solved by decomposing the displacement correction as

$$\Delta \mathbf{U}^{(i)} = \left({}^t\mathbf{K}^{(i-1)} \right)^{-1} \left({}^{t+\Delta t}\boldsymbol{\lambda}^{(i-1)} \mathbf{R} - {}^{t+\Delta t}\mathbf{F}^{(i-1)} \right) + \Delta \lambda^{(i)} \left({}^t\mathbf{K}^{(i-1)} \right)^{-1} \mathbf{R} \quad (15)$$

and by calculating the load factor correction $\Delta \lambda^{(i)}$ as described above, using the constrain equation

$$\left(\mathbf{U}^{(i-1)} + \Delta \mathbf{U}^{(i)} \right)^T \left(\mathbf{U}^{(i-1)} + \Delta \mathbf{U}^{(i)} \right) = \Delta l^2. \quad (16)$$

Note that $\Delta \lambda^{(i)}$ can be positive or negative. Moreover, two values of $\Delta \lambda^{(i)}$ are computed because the constraint equation is quadratic in the displacement correction $\Delta \mathbf{U}^{(i)}$. In order to select the correct root, several techniques were adopted by different authors. In general, if the equilibrium path is characterized by smooth limit points, the method consists in selecting the root that leads to the minimum angle between the new displacement increment vector and the displacement increment vector computed at the previous increment. In other words, the root that leads to the maximum value of γ should be selected [18], where γ is scalar product of two subsequent displacement increment vectors

$$\gamma = \left(\mathbf{U}^{(i-1)} \right)^T \mathbf{U}^{(i)} \quad (17)$$

When convergence is achieved at time t , the arc-length value has to be updated for the subsequent increment. Before updating the arc-length value, additional checks on the converged iteration can be introduced to make sure that the calculated equilibrium configuration is not too distant from the previous equilibrium configuration, thereby ensuring detailing of the equilibrium path. In [18], for example, an additional constraint equation required the norm of the displacement increment at converged iteration to be less than a multiple of the displacement increment computed at time 0

$$\| {}^t\mathbf{U} \| \leq \alpha \| {}^0\mathbf{U} \| \quad (18)$$

This condition was used in an algorithm that required the user to specify a nodal displacement as initial load control parameter, thereby relying on displacement based inputs more so that force based inputs from the user.

If such constraint is not violated, the equilibrium configuration is accepted and the solution can proceed to the next increment. The arc-length value is updated based on convergence rate using the following basic relation [19]

$$\Delta l_{new} = \sqrt{\frac{N_d}{N_t}} \Delta l_{old} \quad (19)$$

where N_d is the desired number of iterations, whereas N_t is the number of iterations that were necessary to find a converged equilibrium at time t . The new arc-length value could also depend on the desired displacement increment, as proposed in [18]

$$\Delta l_{new} = \sqrt{\frac{N_d}{N_t}} \frac{\alpha \| {}^0\mathbf{u} \|}{\| {}^t\mathbf{u} \|} \Delta l_{old} \quad (20)$$

If condition (18) is not verified, the equilibrium configuration is discarded and the solution restarts from the end of increment $t - \Delta t$ with a new arc-length value computed as follows

$$\Delta l_{new} = \frac{\alpha \| {}^0\mathbf{u} \|}{\| {}^t\mathbf{u} \|} \Delta l_{old} \quad (21)$$

The arc length is therefore scaled linearly based on the desired convergence rate. Its value is not specified by the user then, including the first increment where the arc length is calculated based on the load increment $\Delta\lambda^{(0)}$, which is the only user input. For any other increment other than the first, the initial load factor increment is computed at the beginning of the increment as

$$\Delta\lambda^{(1)} = \pm \Delta l / \sqrt{(\mathbf{u}_T^{(1)})^T \mathbf{u}_T^{(1)}} \quad (22)$$

where the sign follows that of the previous increment unless the determinant of the tangent stiffness matrix has changed sign, in which case, a sign reversal is applied.

This load-displacement-constraint procedure enabled to successfully trace very elegantly the equilibrium path of problems with relatively smooth structural response, such as snapping and buckling of thin shells. However, it was shown in [20][9][1] that this method failed to compute the correct advancement direction along the equilibrium path if sharp snap-back points were present, as in softening solids due to crack propagation. In these case the root selection criteria has to be tailored on the specific problem. In [20] an

algorithm was proposed that computed the solution relevant to the advancement direction of both the roots, and then selected the one that led to the minimum residuals. The effectiveness of the method was demonstrated also in [1]. Besides stability and correctness of the solution, also the computational efficiency of the arc-length method is degraded by sharp snap-back instabilities, because of the drastic decrements of the arc-length in proximity of the critical point, as shown in [21] and [20].

Other authors documented the lack of robustness of the conventional arc-length method when applied to crack propagation problems and attributed this shortcoming to the fact that the failure that led to the singularity was localized. For this reason only a few nodes contribute to the change in the norm of displacement increments, thereby providing a weak sensitivity of the method to crack advance. More robust algorithms were developed by constraining only a local set of degrees of freedom. In [22], for example, a cohesive FEA model of a 3PB concrete beam with a crack was solved using a cylindrical arc length method to constraint only the degrees of freedom that described the crack mouth opening. The conventional cylindrical constraint relation (5), or its linearized version called updated normal path method

$$(\mathbf{U}^{(i)})^T \mathbf{U}^{(i-1)} = \Delta l^2 \quad (23)$$

failed to trace the post-critical equilibrium path. By using only a function of few degrees of freedom representing the crack mouth opening

$$(\mathbf{u}^{(i)})^T \mathbf{u}^{(i-1)} = \Delta l^2 \quad (24)$$

The authors were able to compute the post-critical response. Similar methods were also successfully adopted in [23][24][25]. All of them adopted CZM.

Particularly challenging is the case of simultaneous occurrence of delamination and local laminate buckling, known as delamination buckling, because of the very sharp snap-back structural response resulting from the sudden section loss. In [9] it was documented that the arc-length method failed to solve this problem and a dynamic analysis with implicit time integration, a sort of combined static-dynamic analysis as defined by [1], was necessary to solve the problem. This technique however implies that the

damping coefficients of the structure have to be measured or assumed, and then a sensitivity analysis has to be performed to make sure that damping does not affect the structural response and crack propagation. In conclusion, the review of the literature showed that the arc-length method can be modified to solve a particular delamination propagation problem using CZM. However, by modifying the arc-length method to cope with crack propagation problems, the constraint equation becomes problem dependent. As a consequence the method loses some of its generality and elegance.

3.4 On the use of the virtual crack closure technique (VCCT) in conjunction with the arc-length method

Several commercial geometric nonlinear finite element codes allow to simulate delamination propagation through LEFM by using the VCCT. However, the simulation of post-critical unstable delamination growth with the current techniques is unduly expensive because of the repeated load cutbacks that occur at any delamination advancement through the fine mesh. In addition, only the dynamic “snapping” of the structure can be predicted, with all the well-known challenges associated with the convergence of the implicit finite element solution attempting to simulate an unstable mechanical system [26][27]. In order to trace also the virtual branches of the equilibrium path, the VCCT could be coupled with the arc-length method [1]. When the arc-length method is utilized together with a VCCT modeling of the delamination propagation, the modifications to the conventional arc-length method described above are also necessary to overcome random snap-backs that are not due to singularities in the response of the physical system, but are due to spurious oscillation in the finite element model response. These oscillations occur when G_T exceeds the critical value and a couple of coincident crack tip nodes are released in order to allow the delamination to advance by a finite amount, which coincided with the element length. At the iteration that corresponds to this event the force imbalance in the model suddenly increase because of the deletion of the large internal forces that constrained the crack tip modes to remain coincident. This local discontinuity in the out-of-balance forces is so severe that several iterations are required in order to reduce the force residuals within the convergence threshold, Figure 4. For this reason, for a given structural problem, the

number of iterations increases exponentially with the mesh density of the interface undertaking delamination. The sudden softening response that occurs at delamination advancement leads to a localized snap back in the computed equilibrium path [1], which is characterized by spurious oscillations, Figure 5. Depending on the software, single or multiple advancements can occur in an increment. Abaqus allows the user to choose between the two techniques.

In the first case the increment size is cutback if the ratio of total and critical ERR exceeds a release tolerance r_{tol} . The ERR convergence criterion to be satisfied is

$$\frac{G_T}{G_C} \leq 1 + r_{tol} \quad (25)$$

If the condition not satisfied the result of the increment is discarded and the increment is restarted from the beginning with a size reduction. In other words the load increment is cutback.

The delamination is propagated when

$$1 \leq \frac{G_T}{G_C} \leq 1 + r_{tol} \quad (26)$$

The release tolerance r_{tol} has to be set small enough to ensure solution accuracy and avoid errors that can accumulate and lead to gross and undetectable errors and even to solution instabilities.

In the second case, the user can decide to allow multiple nodes to be released within the increment, until relation (25) is satisfied. The nodes are then released one by one and equilibrium has to be achieved after each node is released. This implies that one or more iterations are required at any delamination advancement. However, if convergence cannot be achieved according to the automatic increment control scheme settings (i.e. if the maximum number of crack extension attempts is achieved for example), the increment is eventually cutback. The increment reduction is established automatically based on the Abaqus increment control scheme settings [28].

This procedure is particularly inefficient in case of fine meshes and unstable delamination propagation. When a snap-back instability occurs, for example, a large number of increment cutbacks are required in order to reduce the increment to a size that is small enough to capture the vertical load drop.

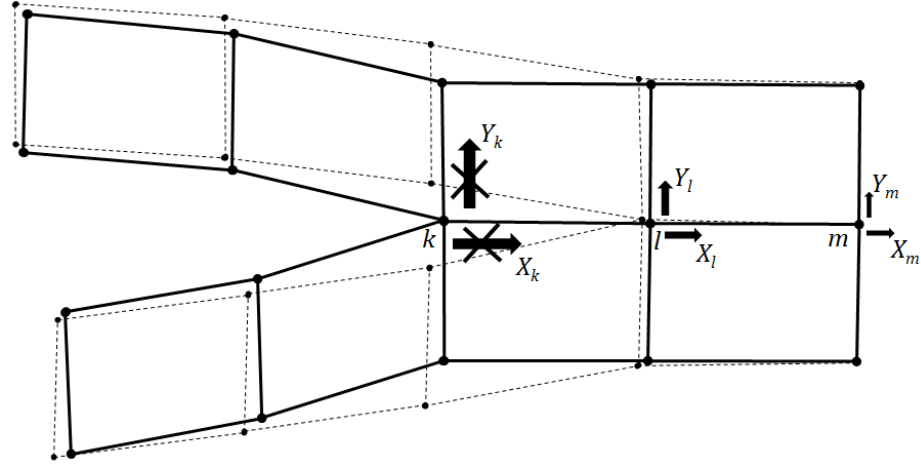


Figure 4. Deletion of MPC forces at crack tip node k (solid lines) and subsequent softening achieved at the end of the increment (dotted lines).

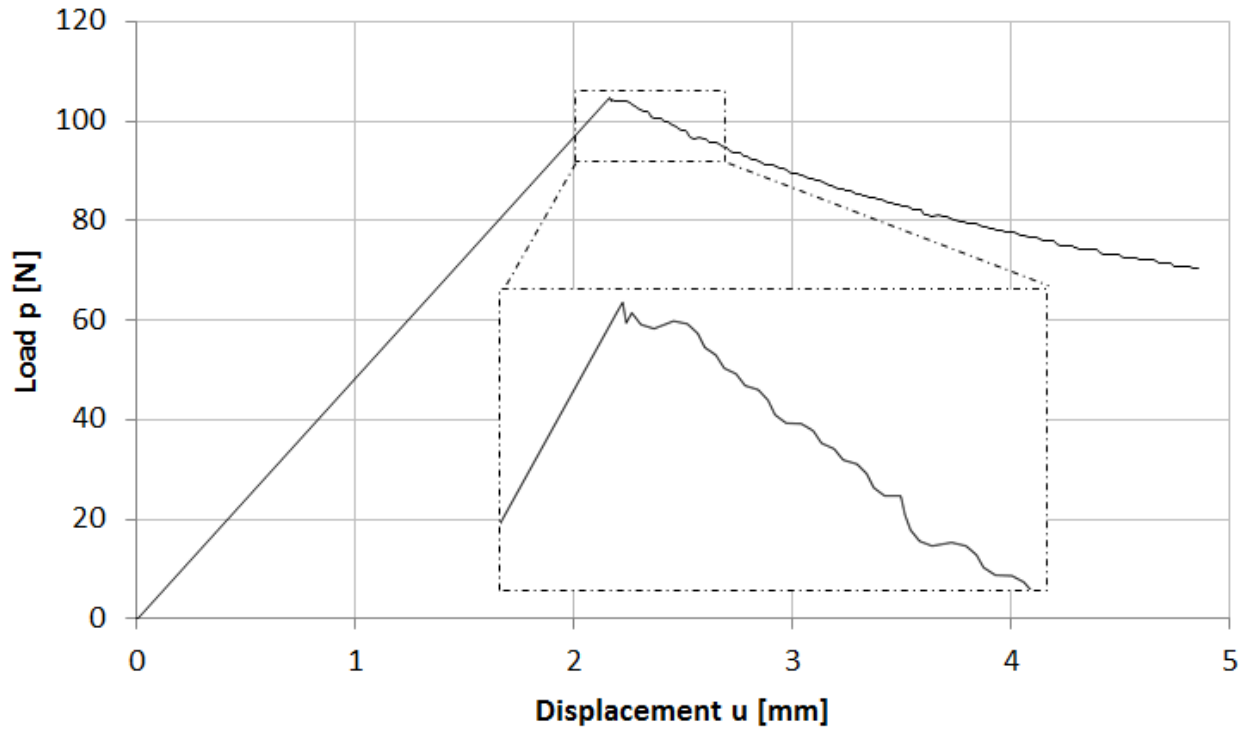


Figure 5. Spurious oscillations in the load-displacement response of a double cantilever beam (DCB) test computed by VCCT.

When the arc-length method is used in combination with the VCCT method, the iteration (1) is conducted according to the previously described procedure, equations (6-9). At the end of iteration (1), if G_T is larger than G_c , the delamination is propagated until the crack is not critical. This propagation can be

operated, for example, under constant displacement boundary conditions in order to minimize the force residuals and increase the convergence rate. The delamination propagation is indicated with curve $A - B$ in Figure 6. When solving for a softening problem due to buckling or due to crack propagation modeled with cohesive damage mechanics, Figure 3, the tangent stiffness matrix linearizes the structural response by determining the tangent to the equilibrium path. For the VCCT modeling, on the contrary, the linearized response is, in general, not tangent to the equilibrium path, Figure 6. For this reason, the critical crack tip condition can be largely overshoot at the end of iteration (1), leading to high out-of-balance forces at the beginning of iteration (2). Moreover, the larger the arc-length the larger force imbalance and the longer propagation length is required after iteration (1). Hence, the number of iterations n_{AB} required to extend the delamination length, exponentially increases with the arc-length. The reason for the exponential increase is that a larger delamination extension is necessary in phase $A - B$ as the arc-length increases, therefore more finite propagations through the mesh have to be computed, and more than one iteration is required for each finite propagation to converge. In other words, the stepping nature of the equilibrium path due to discrete crack propagation is amplified by the arc-length method.

Once the crack is extended enough for G_T to drop below G_c , another iteration with constrained $\mathbf{U}$ and λ is performed as follows. The out-of-balance force vector at the beginning of iteration ($n_{AB} + 2$) is

$$-g(t+\Delta t, \lambda^{(n_{AB}+2)}) = {}^{t+\Delta t}\lambda^{(1)}\mathbf{R} - {}^{t+\Delta t}\mathbf{F}^{(n_{AB}+1)} + \Delta\lambda^{(n_{AB}+2)}\mathbf{R} \quad (27)$$

which gives the displacement correction

$$\Delta\mathbf{U}^{(n_{AB}+2)} = ({}^\tau\mathbf{K}^{(n_{AB}+1)})^{-1}({}^{t+\Delta t}\lambda^{(1)}\mathbf{R} - {}^{t+\Delta t}\mathbf{F}^{(n_{AB}+1)}) + \Delta\lambda^{(n_{AB}+2)}({}^\tau\mathbf{K}^{(n_{AB}+2)})^{-1}\mathbf{R} \quad (28)$$

The value of the load factor $\Delta\lambda^{(n_{AB}+2)}$ is calculated by substituting equation (28) using the constraint equation

$$(\mathbf{U}^{(1)} + \Delta\mathbf{U}^{(n_{AB}+2)})^T(\mathbf{U}^{(1)} + \Delta\mathbf{U}^{(n_{AB}+2)}) = \Delta l^2 \quad (29)$$

If at the configuration ${}^{t+\Delta t}\mathbf{U}^{(n_{AB}+2)}$ the crack is critical, it should be propagated again as described before and as indicated in Figure 6 with segment $C - D$.

The procedure is repeated until convergence to the desired arc-length. It should be noted that because of the non-tangency between the equilibrium path and the stiffness matrix, the control of the arc-length can be quite inaccurate, as shown in Figure 6. However, this is a minor shortcoming. Instead, an important disadvantage of the VCCT employed in conjunction with the arc-length method is that the use of the arc-length method likely decreases the convergence rate with respect to the conventional NR solution method. In elastic instability problems, on the other hand, the arc-length method improves the convergence rate of iterative incremental methods over stable branches of the equilibrium path [19][18]. If VCCT is employed, the repeated delamination propagations with large out-of balance forces lead to severe discontinuities that require a large number of iterations to be resolved. These discontinuities can also be seen as large spurious oscillations in the solution process, as depicted in Figure 6 by branches $A - B$ and $C - D$.

In summary, the use of the arc-length method for delamination propagation problems requires the method to be modified in order to capture sharp snap-back responses due to local failure, thereby losing its generality and elegance. In addition, the use of the arc-length method in conjunction with the VCCT decreases the convergence rate of the NR method. Moreover the NR method in conjunction with the VCCT require numerical experimentations and considerable knowledge and judgment by the user to assure a stable and accurate solution or propagation problems. These available methods do not satisfy the requirement of computational efficiency and robustness to perform the parametric design study of the TFB-CFRP laminate. For these reasons, a novel automatic solution method was developed.

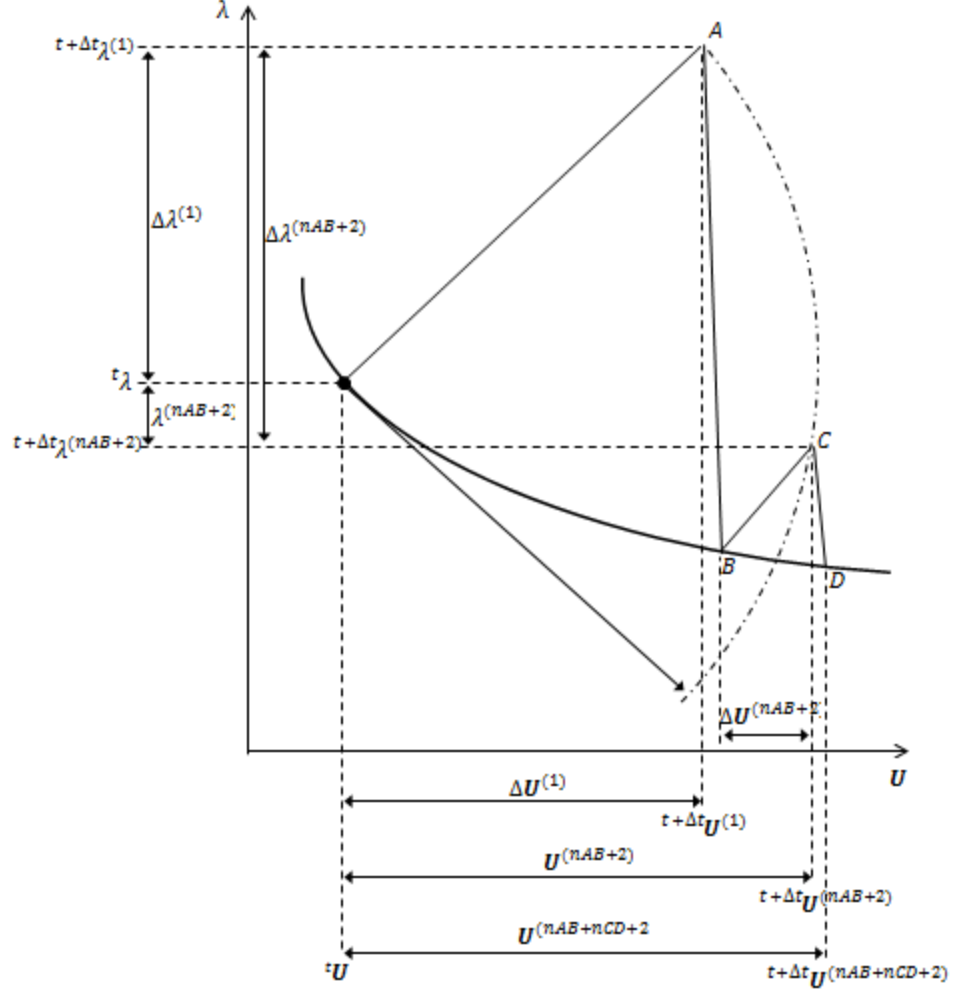


Figure 6. Schematics of the spherical arc-length method with VCCT delamination propagation in a bi-dimensional problem

3.5 The crack-length control scheme

The proposed method consists of an automatic incremental solution algorithm for implicit nonlinear static finite element equations, designed to compute local and global structural collapse by stable and unstable delamination propagation and delamination buckling. Like the arc-length method, this load-displacement-constraint method iterates in the load-displacement space utilizing the Newton-Raphson method, allowing to trace out the complete equilibrium path of the structure. The solution control parameter remains the coordinate s that follows the equilibrium path, and its approximate increment, the so called arc-length Δl .

However, the constrained variables are not the load and the displacements, or a set of displacements, but the load and the delamination length.

The idea of utilizing the crack length as a control parameter for solving snap-back instabilities was proposed by [29] [11]. However, in the current research effort the delamination length is not the control parameter, but a constrained variable.

In order to define a delamination length, the structural problem has to be idealized by a bi-dimensional model. Moreover, the loading is supposed to be dependent on a single intensity parameter λ that controls the magnitude of all the applied loads, which are therefore proportionally varying. In a finite element discretization the intensity parameter can be defined as the norm of the vector of the applied nodal forces divided by a reference force magnitude α

$$\lambda = \frac{\|\mathbf{R}\|}{\alpha} \quad (30)$$

The numerator of equation (30) is the magnitude of the applied loads resultant, which is named p .

Since we don't use nodal displacements as constrained variables, the analyst can arbitrarily define the nondimensional load-displacement space of interest, which is the space where the arc-length Δl is defined. In many practical cases the structural response is wanted with respect to a single scalar displacement u . For the proposed method the load-displacement response has to be defined in a bi-dimensional space $\lambda - u/\beta$, where β is a reference displacement, Figure 7. The coefficients α and β are defined by the user as the maximum expected load and displacement respectively, possibly multiplied by a common factor to avoid numerical truncation errors in case of small values.

It is assumed that an appropriate bi-dimensional finite element representation of the physical problem has been selected and only the solution of the governing static equations is discussed hereinafter. Assuming that the structural configuration is known at time t and using the notation on [16], these equations are

$${}^{t+\Delta t}\mathbf{R} - {}^{t+\Delta t}\mathbf{F} = 0 \quad (31)$$

where ${}^{t+\Delta t}\mathbf{R}$ is the vector of externally applied nodal forces, and ${}^{t+\Delta t}\mathbf{F}$ is the vector of nodal point forces equivalent to the internal element stresses, both being evaluated at time $t + \Delta t$. By applying an implicit incremental method [16], the solution of equations (31) results in the following iterative scheme

$${}^\tau\mathbf{K}^{(i-1)}\Delta\mathbf{U}^{(i)} = {}^{t+\Delta t}\mathbf{R} - {}^{t+\Delta t}\mathbf{F}^{(i-1)} \quad (32)$$

where ${}^\tau\mathbf{K}^{(i-1)}$ is a tangent stiffness matrix and $\Delta\mathbf{U}^{(i)}$ is the correction at iteration i of the current displacement vector

$${}^{t+\Delta t}\mathbf{U}^{(i)} = {}^{t+\Delta t}\mathbf{U}^{(i-1)} + \Delta\mathbf{U}^{(i)} \quad (33)$$

Depending on how the tensors in equation (32) are assembled and updated, the proposed method can be applied to problems that exhibit geometric and material nonlinearities as well as large strains.

In a conventional solution method, the new equilibrium configuration ${}^{t+\Delta t}\mathbf{U}^{(i)}$ is computed by solving through FNR or quasi Newton-Raphson (QNR) iterations the system of N simultaneous equilibrium equations (32) for the displacement corrections $\Delta\mathbf{U}^{(i)}$. The increment Δt is identified by the arc $A - C$ in Figure 7. Instead of following the physical equilibrium path, the increment Δt is split into a growth sub-increment, identified with the equilibrium path $A - B$, which is solved first, and a subsequent equilibrium sub-increment, $B - C$, which computes the new equilibrium configuration ${}^{t+\Delta t}\mathbf{U}^{(i)}$. In terms of displacements we have

$${}^{t+\Delta t}_g\mathbf{U}^{(gi)} = {}^t_g\mathbf{U} + \mathbf{U}^{(gi)}, \quad \mathbf{U}^{(gi)} = \sum_{m=1}^{gi} \Delta\mathbf{U}^{(m)} \quad (34)$$

$${}^{t+\Delta t}\mathbf{U}^{(i)} = {}^{t+\Delta t}_g\mathbf{U} + \mathbf{U}^{(ei)}, \quad \mathbf{U}^{(ei)} = \sum_{n=1}^{ei} \Delta\mathbf{U}^{(n)} \quad (35)$$

where

$$i = {}_g i + {}_e i \quad (36)$$

Having a control parameter implies that the value of a variable, or a set of variables, has to be constrained by an additional equation that includes the control parameter. The constrained variable of the growth sub-increment is the delamination length a . During growth the delamination length is increased by a finite value Δa . The propagation is achieved by releasing one or more couples of coincident interface nodes at

the beginning of the first iteration. For simplicity the model was not remeshed and the delamination was propagated through the number of finite elements that best approximated Δa . As a constrained variable, Δa is computed by solving a scalar constraint equation as discussed below. In order to solve the severe discontinuity of a finite crack opening, FNR iterations (g^i) are employed while the applied displacement is maintained constant, which is accomplished by conducting the growth sub-increment under displacement control. The equilibrium equation to be solved is then

$${}^{t+\Delta t}_g \mathbf{K}({}^{g^{i-1}}) \Delta \mathbf{U}({}^{g^i}) = {}^{t+\Delta t}_g \mathbf{R}({}^{g^i}) - {}^{t+\Delta t}_g \mathbf{F}({}^{g^{i-1}}) \quad (37)$$

Maintaining the applied displacement constant allows to minimize the out-of-balance forces, thereby increasing the convergence rate. In case of a distributed applied load, it could be more efficient to perform the increment under constant loading. When concentrated forces are applied the first method led to higher computational efficiency.

The equilibrium sub-increment consists of converging to the configuration of incipient growth under constant delamination length $a + \Delta a$, starting from the converged configuration at the end of the preceding growth sub-increment. This is usually a relatively small load increment with generally mild geometric nonlinearities, which is solved with the FNR method or the BFGS method [30]. The constrained variable of the equilibrium sub-increment is the applied load vector, which is controlled by scaling the applied load vector computed by the last converged iteration of the growth sub-increment. Scaling is performed by a load increment intensity parameter φ

$${}^{t+\Delta t} \tilde{\mathbf{K}}({}^{e^{i-1}}) \Delta \mathbf{U}({}^{e^i}) = \varphi {}^{t+\Delta t}_g \mathbf{R} - {}^{t+\Delta t} \mathbf{F}({}^{e^{i-1}}) \quad (38)$$

The parameter φ can be greater or smaller than 1. In fact, automatic loading should occur when the delamination growth is stable, whereas automatic unloading should occur under unstable growth. The parameter φ , as for Δa in the growth sub-increment, is computed by solving another constraint equation. Hence the applied load is not under user control and becomes an additional variable. In the current implementation of the method, φ is computed at the beginning of each equilibrium sub-increment and is

kept constant within the increment. It is important to remark that in equation (37) and (38) the applied load was assumed deformation independent.

The BFGS method, which is a QNR method was introduced for the purpose of minimizing the number of matrix ${}^{t+\Delta t}\mathbf{K}^{(e^{i-1})}$ factorizations by updating it with an approximate secant stiffness matrix ${}^{t+\Delta t}\tilde{\mathbf{K}}^{(e^{i-1})}$, thereby increasing computational efficiency in certain circumstances. The performance of the BFGS and FNR methods in the solution of the equilibrium sub-increments was characterized with benchmark examples. The results are discussed in the next section.

Line searching was necessary to improve the robustness and convergence rate of the BFGS method. When the BFGS method is employed, the displacement vector is updated by adding a multiplier β^{ls} to the displacement correction

$${}^{t+\Delta t}\mathbf{U}^{(i)} = {}^{t+\Delta t}\mathbf{U}^{(i-1)} + \beta^{ls} \Delta \mathbf{U}^{(e^i)} \quad (39)$$

A search algorithm computes the value of β^{ls} that minimizes the force residuals along the direction defined by the displacement correction vector $\Delta \mathbf{U}^{(e^i)}$. The force residuals at iteration (e^i) are compared to the force residuals of the last converged iteration $(e^i - 1)$ until the following convergence condition is satisfied

$$\frac{(\Delta \mathbf{U}^{(e^i)})^T (\varphi {}^t\mathbf{R} - {}^{t+\Delta t}\mathbf{F}^{(e^i)})}{(\Delta \mathbf{U}^{(e^i)})^T (\varphi {}^t\mathbf{R} - {}^{t+\Delta t}\mathbf{F}^{(e^{i-1})})} \leq S_{tol} \quad (40)$$

The nodal forces ${}^{t+\Delta t}\mathbf{F}^{(e^i)}$ correspond to the displacements ${}^{t+\Delta t}\mathbf{U}^{(i)}$ and are computed through back substitution for different values of β^{ls} until the above ratio is equal or smaller than the convergence tolerance S_{tol} . If the convergence tolerance is not achieved within the maximum number of line search iterations, N^{ls} , the algorithm exits the line search loop. The line search also ceases if the change in β^{ls} is less than η^{ls} times its current value. The minimization is constrained by the following constraint conditions

$$s_{min}^{ls} \leq \beta^{ls} \leq s_{max}^{ls} \quad (41)$$

For the present work η^{ls} was set to 0.01; s_{min}^{ls} and s_{max}^{ls} were set to 0.1 and 1.5 respectively. The values for S_{tol} and N^{ls} were established through a parametric study reported in the following section.

The elastic branch of the equilibrium path that precedes delamination propagation is solved as an equilibrium sub-increment. This initial loading phase can span through a large load increment and sometimes is affected by large displacements. For these reason the FNR method is always employed in this phase.

The stiffness matrices that are sent to the solver by alternating growth and equilibrium sub-increments are positive or negative definite, except when buckling occurs. In this cases the stiffness matrix could be singular. In any case the incremental scheme described above remains unchanged, but well established artificial stabilization methods, such as viscous forces, are automatically activated.

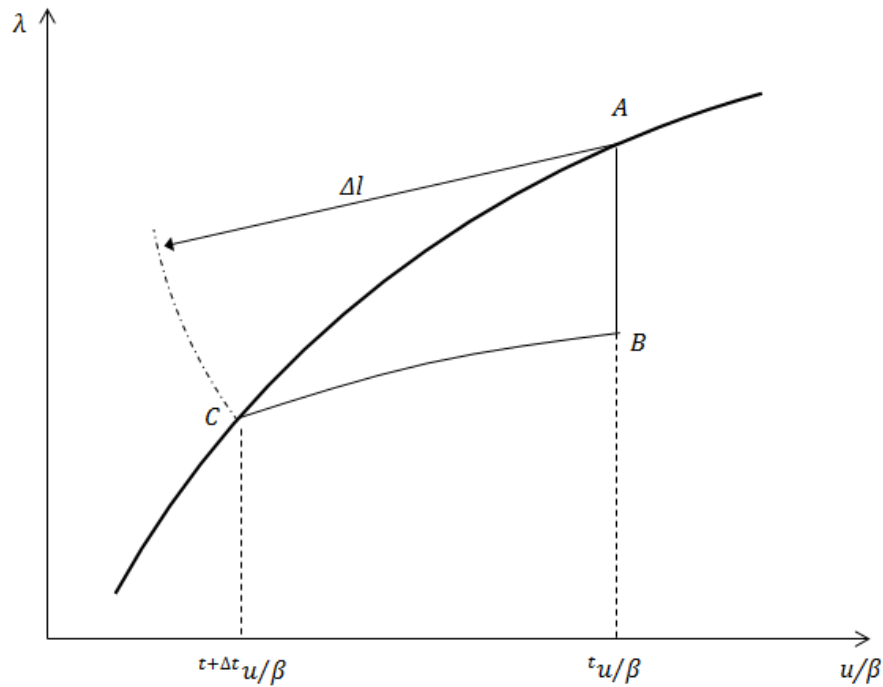


Figure 7. Schematics of a crack-length control scheme increment in a nondimensional load - displacement diagram.

In order to develop the constraint equations that relate Δl to Δa and φ , let us consider a known configuration at time t . An approximate relation between the delamination length increment $^{t+\Delta t}\Delta a$ and the arc-length that spans from t to $t + \Delta t$ is derived from an explicit computation of the arc-length components using the forward Euler method, Figure 8. The time variable associated with these linearly extrapolated variables is indicated with $\tilde{t}$ to remark that they are an approximate estimation of the corresponding values computed by FEA. The length $^{t+\Delta t}\Delta a$ is simply indicated as Δa for better clarity. The arc-length component of the growth sub-increment equals the nondimensional variation of the applied load, which is expressed by a first order Taylor series expansion

$$_g\Delta\tilde{l} = \frac{1}{\alpha} \left(^{t+\Delta\tilde{t}}_gp - ^tp \right) \quad (42)$$

$$_g\Delta\tilde{l} = \frac{1}{\alpha} \left(\frac{dp}{da} \right)_u \Delta a \quad (43)$$

Likewise the load component of the equilibrium sub-increment path is

$$_e\Delta\tilde{l}_p = \frac{1}{\alpha} \left(^{t+\Delta\tilde{t}}_p - ^{t+\Delta\tilde{t}}_gp \right) \quad (44)$$

$$_e\Delta\tilde{l}_p = \frac{\varphi-1}{\alpha} \left[\left(\frac{dp}{da} \right)_u \Delta a + ^tp \right] \quad (45)$$

In order to define the displacement component of the equilibrium path, we introduce the compliance, C , which refers to the load p and the displacement u .

The displacement component is then equal to

$$_e\Delta\tilde{l}_u = \frac{1}{\beta} \left(^{t+\Delta\tilde{t}}_gC \left(^{t+\Delta\tilde{t}}_p - ^{t+\Delta\tilde{t}}_gp \right) \right) \quad (46)$$

$$_e\Delta\tilde{l}_u = \frac{\varphi-1}{\beta} \left[\left(\frac{dp}{da} \right)_u \Delta a + ^tC \left(^tp + \left(\frac{dC}{da} \right) \Delta a \right) + \left(\frac{dC}{da} \right) \left(\frac{dp}{da} \right)_u \Delta a^2 \right] \quad (47)$$

If we neglect the last member of the addition we obtain

$$_e\Delta\tilde{l}_u \cong \frac{\varphi-1}{\beta} \left\{ \left(\frac{dp}{da} \right)_u \Delta a + ^tC \left(^tp + \left(\frac{dC}{da} \right) \Delta a \right) \right\} \quad (48)$$

The first constraint equation can be assembled by substituting equations (43), (45) and (48) into the following arc-length condition

$$({}_g\Delta\tilde{l} + {}_e\Delta\tilde{l}_p)^2 + {}_e\Delta\tilde{l}_u^2 = \Delta l^2 \quad (49)$$

Equation (49) is a quadratic equation in the unknown Δa and can be written as

$$X\Delta a^2 + \Psi\Delta a + \Omega = 0 \quad (50)$$

where

$$X = \frac{\varphi^2}{\alpha^2} {}^t\left(\frac{dp}{da}\right)_u^2 + \frac{(\varphi-1)^2}{\beta^2} \left[{}^tC {}^t\left(\frac{dp}{da}\right)_u + {}^t\left(\frac{dC}{da}\right) {}^tp \right]^2 \quad (51)$$

$$\Psi = \frac{2(\varphi^2-\varphi)}{\alpha^2} {}^t\left(\frac{dp}{da}\right)_u {}^tp + \frac{2(\varphi-1)^2}{\beta^2} \left[{}^tC^2 {}^tp {}^t\left(\frac{dp}{da}\right)_u + {}^tC {}^tp^2 {}^t\left(\frac{dC}{da}\right) \right] \quad (52)$$

$$\Omega = (\varphi - 1)^2 \left[\frac{1}{\alpha^2} {}^tp^2 + \frac{1}{\beta^2} {}^tC^2 {}^tp^2 \right] - \Delta l^2 \quad (53)$$

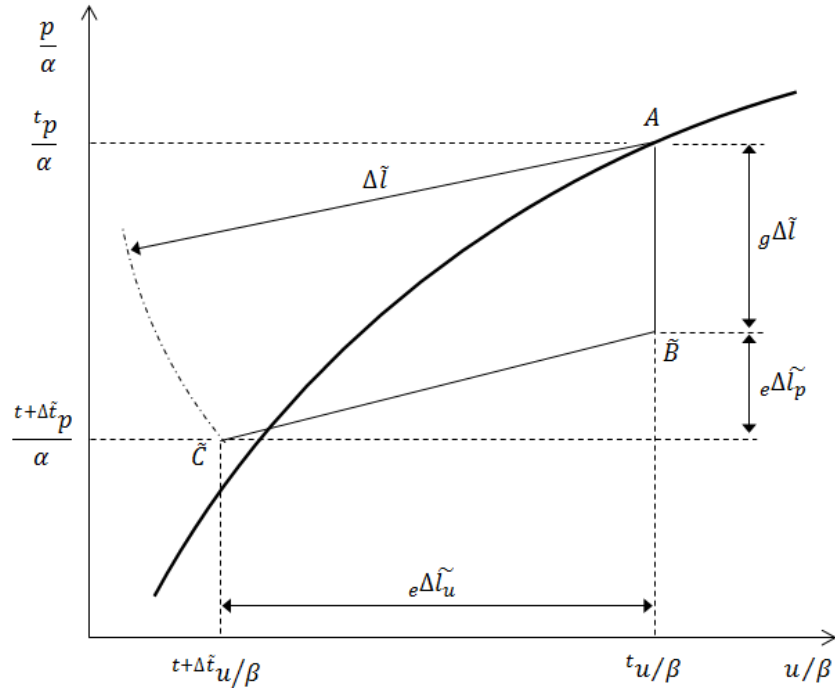


Figure 8. Schematics of the linear extrapolation of the arc-length components.

Equation (50) is a scalar equation that, given a desired arc-length Δl , can be used to compute the delamination length increment Δa to be applied in the growth sub-increment. The equation must be solved at the beginning of each growth sub-increment in order to update the delamination length. After being updated, the delamination length remains constant within the increment. A root of the equation is a

positive value that corresponds to the delamination length increment that allows the solution to proceed forward along the equilibrium path. The other root is a negative value that corresponds to the opposite direction. Since a is monotonically increasing with time, always the positive root is selected.

In order to solve equation (50), however, the values of the derivatives of the load and the compliance with respect to the crack length and the value of the load increment intensity parameter φ have to be known. The two derivatives were computed by backward difference or analytic models. The results presented in the next section were obtained with analytic sensitivities. The description of the numerical computation of the derivative by backward difference is omitted, whereas the derivation of the analytic sensitivities is described in detail.

An analytic relation between the compliance and the crack length increment was found by assuming a linear elastic response. Within this assumption we can apply the Castigliano's theorem to a generic cracked body with an applied resultant load p and derive the relation between the variation of compliance and strain energy U upon an infinitesimal crack surface growth dA

$$dC = \frac{2dU}{p^2} \quad (54)$$

Noting that for a constant applied load the potential energy Π is given by

$$\Pi = -U \quad (55)$$

we can substitute equations (54) and (55) into the definition of the strain energy release rate

$$G_T = -\frac{d\Pi}{dA} \quad (56)$$

and obtain the analytic sensitivity equation [31]

$${}^t\left(\frac{dC}{da}\right) = \frac{2b}{{}^tp^2} {}^tG_T \quad (57)$$

where b is the width of the bi-dimensional structural problem under consideration.

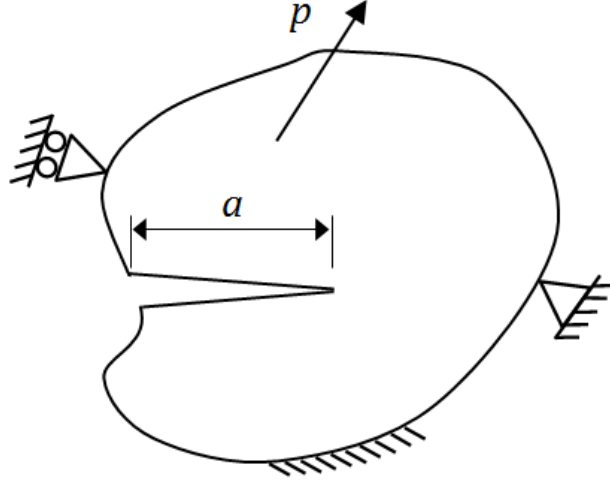


Figure 9. Generic linear elastic solid body with a crack

Another condition for the application of the Castigliano's theorem is that the displacement u must be the displacement at the point of application of the resultant force p , in the direction of p .

Under the same assumptions, the load sensitivity to an infinitesimal crack length increase in a displacement controlled structure is [31]

$${}^t \left(\frac{dp}{da} \right)_u = - \frac{2b}{t_u} {}^t G_T \quad (58)$$

The load increment intensity parameter can be approximated by the extrapolated load magnitude at the end of the equilibrium sub-increment ${}^{t+\Delta\tilde{t}}p$ divided by the extrapolated load magnitude at the end of the growth sub-increment ${}^{t+\Delta\tilde{t}}_g p$

$$\tilde{\varphi} = \frac{{}^{t+\Delta\tilde{t}}p}{{}^{t+\Delta\tilde{t}}_g p} \quad (59)$$

By substituting equation (57) into (59) and noting that, for small increments, the derivative of the compliance with respect to the crack length can be considered constant throughout an equilibrium sub-increment, we obtain

$$\tilde{\varphi} = \left(\frac{{}^{t+\Delta\tilde{t}}G_T}{{}^{t+\Delta\tilde{t}}_g G_T} \right)^{1/2} \quad (60)$$

After imposing the condition of incipient delamination propagation, the following constraint equation for the equilibrium sub-increment is found

$$\tilde{\varphi} = \left(\frac{{}^t G_C}{{}^{t+\Delta\tilde{t}}_g G_T} \right)^{1/2} \quad (61)$$

In the above equation the mode mixity is assumed constant between two consecutive increments. In fact, if the mode mixity does not change between the end of time increment t and the end of increment $t + \Delta t$, the critical energy release rate ${}^{t+\Delta t} G_C$ must be equal to ${}^t G_C$. The invariance of the mode mixity between two consecutive increments is expected to be verified provided that elastic instabilities do not occur. The error associated with this assumption can be expected to be small provided that the delamination length increments are small enough.

The strain energy release rate ${}^{t+\Delta\tilde{t}}_g G_T$ was computed with the following generic second order numerical method

$${}^{t+\Delta\tilde{t}}_g G_T = {}^t G_T + {}^t \left(\frac{dG_T}{da} \right)_u \Delta a + \frac{1}{2} \frac{{}^t \left(\frac{dG_T}{da} \right)_u - {}^{t-\Delta t} \left(\frac{dG_T}{da} \right)_u}{{}^t \Delta a} \Delta a^2 \quad (62)$$

where ${}^t \Delta a$ is the growth value adopted in the previous increment.

The derivatives of the energy release rate were computed with backward difference between the end (${}^{\tau-\Delta t}_g G_T$) and the beginning (${}^{\tau-\Delta t} G_T$) of the preceding growth sub-increment

$${}^{\tau} \left(\frac{dG_T}{da} \right)_u = \frac{{}^{\tau-\Delta t}_g G_T - {}^{\tau-\Delta t} G_T}{{}^t \Delta a} \quad (63)$$

The two constraint coupled equations (50) and (61) were solved by numerical iterations, starting from an initial numerical value for Δa equal to ${}^t \Delta a$

$$\begin{cases} X\Delta a^2 + \Psi\Delta a + \Omega = 0 \\ \tilde{\varphi} = \left(\frac{{}^t G_C}{{}^{t+\Delta\tilde{t}}_g G_T} \right)^{1/2} \end{cases} \quad (64)$$

The range of allowable values of Δa was limited by a maximum and minimum value

$$\Delta a_{min} \leq \Delta a \leq \Delta a_{max} \quad (65)$$

The solution was considered converged when the relative change in Δa was within 5%

$$\left| \frac{\Delta a_i}{\Delta a_{i-1}} - 1 \right| \leq 0.05 \quad (66)$$

or the solution was accepted when a maximum number of iterations was achieved. The system of equations (64), which is solved at the beginning of each growth sub-increment, provides the finite delamination length increase to be applied in the current increment. At the beginning of each equilibrium sub-increment the exact load intensity parameter φ can be calculated from the ERR computed at the end of the growth sub-increment

$$\varphi = \left(\frac{t_{G_C}}{t + \Delta t_{G_T}} \right)^{1/2} \quad (67)$$

The strain energy release rates relevant to delamination mode I (G_I) and mode II (G_{II}) are computed with the VCCT, whereas the critical energy release rate G_C is calculated with the BK law

$$G_C = G_{IC} + (G_{IIC} - G_{IC}) \left(\frac{G_{II}}{G_T} \right)^\eta \quad (68)$$

Since the energy release rate is not available at the beginning of the first increment, a load perturbation increment, of the size of a small fraction of the maximum expected load, must be applied at the beginning of the analysis. The perturbation increment provides the ERR data required by the subsequent equilibrium sub-increment. Once the solution is converged at the critical loading condition, the delamination is propagated in the first growth sub-increment by a user defined amount Δa_0 . The initial delamination length increment Δa_0 , together with Δa_{min} , Δa_{max} and the normalizing factors α and β , are the user inputs for the *crack length control scheme*.

In the subsequent increments, Δa is automatically computed from equations (64), whereas the arc-length value is updated at the beginning of each growth sub-increment using the following relation

$$\Delta l_{new} = \sqrt{\frac{n_d}{n_t}} \Delta l_{old} \quad (69)$$

where n_d is the desired number of iterations per increment (total of growth and equilibrium sub-increment), and n_t is the total number of iterations of the previous increment. The arc length is therefore controlled by the convergence rate.

The computation of ${}^{t+\Delta\tilde{t}}_g G_T$, as described in equations (62) and (63), requires data from two converged growth sub-increments. Since at the second growth sub-increment only one data point is available, ${}^{t+\Delta\tilde{t}}_g G_T$ is computed by forward Euler.

An increment is considered converged if certain convergence criteria are met. For the growth sub-increments, a force and a displacement criteria have to be satisfied. The convergence criteria for forces is evaluated against the largest force residual at the end of iteration (i), defined as

$$r_{max}^{(i)} = \max |{}^{t+\Delta t} \mathbf{R}^{(i)} - {}^{t+\Delta t} \mathbf{F}^{(i)}| \quad (70)$$

We define the average magnitude of all the nodal force components, either internal or externally applied forces, throughout the entire model as

$${}^{t+\Delta t} \bar{q}^{(i)} = \frac{\sum_{e=1}^E \sum_{n_e=1}^{N_{n_e}} \sum_{j=1}^{N_{n_e}} |{}^{t+\Delta t} q_{j,n_e}^{(i)}| + \sum_{k=1}^{N_{ef}} |{}^{t+\Delta t} q_k^{(i)}|}{\sum_{e=1}^E \sum_{n_e=1}^{N_{n_e}} N_{n_e} + N_{ef}} \quad (71)$$

where:

E is the total number of elements;

N_{n_e} is the number of nodes in element e ;

N_{n_e} is the number of degrees of freedom at node n_e ;

N_{ef} is the total number of externally applied nodal force components;

${}^{t+\Delta t} q_{j,n_e}^{(i)}$ is the j -component of a nodal force applied to node n_e by element e due to element stresses;

${}^{t+\Delta t} q_k^{(i)}$ is an externally applied nodal force component.

The overall time average of the nodal forces from the beginning of the analysis, including the current iteration (i), is calculated as

$${}^{t+\Delta t} \tilde{q}^{(i)} = \frac{1}{t_N} \sum_{j=1}^{N_t} {}^t_j \bar{q} + {}^{t+\Delta t} \bar{q}^{(i)} \quad (72)$$

${}^t N$ is the number of converged increments at time t such that

$${}^t_j \bar{q} > 10^{-5} {}^t_j \tilde{q} \quad (73)$$

The value of ${}^{t+\Delta t}\bar{q}^{(i)}$ is recalculated at each iteration of the current increment and also ${}^{t+\Delta t}\tilde{q}^{(i)}$ is updated whenever the condition (73) satisfied. The value ${}^0\tilde{q}$ is initialized to 0.01 at the beginning of the analysis, whereas at the beginning of each increment it is set equal to the value computed in the last iteration of the previous increment.

The time averaged force $\tilde{q}$ can be very small if large parts of the model are inactive or experiencing only rigid body motion. This condition is verified, for example, when a sub-laminate is cut out from the load path by the delamination propagation. This could lead to an unduly strict convergence criterion. For this reason, a degree of freedom is marked as inactive if the corresponding nodal force component does not satisfy the following condition

$$\left| {}^{t+\Delta t}q_j^{(i)} \right| \geq 10^{-5} {}^{t+\Delta t}\tilde{q}_{max}^{(i)} \quad (74)$$

where ${}^{t+\Delta t}\tilde{q}_{max}^{(i)}$ is the largest time-averaged nodal force component, due to either external or internal forces. If the largest nodal force component ${}^{t+\Delta t}q_{max}^{(i)}$ is such that

$$\left| {}^{t+\Delta t}q_{max}^{(i)} \right| \geq 0.1 {}^{t+\Delta t}\tilde{q}_{max}^{(i)} \quad (75)$$

the spatial average is computed only across the active parts of the model, otherwise all inactive parts of the model are reclassified active. Finally, for a converged iteration the largest force residual must be less than 0.5% than the average nodal force

$$r_{max}^{(i)} \leq 0.005 {}^{t+\Delta t}\tilde{q}^{(i)} \quad (76)$$

For the displacement criterion, the maximum nodal displacement correction computed in the current iteration must be small compared to the largest nodal incremental displacement computed in the current increment, namely

$$\max |\Delta U^{(i)}| \leq 0.01 \max |{}^{t+\Delta t}U^{(i)} - {}^tU| \quad (77)$$

or the maximum displacement correction must be small compared to the estimated largest correction that would occur with one more iteration

$$\frac{r_{max}^{(i)}}{\min(r_{max}^{(i-1)}, r_{max}^{(i-2)})} \max |\Delta U^{(i)}| \leq 0.01 \max |{}^{t+\Delta t}U^{(i)} - {}^tU| \quad (78)$$

Conditions (76) and (77), or (76) and (78), have to be simultaneously satisfied. The solution of a sub-increment is initially attempted in one step. However, if either the force or the displacement criterion is not satisfied within 16 iterations, Δa is progressively cut down according to an automatic increment size control scheme and the solution of the growth sub-increment is attempted in multiple steps.

Equilibrium sub-increments must satisfy the same convergence criteria defined above, which ensure that the equilibrium equations are satisfied, but they also must satisfy the following ERR criteria

$$\left| \frac{t+\Delta t_{G_T^{(i)}}}{t+\Delta t_{G_C^{(i)}}} - 1 \right| \leq c_{tol} \quad (79)$$

which ensures convergence to incipient delamination growth.

If either the force or the displacement condition is not satisfied within 16 iterations, the sub-increment is repeated using the FNR method (this applies only when the BFGS method is employed). If convergence is not achieved again, the sub-increment is then reattempted through multiple FNR increments by cutting down the load increment size. The automatic increment control scheme consists in cutting down the initial equilibrium sub-increment size into 10 reduced increments of constant size. If necessary, the increment size is further reduced to obtain 100 reduced increments.

If the force and displacement criteria are satisfied, the solution is accepted because an equilibrium configuration has been found. However, if the ERR criterion is overshoot, the equilibrium sub-increment restarts from the new configuration with the automatic increment size cut down described above. Whereas if the ERR criterion has not been overshoot, the increment size is not cut-down.

When the increment cut down process is activated, the solution always restarts from the last converged iteration. Moreover, φ is updated at the beginning of each increment, based on the total ERR computed at the end of the previous increment. A peculiar characteristic of the *crack length control scheme* is that at any iteration, regardless if there are remaining increments to complete the set of reduced increments, an equilibrium sub-increment is declared converged when all three convergence criteria are satisfied, allowing the solution to move forward to the next growth sub-increment. This last feature, together with

the increment size reduction, avoids solution oscillations with repeated overshooting of the equilibrium condition at incipient delamination growth.

In any case, if the reduction of the increment size is not successful, artificial stabilization techniques based on viscous forces are automatically applied for either growth or equilibrium sub-increments. Since the method was implemented in Abaqus/Standard, two available techniques, namely contact stabilization and automatic stabilization [28], were employed. The first adds stiffness only to the nodes that are adjacent to the delamination surfaces, whereas the second to the whole model. Contact stabilization is first employed to reduce the large out-of-balance forces that arise upon crack opening or contact chattering. If convergence is not achieved the solution is restarted from the last converged increment with automatic stabilization.

These viscous nodal forces could introduce not negligible errors in the VCCT computation of the ERR that could accumulate and lead to gross and undetectable errors. Therefore a stabilization-free increment is always performed to compute the final equilibrium configuration. This ensures the accuracy of the nodal variables used by the VCCT.

The physical meaning and numerical repercussions of the convergence tolerance c_{tol} on the accuracy of the solution are the same as the previously discussed r_{tol} . For both of them, if the convergence tolerance is too large inaccurate results are obtained, whereas if the tolerance is too tight much computational effort is spent to obtain needless accuracy. The remarkable difference is that in a conventional VCCT implementation a converged equilibrium configuration is discarded if the ERR convergence criteria is not satisfied, equation (25). On the other hand, the *crack length control scheme* uses that converged equilibrium as a restart configuration.

Among the input parameters that were mentioned in this section, seven are peculiar to the proposed method and are listed in Table 1, whereas the others are required by any nonlinear analysis.

The possibility of extending the method to multi-delamination problems and to three-dimensional models has not been investigated at this time. Although the analytic model utilized to compute the applied load and compliance derivatives is capable to predict the epastic response of mutli-delaminated and three-

dimensional structures, further research is necessary to assess the feasibility of constraining a delamination area instead of the delamination length, as well as finding the equilibrium at incipient growth on a crack front instead of a crack tip. The capability of correctly propagating the delamination by a variable finite amount along the crack front seems to be the biggest challenge. The required software would likely be an elaborate and cumbersome addition to a finite element solver.

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3.6 Benchmarks

The crack-length control scheme was implemented in the commercial finite element software Abaqus/Standard, thereby taking advantage of the existing updated Lagrangian formulation and the FE solver. The load-displacement-constraint technique described in the previous section was implemented by means of a Python script that called a restart job for each sub-increment.

The simulation of a single leg bending test (SLB), a double cantilever beam test (DCB) and a delamination buckling problem were conducted to assess the effectiveness of the method in case of unstable delamination propagation, stable propagation and elastic instability respectively. Changes in the solution parameters settings were also discussed.

Only structural problems featuring a single concentrated applied nodal force were solved in order to characterize the basic performances of the method. Therefore growth sub-increments were conveniently performed under displacement control at the loading node. For problems where the loading is applied through distributed pressure forces on large surfaces or through body forces, delamination propagation under constant load control might perform better and be simpler to implement. In addition, in all the examples the vector of external loads was independent from the deformation. Several mesh sizes were utilized to test the computational efficiency of the method at varying number of degrees of freedom. In any case the mesh was structured so that the aspect ratio (length to width) for the elements always ranged between 0.1 and 10. The transitions between regions with different mesh sizes were always modeled with a progressive change in element dimension. The material model was linear elastic.

The performance of the proposed method was compared to conventional Abaqus/Standard VCCT solutions. All the Abaqus VCCT solutions were obtained in a single FNR step, with an initial increment size of 0.001 and a maximum increment size of 0.25. The default increment control scheme settings were utilized [28], but the maximum number of solution attempts was increased to 100 because many increment attempts at increasing crack lengths were needed to resolve unstable propagation. The VCCT release tolerance r_{tol} was set to 0.1. The settings for the *crack length control scheme* are listed in Table 1.

	E_1 [GPa]	$E_2=E_3$ [GPa]	$G_{12}=G_{13}$ [GPa]	G_{23} [GPa]	$\nu_{12}=\nu_{13}=\nu_{23}$	G_{IC} [MPa]	G_{IIC} [MPa]	η^1	Ply thickness [mm]
Material 1	146.86	10.62	5.45	3.99	0.33	248 ²	865 ²	1.512 ³	0.127
Material 2	162	8.34	4.96	3.11	0.34	316	579	1.6	0.127
1) Exponent of BK law. 2) Mean values of precracked specimens [33]. 3) Calculated based on mode mix ratio reported in [34].									

Table 1. Unidirectional material data.

	Δa_0 [mm]	Δa_{min} [mm]	Δa_{max} [mm]	α [N ⁻¹]	β [mm ⁻¹]	c_{tol} [-]	n_d [-]
SLB	5	1	10	1	0.018	0.005	12
DCB	1	1	2.5	1	0.0374	0.005	12
Buckling	2	2	5	1000	0.002	0.01	12

Table 2. Solver settings for the crack length control scheme.

The SLB is a test method designed to measure the fracture toughness of bi-material interfaces under mixed fracture mode I-II and for a wide range of mode mixity ratios [32]. Existing experimental data, closed form solutions and numerical simulations for the particular material and specimen configuration adopted in the present work can be found in the literature [33] [34]. The modeling technique adopted was preliminarily validated by comparing the computed structural response and mode mixity to these previous results. As reported by [34], geometric nonlinearities occurred before failure. In the present work the delamination was propagated to an extended length because the purpose was to perform a benchmark evaluation for the *crack length control scheme* method. For this reason the applied loading was such that large displacement occurred.

The SLB specimen consisted of a center-dalaminated, 32-ply laminate with stacking sequence [30/-30/0/-30/0/30/0₄/30/0/-30/0/-30/30/d/-30/30/0/30/0/-30/0₄/-30/0/30/0/30/-30], for a total thickness of 4.064mm. The span was 177.8 mm, the width was 25.4 mm and the delamination length spanned from 15 mm (a_0) to 100 mm. The initial delamination length a_0 was shorter than conventional SLB tests in order to reproduce a sharp snap-back instability in the structural response. The material was named ‘material 1’ and its properties are listed in Table 1. The boundary conditions are illustrated in Figure 10. For simplicity, instead of modeling the contact with support rollers, the specimen was hinged at its ends and the in-plane displacement of the left end hinged node was deemed negligible. For the same reason the force was applied at a mid-span node. The finite element mesh consisted of 8-node biquadratic plane stress quadrilateral elements with reduced integration (CPS8R). The 32 plies were modeled with one element per ply thickness for the two plies per each side adjacent to the crack. The other plies were modeled with one element each 4 plies and the 6 outermost plies with one element, with smeared material properties.

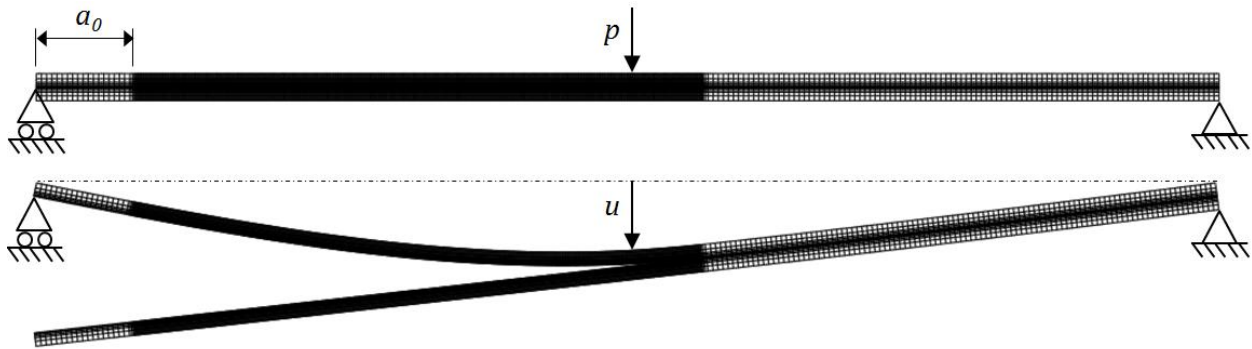


Figure 10. FEM of SLB specimen at zero and maximum unscaled deformation. Mesh size adjacent to delaminated interface 0.125 mm.

The *crack length control scheme* was able to trace the full equilibrium path. The result obtained with BFGS solution of the equilibrium sub-increments and a mesh size at the delaminated interface of 0.125 is shown in Figure 11. The curve markers indicate the system configurations at the end of each converged equilibrium sub-increments. Similar results were obtained with FNR solution of the equilibrium sub-

increments. The load-displacement curve was in good agreement with the prediction of the classical plate theory (CPT) analytic solution [32]. The CPT model diverged from the numerical results when the delamination length exceeded the half span. The reason for this discrepancy was that the change in the crack tip loading, that occurred approaching the point where the force was applied, was not correctly predicted by the analytic model. On the other hand, the FEA captured the change in the delamination propagation characteristics.

All the increments converged without cutting down the delamination length increment (growth) or the load increment (equilibrium). Convergence to incipient growth was always successful at the first attempt, except for the elastic branch. In this case the linear analytic model that was utilized to compute the load increment intensity parameter φ underestimated the critical load because of the geometric nonlinearities associated with the large incremental displacements.

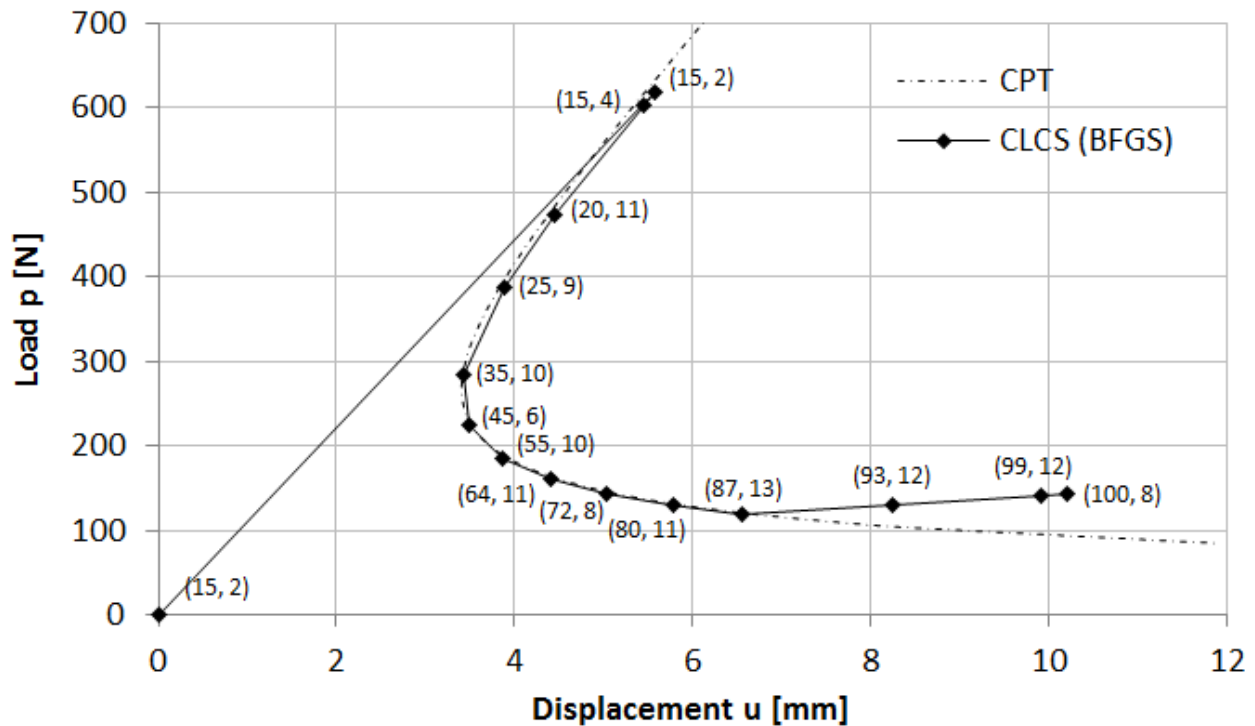


Figure 11. Crack length control scheme (CLCS) and closed form (CPT) solutions. Curve markers indicate converged increments. The delamination length and the total number of iterations required to converge are indicated in parenthesis.

For the same finite element discretization, the conventional Abaqus VCCT solution, obtained under displacement control and by allowing multiple nodes to release within an increment, is reported in Figure 12. As before, each curve marker represents a converged increment. The solution captured the sharp load drop from the critical point to the stable branch of the equilibrium path. To do so, a large number of iterations were required in the process of automatically cutting down the increment size by seven orders of magnitude when the critical load was approached.

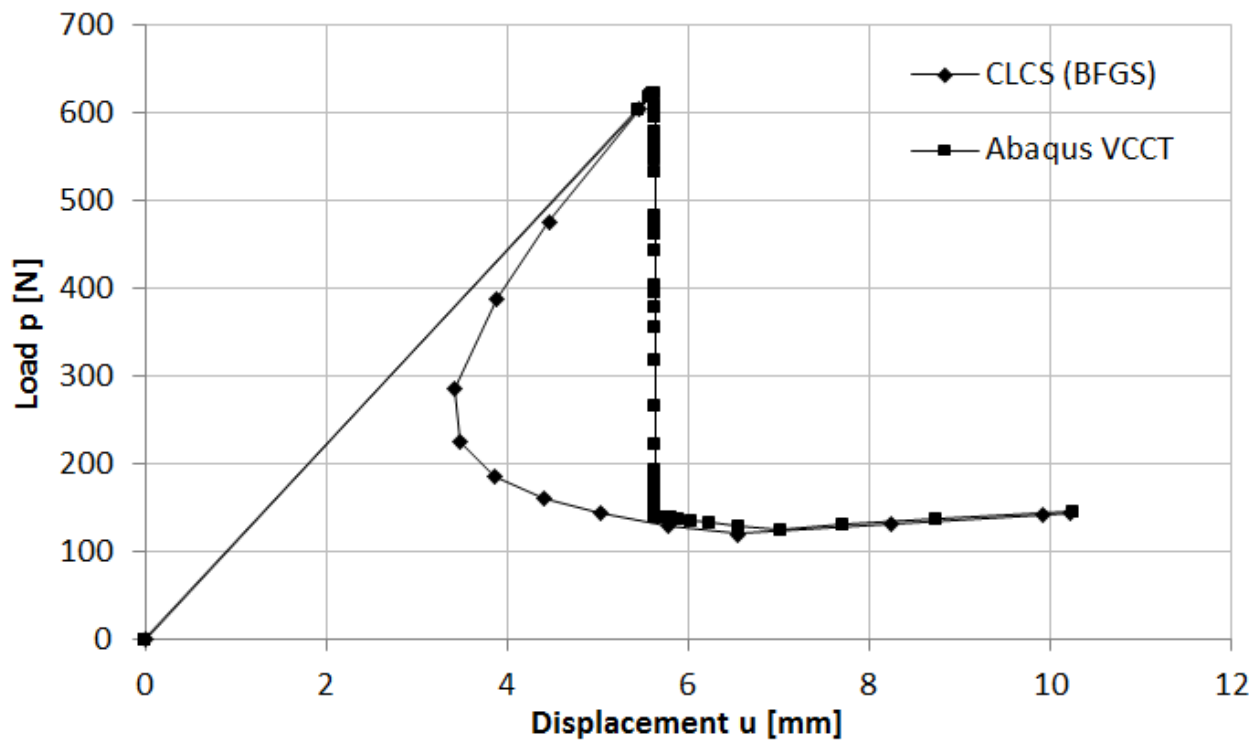


Figure 12. Crack length control scheme (CLCS) and Abaqus VCCT solutions.

The Abaqus VCCT solution required contact stabilization, a stabilization technique based on localized artificial nodal stiffness at the opening delamination surfaces [28]. Although abrupt crack opening also occurs during growth sub-increments, the *crack length control scheme* did not require the use of any artificial stabilization technique to achieve convergence. The reason lied in the difference between the two propagation schemes: in the first method delamination grew under increasing displacement, whereas in the second the applied displacement was fixed throughout the growth sub-increment. The need of a

stabilization technique, besides the already mentioned risk of introducing a not negligible error in the computation of the energy release rate, required a convergence study to find the optimum values for the stabilization coefficients. The convergence study shown in Figure 13 was performed using the single node propagation per increment, which explains the larger number of increments than the Abaqus VCCT solution reported in Figure 12, which was obtained by allowing multiple crack tip nodes to be released within the same increment. Stabilization was only applied to the degrees of freedom that were normal to the delaminated interface.

The results shown in Figure 13 confirm that, when the structural response is unknown a priori, the unstable propagation could remain undetected and the critical load be overestimated if an appropriate convergence study on the stabilization coefficients was not undertaken [26]. A preliminary scan of the equilibrium path with an efficient load-displacement-constraint method could provide useful information to the analyst.

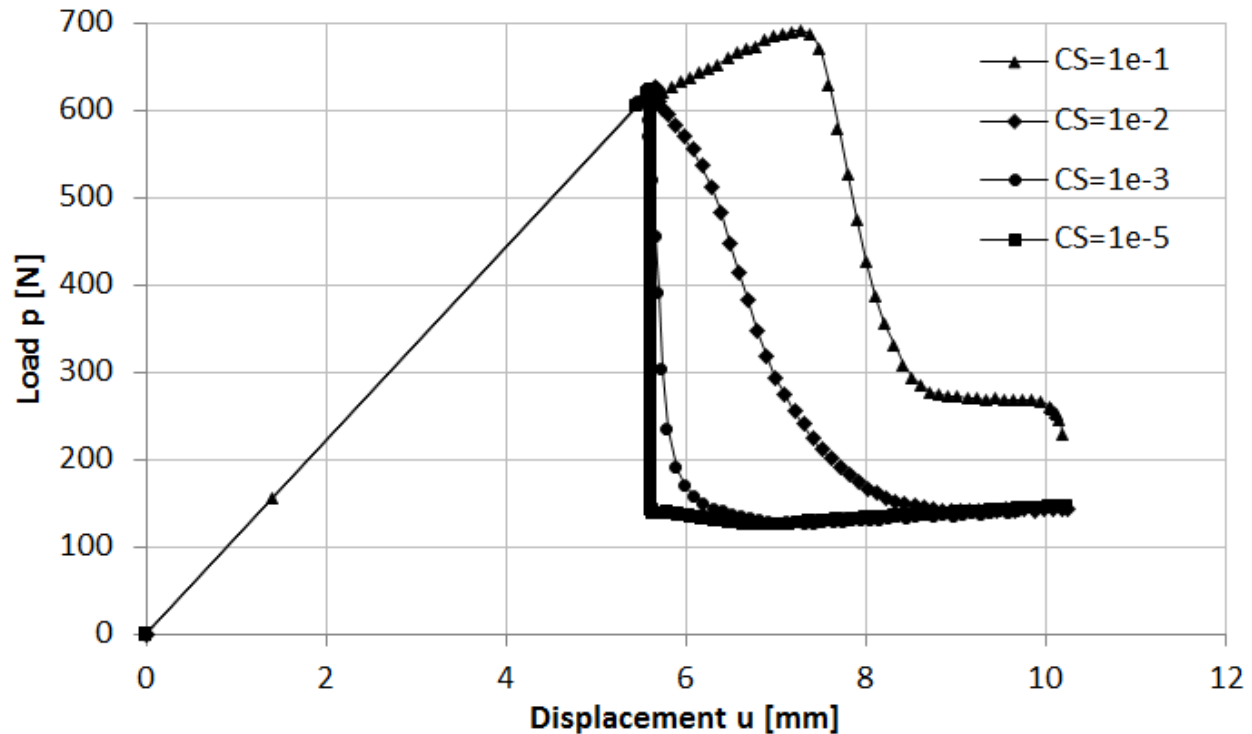


Figure 13. Convergence study on contact stabilization (CS) coefficient.

For the Abaqus VCCT solution, as expected, the finer the mesh at the delaminated interface, the higher the number of iterations required. The propagation condition on the ERR, stated by equation (25), has to be verified at a larger number of nodes if the mesh gets finer. In addition, for each node, multiple iterations are required to achieve convergence due to the severe discontinuity of the crack opening and to the load cutbacks. For these reasons the total number of iterations increased exponentially as the mesh size was reduced, Figure 14. On the contrary, in the proposed method the finite crack propagation length Δa can span across multiple elements and the energy release rate controls the increment size, thereby avoiding unduly expensive load cutbacks and automatically selecting the largest increment size that ensures the desired resolution on the structural response. In fact, the total number of iterations of the *crack length control scheme* showed to be independent from the mesh size and one or two orders of magnitude smaller than the Abaqus VCCT. This characteristic makes the *crack length control scheme* particularly appealing for fine mesh models.

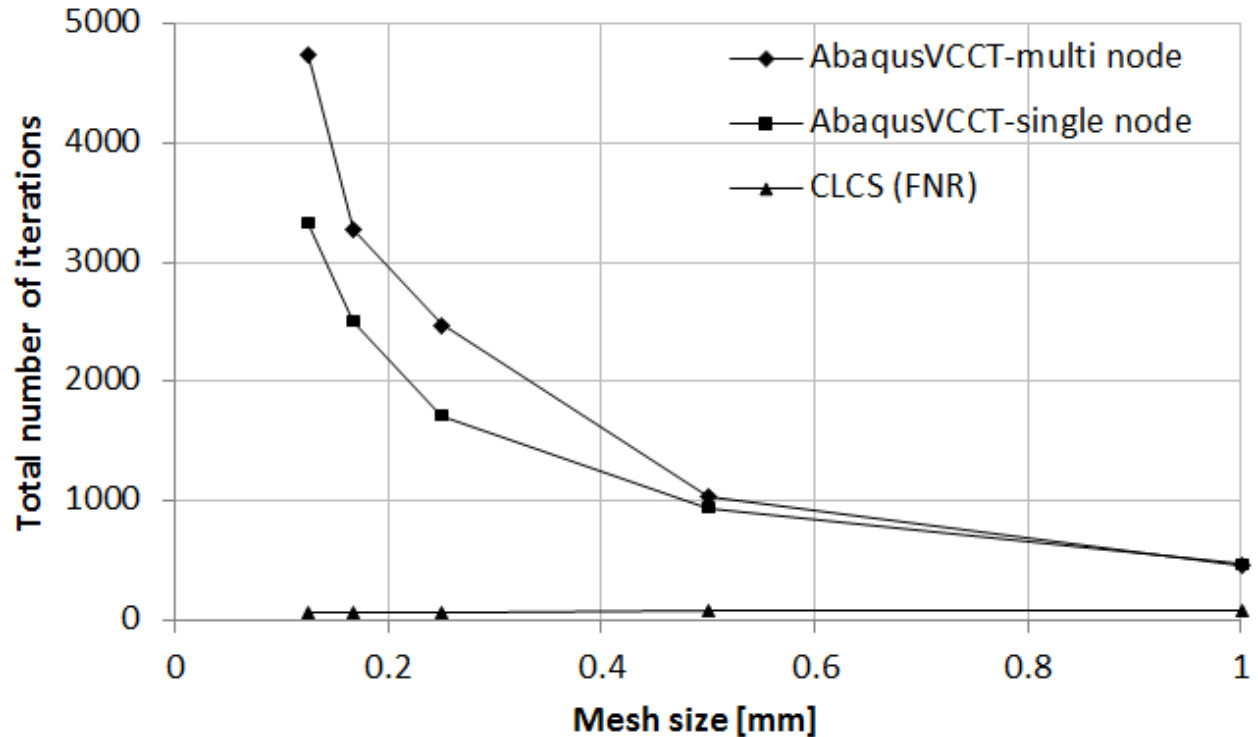


Figure 14. Number of iterations vs. mesh size at delaminated interface.

The optimum settings for the line search algorithm were determined by performing a parametric study on the search tolerance S_{tol} and the maximum number of allowed search iterations N^{ls} . The results are plotted in Figure 15 in terms of the ratio of the average CPU time for solving one equilibrium sub-increment with line searches, ${}_e\bar{t}^{ls}$, to the same average time without line searches, ${}_e\bar{t}$. When N^{ls} is equal to 0, that is when line searches were not performed, the two times coincide and their ratio is equal to unity. The solution time converged to a constant value within 10 search iterations. A small search tolerance caused the BFGS solution to diverge, resulting in the automatic FNR restart of the sub-increment and therefore leading to a sharp increase in ${}_e\bar{t}^{ls}$. Better results were obtained with less aggressive line searches, with the maximum computational efficiency achieved with a search tolerance of 0.9. With these settings, which were adopted for all the other benchmark examples, the line search algorithm allowed to reduce the solution time of the BFGS method by about 40%.

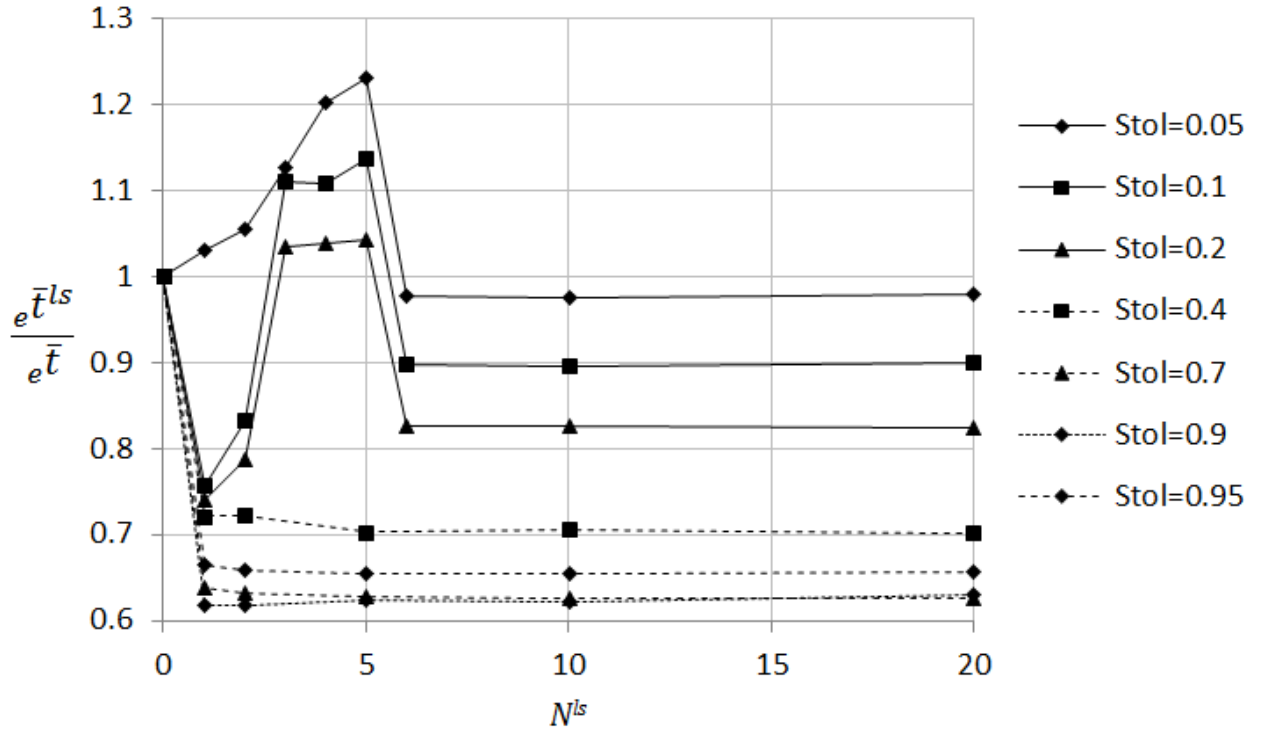


Figure 15. Ratio of average equilibrium sub-increment CPU time with line search (${}_e\bar{t}^{ls}$) to time without line search (${}_e\bar{t}$). N^{ls} is the maximum allowed number of line search iterations. S_{tol} is the line search convergence tolerance.

Nevertheless, the use of the BFGS method to solve equilibrium sub-increments showed to be less computationally efficient than the FNR method for all the five mesh sizes mentioned above, Figure 16. The ratio between solution times remained almost constant with the total number of degrees of freedom ranging from 10k to 64k.

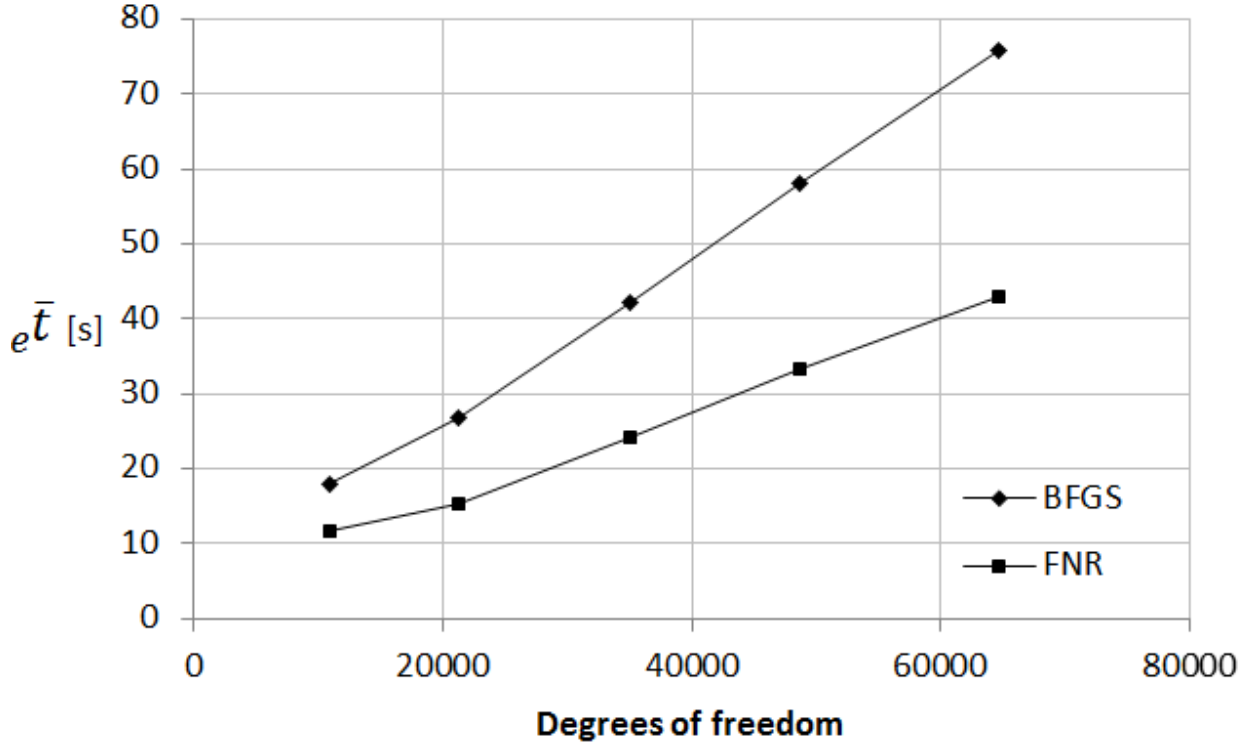


Figure 16. Average equilibrium sub-increment solution time ($e \bar{t}$) for BFGS and FNR solution method.

An arbitrary DCB test was simulated in order to assess the basic capabilities of the proposed analysis method in case of a stable delamination propagation. The specimen length was 150 mm, the width was 25.4 mm and the delamination length spanned from 38 mm (a_0) to 59 mm. The DCB specimen consisted of a 30 plies unidirectional laminate for a total thickness of 3.81 mm. The material was named ‘material 2’ and its properties are listed in Table 1. The finite element model is shown in Figure 17. The plies were modeled with one element per ply thickness for the two plies per each side adjacent to the crack, whereas the length of the elements was 0.125 mm. The other plies were modeled with one element per each of the next 3, 4 and the outermost 6 plies, for a total of 20920 degrees of freedom. The same plane stress elements utilized for the SLB specimen were employed.

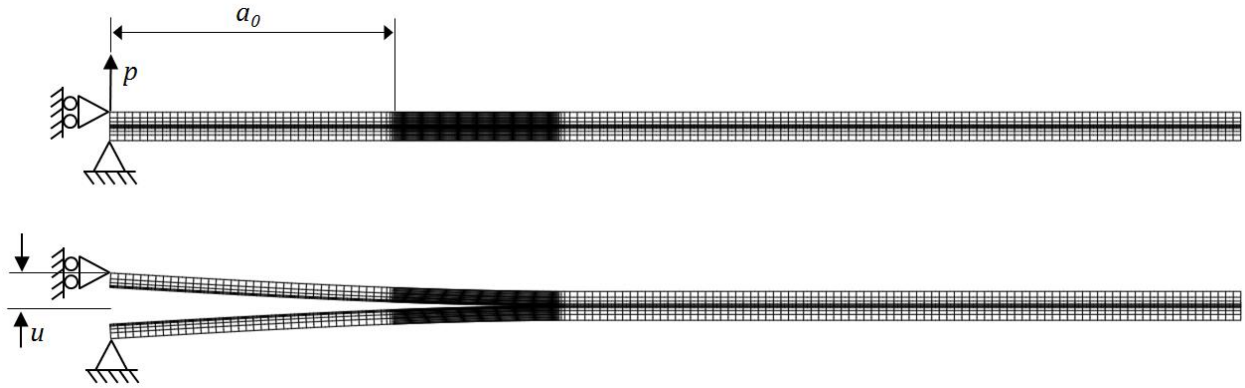


Figure 17. FEM of DCB specimen at zero and maximum unscaled deformation.

Figure 18 shows the structural response calculated by the FNR *crack length control scheme*, the Abaqus VCCT and the closed form solution based on the shear deformable plate theory (SDPT). For clarity, the curve markers of the Abaqus VCCT solution were omitted. The results of the three analysis methods were in good agreement. The branch with negative slope relevant to the Abaqus VCCT was slightly off because of the relatively large value of release tolerance (r_{tol}). After the initial 1 mm delamination length increment Δa_0 , the *crack length control scheme* proceeded by increasing Δa increments until it reached the maximum length increment Δa_{max} of 2.5 mm. This progressive increase took place within five increments. Thereafter the delamination propagated at constant increments lengths. Unlike the unstable delamination growth case, the number of iterations per increment remained constant during propagation, with 6 iterations per increment. Similar to the SLB example, all the increments converged at the first attempt, except for the large increment that traced the elastic branch. In total 68 FNR iterations were necessary for the *crack length control scheme* to solve the problem. Although the Abaqus VCCT solution did not need to dramatically cut down the increment size, because no unstable delamination occurred, it required 856 iterations for the single node propagation method and 805 iterations for the multiple node propagation method. The number of FNR iteration required by the *crack length control scheme* was then about one order of magnitude smaller than the Abaqus VCCT in case of stable propagation. The BFGS method was as computationally efficient as the FNR method in solving the equilibrium sub-increments, taking about the same average CPU time per increment.

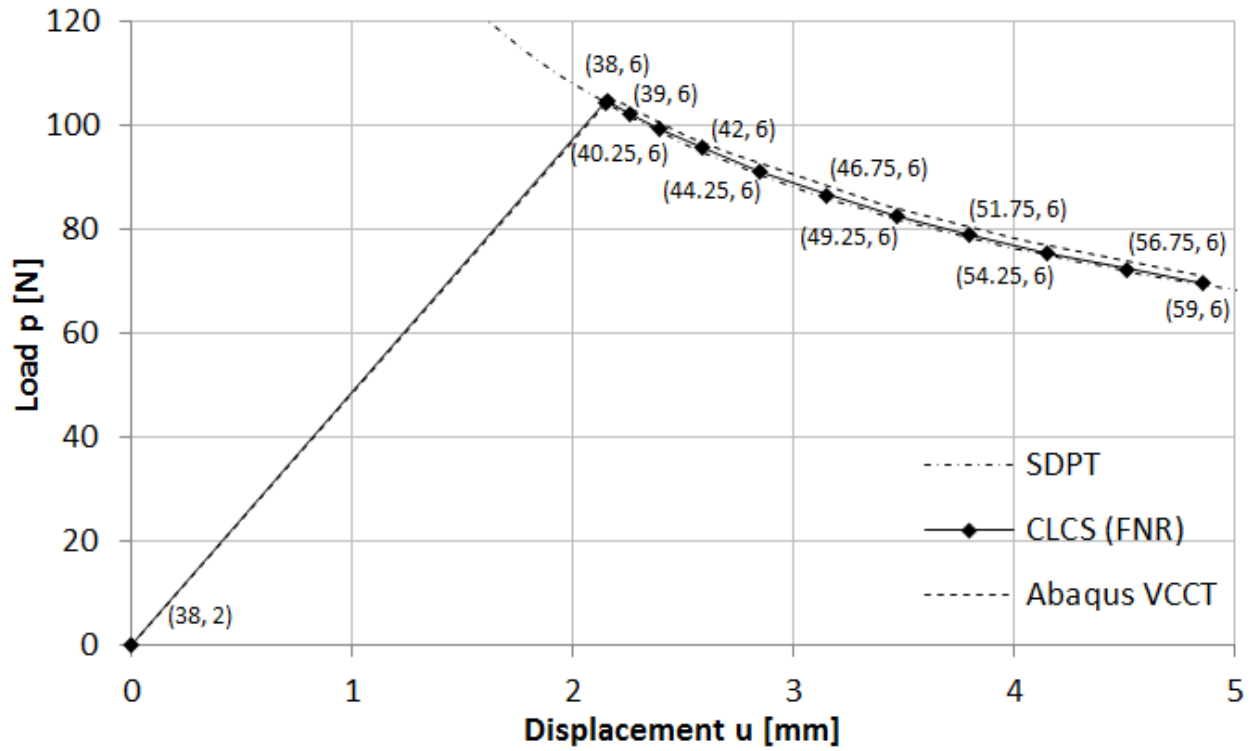


Figure 18. Crack length control scheme (CLCS), Abaqus VCCT and closed form (SDPT) solutions. Curve markers indicate converged increments. The delamination length and the number of iterations required to converge are indicated in parenthesis.

The simultaneous occurrence of delamination and local buckling was reproduced by means of a delaminated plate loaded under uniaxial compression. The plate length was 43 mm, the width was 76.2 mm and the centered delamination spanned from an initial length of 14.7 mm (a_0) to 42.7 mm. The laminate stacking sequence was $[0/45/90/-45/d/(0/45/90/-45)_4]$, for a total thickness of 2.54 mm. The material was ‘material 2’ with degraded G_{IC} and G_{IIC} to 189 J/m² and 347 J/m² respectively, Table 1. The finite element model is shown in Figure 19. The load was applied at the mid-depth of the lower sub-laminate. The plies were modeled with one element per ply thickness, whereas the length of the elements adjacent to the crack surfaces was 0.125 mm. The finite elements were 8-node biquadratic plane strain quadrilateral with reduced integration (CPE8R), for a total of 16644 degrees of freedom.

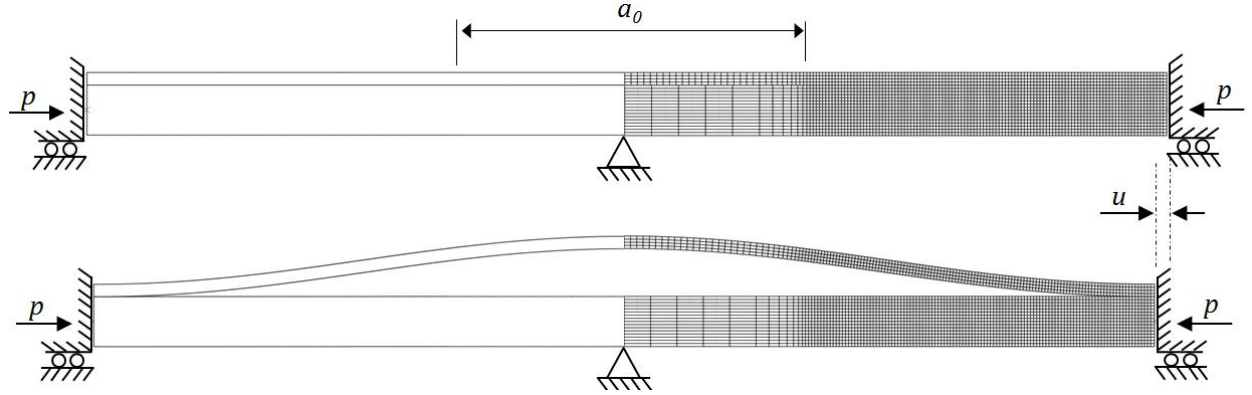


Figure 19. FEM of delamination buckling specimen at zero and maximum unscaled deformation. Mesh size adjacent to delaminated interface 0.125 mm.

The computed structural response is plotted in Figure 20, whereas the values of the solution parameters for the *crack length control scheme* are listed in Table 1.

The buckling of the upper sub-laminate led to the unstable delamination propagation that in turn disbonded a large portion of the laminate. The sudden section loss caused a very sharp snap-back instability in the equilibrium path, which was captured by the *crack length control scheme* method. Following the softening branch, the curve recovered a positive slope, which correctly shows that the laminate was capable of bearing increasing loads after the buckling of a sub-laminate. The Abaqus VCCT solution under displacement control confirmed the snap-back structural instability. Under force control the structure would have been subjected to snap-through instability, with the load-displacement jumping horizontally from the critical point to the branch of the equilibrium path on the right side of the diagram.

In order to converge to incipient growth, the size of the first four equilibrium sub-increments on the softening branch, indicated with hollow markers in Figure 20, was automatically cut down by the *arc length control scheme* method. The reason was the overshoot of the ERR convergence criterion. The overshoot was particularly large for increment A, during which buckling occurred. At the beginning of this increment in fact, before the sub-laminate buckled, the value of φ was very large because of the small value of total energy release rate ${}^{t+\Delta t}G_T$, leading to an amplification of the applied load. To reduce overshooting, a limit to the applied load was introduced in the code based on the critical column buckling load for the upper sub-laminate. The increment was cut down to 1/100 for A and to 1/10 for B-D, thereby

dividing the equilibrium sub-increments into a sequence of 100 and 10 sub-increments respectively. Increments E-H also failed the ERR convergence condition, but G_T did not overshoot G_C . In this case the increment was automatically restarted with an updated φ value but the solution was again attempted in one increment, as described in the previous section. All the growth sub-increments converged at the first attempt. Furthermore, the *crack length control scheme* did not require the use of stabilization techniques, whereas contact stabilization was required for the Abaqus VCCT solution. The settings for the contact stabilization were the same as the SLB example.

The solution of the constraint equations (64) provided large values of Δa , that always exceeded the specified Δa_{max} by more than an order of magnitude. The reason for the error that affected the crack length prediction was the nonlinear structural response caused by the elastic instability and the subsequent large displacements of the upper sub-laminate. The use of analytic sensitivity to compute the load and compliance derivatives led to particularly large errors because of the combined effect of the analytic models that assumed linear elastic structural response, and of the linearization performed by the forward Euler method used to derive equations (64). The numeric sensitivity performed slightly better because only the second effect was present, but the computed values for Δa were still affected by a too large error to be utilized in the arc-length control. Also the nonlinear change in the delamination mode mixity caused by the local buckling contributed to the arc-length control failure. To cope with this shortcoming, the algorithm was modified so that a constant delamination length increment, equal to Δa_{min} , was prudentially used if Δa was larger than $2\Delta a_{max}$ or in case of numerical divergence in the solution of equations (64).

Despite the cut down of the increments and the inability to control the arc-length, the *arc length control scheme* required 293 FNR iterations to solve the problem, compared to the 1698 iterations of the Abaqus VCCT with a single node released per increment and the 612 iterations with multiple nodes released per increment. Similar results were obtained using the BFGS method, which showed about the same computational efficiency of the FNR method.

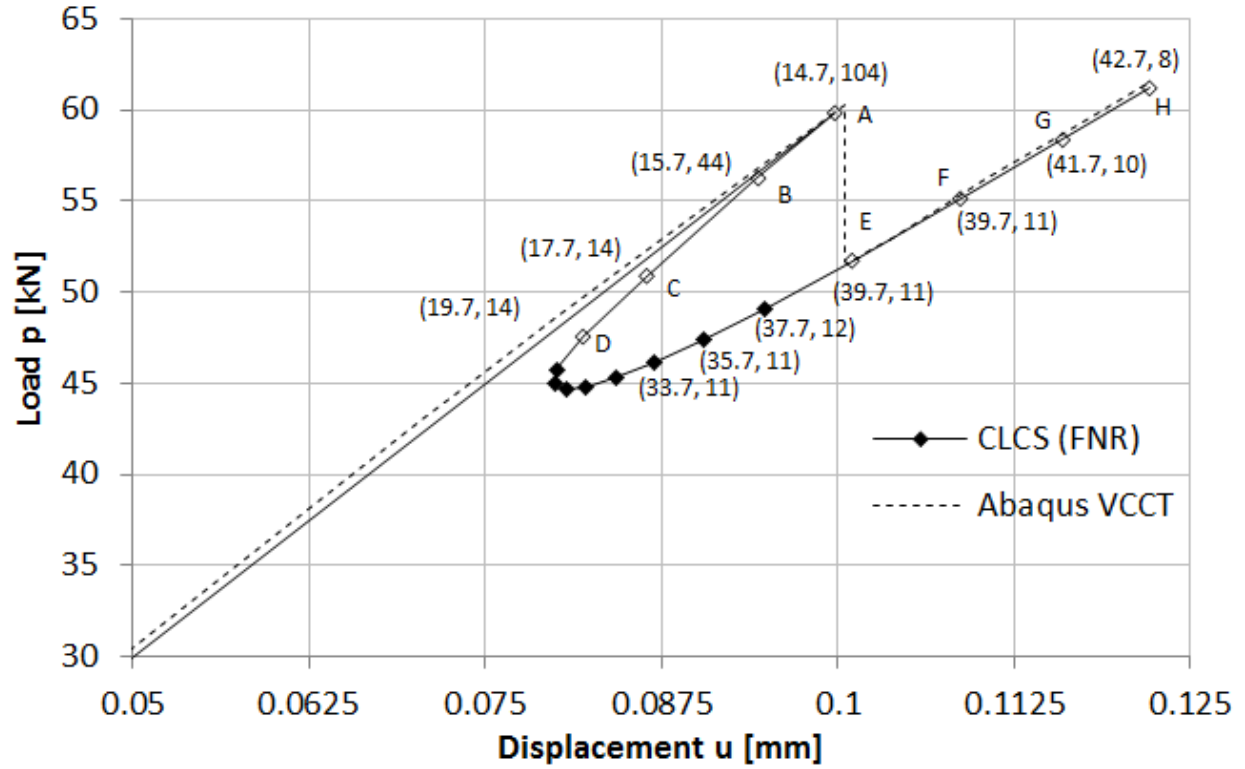


Figure 20. Crack length control scheme (CLCS) and Abaqus VCCT solutions of delamination buckling. Curve markers indicate converged configurations. The delamination length and the number of iterations required to converge are indicated in parenthesis.

3.7 Conclusions

The *crack-length control scheme* proved to be effective in capturing the sharp snap-back instability of the structural response that characterizes the brittle failure mode of laminated structural composites. The method provides the envelope of stable and unstable delamination growth and delamination buckling collapse responses by tracing the complete equilibrium path of the structure with continuation through singular points.

Although it does not guarantee convergence, the *crack-length control scheme* increased the convergence rate and robustness of the Newton-Raphson method in the benchmark assessment presented. The primary reason for the increased computational efficiency was the capability of advancing the delamination by multiple elements, thereby reducing the number of iterations required by a conventional implementation of the VCCT method. Moreover, by extending the delamination under constant applied displacement

intervals, the unstable propagation of the delamination could be computed without the use of an artificial stabilization technique. Also the use of the delamination length as the constrained variable instead of nodal displacements contributed to the robustness of the method. In fact, since the delamination length is monotonically increasing with time, the user can reasonably establish, based on the dimensions of the structure under consideration, an upper and lower limit to the incremental variation (Δa) of the delamination length. These limit values can be utilized in place of the values computed by the constraint equations to overcome inconsistencies in the arc-length control procedure that could arise in case of a highly nonlinear structural response such as delamination buckling.

The main drawback of the *crack length control scheme* consists in the dependency of the constraint equations to the crack topology, i.e. to the element connectivity. As a result, the delamination has to be tracked constantly during the solution process, leading to a cumbersome code addition to a finite element solver. This requirement limits the applicability of the proposed method to bi-dimensional problems. In its current implementation, the method is also not compatible to multi-delamination problems and the possibility to upgrade it has to be assessed.

Possible applications of the method include automated parametric studies with delamination propagation, featuring multiple materials sets with widely varying material properties, like the CFRP-TFB laminate study. A more general application consists in the preliminary analysis of complex structures that can be idealized with a bi-dimensional finite element discretization. When the structural response is unknown a priori and therefore considerable judgment by the user may be required, knowing the full equilibrium path a priori would enable immediate assessment of the correctness of the structural response computed by commercially available software. Furthermore the structural response associated with any initial crack length could be inferred without additional analyses.

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4. Analysis of the multifunctional laminates

4.1 Summary

Stress analysis was employed to determine the critical locations for delamination onset and the *crack length control scheme* analysis method was applied to the simulation of the mechanical tests utilized to experimentally characterize the TFB-CFRP laminates. The results of the progressive failure analysis of several potentially critical delaminations were compared to the experimental results in order to develop a better understanding of the failure mechanism, determine the critical ones and validate the analysis tools. Finally, a numerical parametric study based on finite element analysis allowed to trace performance plots for optimum laminate design.

4.2 Introduction

The TFB-CFRP laminate is a novel problem in the analysis of composite structures. It shows geometrical similarities to typical designs that were analyzed by a number of authors, such as ply drop [1, 2], embedded defect, tiled composites [3]. The literature relevant to hybrid composites [4, 5] and bi-material and tri-material bonded joints [6] should also be taken into account for the mechanical analysis of a laminate that is characterized by the presence of multiple constituents with inhomogeneous material properties. However, unlike any other structural composite material system, the TFB-CFRP laminate is comprised of layers with widely varying mechanical properties. While the relation between these properties and the overall structural efficiency are obvious, their effects on the strength and damage tolerance of the multifunctional laminate are unknown and unintuitive.

Another peculiarity of the analysis of the TFB-CFRP laminate consists in the specific mechanical failure modes that lead the electrical failure. The local delamination buckling of the packaging layers, for example, leads to contamination of the active components (type II failure). Moreover, the delamination

between the active components or between the active components and the lower substrate, where the current collectors are, leads to electrical discontinuity. Both cases result in the electrical failure.

The objective of the numerical analysis presented hereinafter was to determine the mechanical properties of the single constituents that ensured the integrity of the laminate under static loading. Based on this fundamental knowledge, a preliminary design study of a TFB-CFRP laminate could be performed and the achievable structural efficiency in terms of specific strength and specific apparent modulus could be assessed.

The conventional analysis approach applied to composites materials is comprised of experiments to determine the critical failure modes, followed by stress analysis for determining the location of the first matrix cracking, and finalized by the progressive failure analysis. Within this framework, the experimental characterization presented in chapter 2 [7], which was enabled by the materials and processing compatibility study reported in chapter I [8], provided evidence that unstable delamination propagation at the CFRP/lower substrate and active components/sealant interfaces were among the critical failure modes for the externally bonded TFB configuration. The stress analysis and computational fracture mechanics analysis reported hereinafter were conducted in order to provide a theoretical explanation of the experimental evidence and to determine the critical failure modes for the embedded battery configuration, which were still unknown. Upon successful matching of the experimental results, performance plots under varying constituents materials were traced for the TFB-CFRP preliminary design.

The progressive failure analysis was divided into two phases: experimental match and parametric study. In the first phase the crack length control scheme (see chapter 3) was utilized to compute the delamination characteristics at all the critical interfaces and compare the results to the experimental results. In the parametric study the distributions of G_I , G_{II} and G_C were traced under constant ultimate loads for a set of different design variables. The results allowed tracing a design space for materials moduli and fracture toughness. The main motivation of this approach was driven by the need to understand the influence of the elastic moduli of the packaging materials on the fracture parameters. This knowledge would enable to

functionally grade the TFB substrate and sealant in order to relax the fracture toughness requirement for the active components. In fact, developing materials with high fracture toughness seems to be the most difficult design challenge for the state-of-the-art of nanostructured electrode and electrolyte materials. This study would provide design guidelines for the next generation of airborne TFB-CFRP laminates. For simplicity, the parametric study was performed without varying the geometry and loading conditions, which were the same as the experimental test campaign.

Due to the preliminary design nature of this study, in all the phases of the analysis the laminates were idealized through bi-dimensional models to reduce the modeling time and increase the computational efficiency. Maximizing the computational efficiency was necessary because the localized nature of the delamination analysis, together with the multiscale structure of the TFB-CFRP laminate (i.e. small relative thickness of the active components), required fine finite element meshes

The FE models of the mechanical tests described in chapter 2 consisted of laminate longitudinal sections discretized with plane strain elements. This simplification did not allow to capture mode III fracture and anticlastic bending, which were considered negligible. Also the TFB longitudinal edges were considered not critical for delamination initiation and propagation. The plane strain assumption overconstrained the displacements by not allowing out-of-plane displacement of the longitudinal section. Out-of-plane displacement would be particularly important in the proximity of angle plies due to the high localized Poisson's ratio caused by the scissoring shear affect. Since these displacements lead to mode III loading of the crack tip, it would not be conservative to neglect them. However, we did not consider any case of delamination adjacent to an angle ply, therefore this contribution was neglected.

By defining a Cartesian coordinate frame that features the $z - axis$ normal to the longitudinal section and the $x - axis$ parallel to the longitudinal direction of the specimen, the plane strain condition assumed that ε_z , γ_{xz} and γ_{yz} are equal to zero. Similarly, plane stress would assume that σ_z , τ_{xz} and τ_{yz} are equal to zero. The first assumption approximated well the strain field that occurs at the specimen centerline and the error introduced is more marked if angle plies are present because of their high Poisson's ratio. The second assumption does not constrain the displacement in the transverse direction, similarly to what

happens at the free edges of a panel. Plane stress and plane strain can therefore be considered two extremes, whereas a three-dimensional model would provide an intermediate response between the two. In particular, results from plane strain analyses may not be regarded as reliable near the free edges of the specimen, nor can the results from plane stress analysis be interpreted as representative of the center of the specimen [9]. Furthermore, the three-dimensional stress-strain field caused by the section change of the specimen, determined by the presence of the TFB, was considered a secondary effect and was also neglected.

Another assumption was that because of the high displacement constraint, the plastic deformation was confined to a small local region adjacent to the crack tip. This implied a high cohesive strength in the model and in such circumstances fracture is governed by a single parameter, G_C . In other words, small scale yielding (SSY) was assumed. This allowed to model the delamination propagation by applying the LEFM through the VCCT. This assumption was conservative and therefore appropriate for a preliminary design. It was conservative because yielding in the fracture process zone would reduce the strain energy released upon crack advance, leading to a lower ERR.

Geometric nonlinear FEA was employed to solve the crack surface and contact creation within the laminate. Incremental solution was also necessary to model the crack growth. A large displacement formulation was always selected in order to correctly compute the large rotations achieved in the 4PB and delamination buckling cases.

The materials were all linear elastic. Therefore the results of the parametric study are applicable to materials that have a linear elastic response within a maximum strain of about 5% (small strains). Material that show a hyperelastic response, such as rubber materials, were not considered as potential sealant materials for the TFB. To obtain correct results for such materials, their small compressibility ($\nu > 0.475$) should be modeled correctly, in particular when the material is highly confined such as in a plane strain condition.

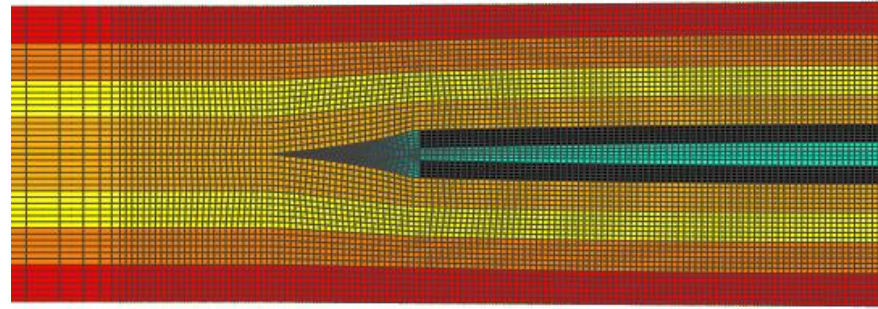
The dependence of G_C on temperature was not considered because it was experimentally demonstrated that the TFB generated negligible heat. This was assessed by monitoring the surface temperature of a battery with an infrared (IR) camera in the worst case of a short circuit.

4.3 High fidelity modeling

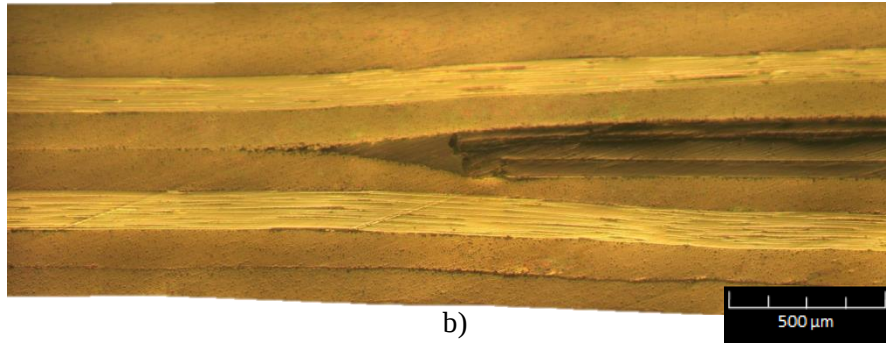
A realistic TFB termination geometry and smooth ply and resin pocket curvatures were generated at the TFB ends. Several geometrical parameters were adjusted in order to reproduce the transition observed in the micrographs of the sections, Figure 1.

For the embedded configuration, as shown in Figure 2, the TFB thickness dropped from the nominal thickness of 0.2 mm to 0.16mm at TFB end. This thickness reduction took place in a span of 1.6 mm through an arc that is tangent to the TFB outer surface. The resin pocket was 0.5 mm long from the TFB end to the pocket vertex. In order to avoid a singular pocket vertex a line inclined by 6 degrees from the horizontal and 0.1 mm long was used to approximate the pocket vertex. An arc tangent to this line and passing through the TFB end corner was then drawn and cut at a distance of 0.07 mm from the TFB end (yellow dotted line). Another arc tangent to the TFB outer surface and passing through the endpoint of the previous arc was drawn. The connection between the two curves was smoothed out by a 1 mm radius fillet.

The ply interfaces located before the resin pocket were parallel and equally spaced as per their nominal thickness of 0.125 mm. On the other hand, the first three ply lines adjacent to the TFB were created by offsetting the TFB outer surface curve, Figure 3. The offset values for these three curves, starting from the TFB surface, were 0.1, 0.1 and finally 0.105. Starting from the fourth ply, only the straight portion of the ply curves was offset by the following values: 0.11; 0.115; 0.12; 0.125. The last offset value was repeated up to the outer laminate surface (the 0.13 mm quoted in the picture is incorrect). The curves were then connected with straight lines. Fillets with 2 mm radii were finally created at the corners indicated in picture.



a)



b)

Figure 1. Embedded TFB configuration. a) High fidelity FEM of TFB termination. b) Micrograph of TFB termination.

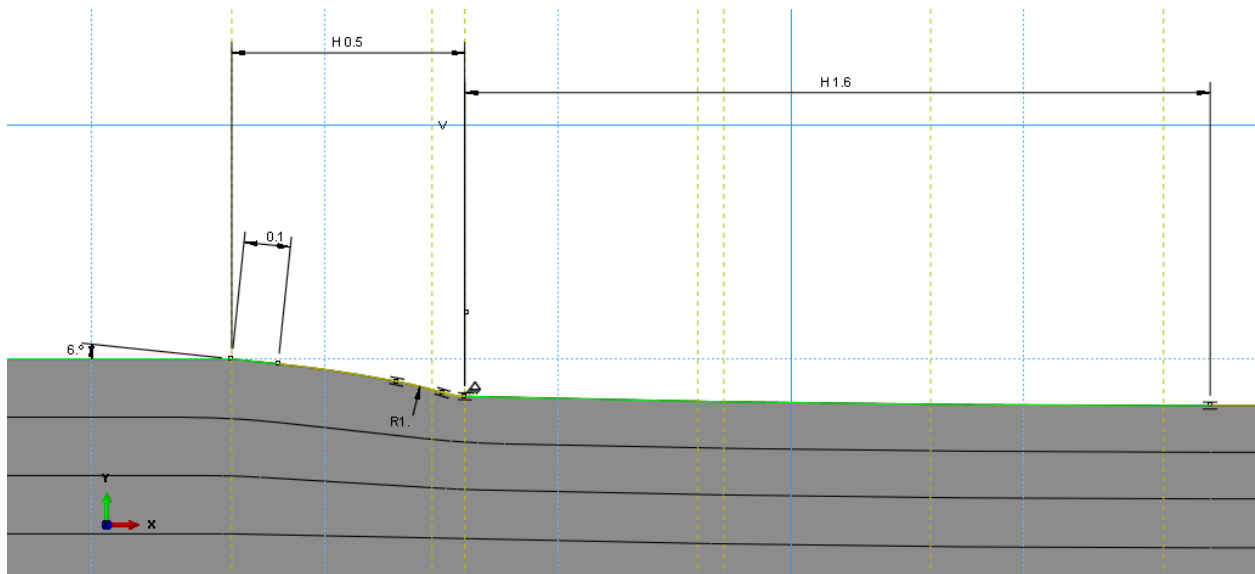


Figure 2. Geometrical description of TFB tapered end and resin pocket.

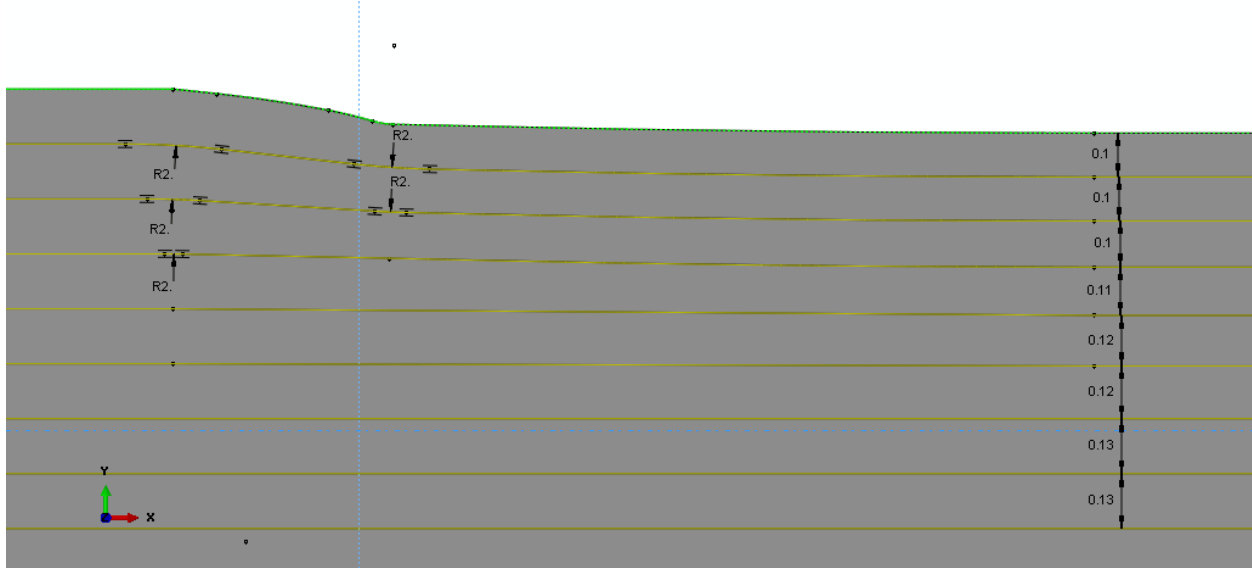


Figure 3. Geometrical description of ply interfaces at TFB termination.

The externally bonded TFB was modeled without tapered thickness, but with a triangular resin ramp at the TFB ends. The ramp was 0.29 mm high, corresponding to TFB thickness plus the thickness of the adhesive film, and 0.5 mm long, Figure 4.

The finite element mesh employed only quadrilateral elements. For the stress analysis, eight noded plane strain elements with quadratic interpolation functions and reduced (2x2) integration scheme (CPE8R) were employed. The baseline mesh refinement featured an element length of 0.02 mm in the critical zones, coarsening gradually to 0.2 mm at the extremities of the specimen. Each CFRP ply was modeled with 6 elements through the thickness, whereas TFB substrate of sealant was modeled with 5 elements. This choice of baseline mesh refinement was selected after various mesh sensitivity studies with up to 10 elements through the thickness of each ply and element lengths down to 0.01 mm.

The discretization utilized for the delamination propagation analysis for the experimental match and parametric study was coarser to increase computational efficiency. The mesh size adjacent to the delaminated interfaces was maintained equal to 0.02 mm, but the elements through the thickness of the TFB packaging layers were reduced to 3 and to a maximum of 7 and a minimum of 1 for the CFRP plies, with gradual mesh size increase with the distance from the delaminated interfaces, Figure 4. The number

of degrees of freedom was also reduced by utilizing four noded linear elements with reduced integration (CPE4R). In any case, the mesh was structured so that the aspect ratio (length to width) for the elements always ranged between 0.1 and 10. All the bi-material interfaces were modeled with matched nodes for a more accurate computation of the ERR.

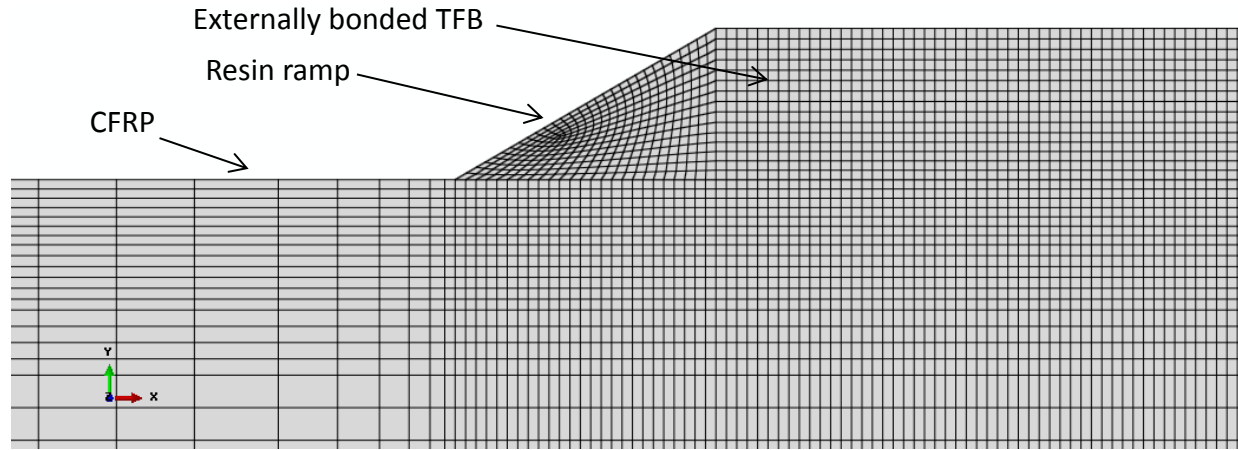


Figure 4. Externally bonded TFB configuration. Close-up on mesh at battery termination.

For all the models, the active components were homogenized in one layer with uniform mechanical properties of pure Lithium, Table 1. The reason for this simplification depended on the consideration that the delamination characteristics of a crack propagating through the active component were not substantially different from a delamination propagating between the active components and the sealant. This consideration was based on the fact that the thickness of the active components was ten times smaller than the packaging layers, therefore the loading of the crack tip elements did not differ much from an interface to the other. The optimum fracture toughness values computed by the parametric study for the interface between the active components and the sealant can therefore be applied also to the interfaces between electrodes and electrolyte. Moreover, the thickness of the active components was increased from the nominal 0.01 mm to 0.02 mm to increase the mesh size and reduce the number of degrees of freedom. All the relevant mechanical material properties are listed in Table 1. These properties are defined in the material coordinate system, which is defined by rotating the model coordinate system defined above by

90° around the $x - axis$. As noted in the table, the values for the exponent η of the BK law were assumed to the equal to 2 for all the materials.

Property	Symbol	Muscovite ¹⁾	Surlyn	IM7/977-3 ²⁾	977-3 ²⁾	AF 163-2 ³⁾	Lithium ¹²⁾
Tensile modulus of elasticity [GPa]	E_1	161	0.28 ⁴⁾	162	3.79 ¹¹⁾	1.10	1.9
	E_2	161	0.28 ⁴⁾	8.34	3.79 ¹¹⁾	1.10	1.9
	E_3	56.8	0.28 ⁴⁾	8.34 ¹⁰⁾	3.79 ¹¹⁾	1.10	1.9
Shear modulus of elasticity [GPa]	G_{12}	14.1	0.11 ⁵⁾	4.96 ¹⁰⁾	1.46 ⁵⁾	0.41	0.73 ⁵⁾
	G_{13}	14.1	0.11 ⁵⁾	3.11 ¹⁰⁾	1.46 ⁵⁾	0.41	0.73 ⁵⁾
	G_{23}	65.2	0.11 ⁵⁾	4.96	1.46 ⁵⁾	0.41	0.73 ⁵⁾
Poisson's ratio	ν_{12}	0.27	0.3 ⁷⁾	0.34 ¹⁰⁾	0.3 ⁷⁾	0.34	0.3 ⁷⁾
	ν_{13}	0.24	0.3 ⁷⁾	0.34	0.3 ⁷⁾	0.34	0.3 ⁷⁾
	ν_{23}	0.10	0.3 ⁷⁾	0.34 ⁷⁾	0.3 ⁷⁾	0.34	0.3 ⁷⁾
Strain to tension failure [%]	ε_1^{TU}	unkn.	8.00 ⁴⁾	1.46	unkn	unkn.	unkn.
	ε_2^{TU}	unkn.	8.00 ⁴⁾	0.77	unkn	unkn.	unkn.
Mode I fracture toughness [J/m ²]	G_{IC}	1.30 ⁸⁾	1200 ⁹⁾	316	217	3682	unkn.
Mode II fracture toughness [J/m ²]	G_{IIC}	unkn.	4600 ⁹⁾	579	unkn	unkn	unkn.
BK law exponent ⁷⁾	η	2	2	2	2	2	unkn.

¹⁾ Engineering constants calculated by approximating the monoclinic stiffness matrix in McNeil et al., *J Phys.: Condens. Matter* 5, 1681 (1993) to a transversely isotropic stiffness matrix relative to the 1-3 plane.
²⁾ Cytec IM7/977-3 data sheet.
³⁾ 3M Scotch-Weld Structural Adhesive Film AF 163-2 data sheet.
⁴⁾ Yield strain, ref. NCAR Report FRB-4 (1965).
⁵⁾ Calculated assuming isotropy and $\nu = 0.3$.
⁶⁾ Calculated assuming isotropy in the battery plane.
⁷⁾ Assumed.
⁸⁾ Hill et al., *Int. J. Fracture* 119/120, 365 (2003).
⁹⁾ Compston et al., *J Mater. Sci. lett.* 20, 509 (2001).
¹⁰⁾ Calculated assuming transverse isotropy relative to the 2-3 plane.
¹¹⁾ Flexural modulus
¹²⁾ 92.5% Li-7, 7.5% Li-6. Schultz R., Fermilab-TM-2191 (2002).

Table 1. Material properties defined in the material coordinate system.

4.4 Stress analysis

The stress analysis was performed using the nonlinear static implicit solver Abaqus/Standard. A full Newton-Raphson method was utilized to iteratively solve the equilibrium equations with an updated Lagrangian formulation to account for large displacements. The nonlinear solver was preferred to a linear analysis because geometric nonlinearities occurred due to the large displacement experienced by the bending tests, and not negligible stiffening membrane effect at TFB location occurred for the externally bonded configuration under any loading condition. This geometric nonlinear behavior was confirmed by other authors for similar bonded joints configurations such as skin/stiffener structures [10].

The laminate configurations considered in the analysis were the four point bending tests of the quasi-isotropic laminates $[(0/45/90/-45)_3/\text{TFB}/(-45/90/45/0)_3]$ and $[(0/45/90/-45)_{3S}/\text{AF}/\text{TFB}]$ for the embedded and externally bonded case respectively, and the corresponding uniaxial tension configurations $(0/45/90/-45/\text{TFB}/-45/90/45/0)$ and $[(0/45/90/-45)_S/\text{AF}/\text{TFB}]$. The models and boundary conditions are illustrated in Figure 5 and Figure 6. The analysis of the combined curvature and tension case for the externally bonded battery was omitted. While the bending specimens were modeled according the actual dimensions in order to correctly apply the loading, the uniaxial tension specimens were shortened to 50.8 mm for the sake of computational efficiency. Only half specimen was modeled for all the load cases because of symmetry. The applied loads were the critical loads experimentally determined in chapter 2.

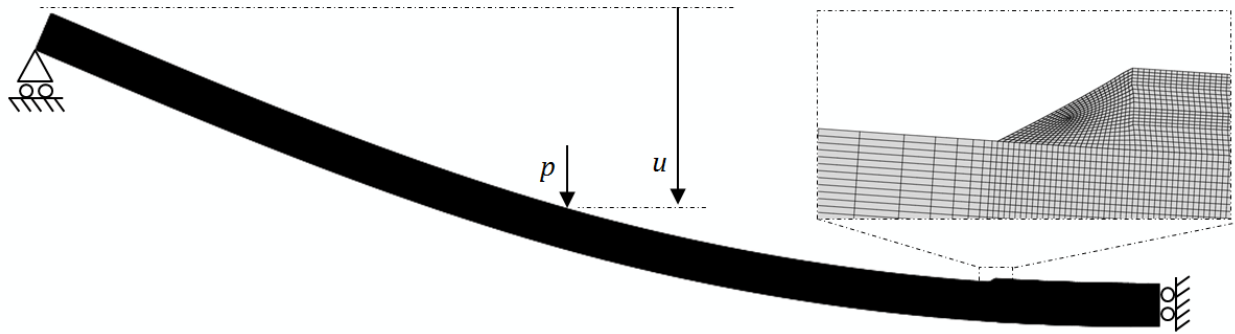


Figure 5. FEM of four point bend test of configuration $[(0/45/90/-45)_{3S}/\text{AF}/\text{TFB}]$ for stress analysis.

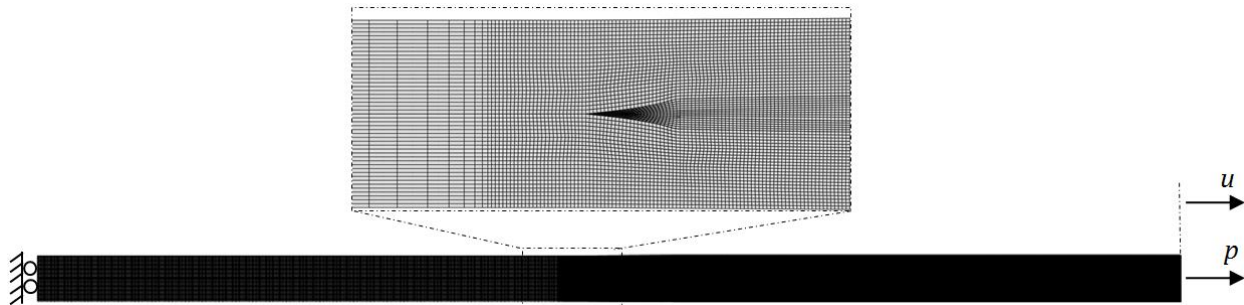


Figure 6. FEM of uniaxial tension test of configuration $(0/45/90/-45/\text{TFB}/-45/90/45/0)$ for stress analysis.

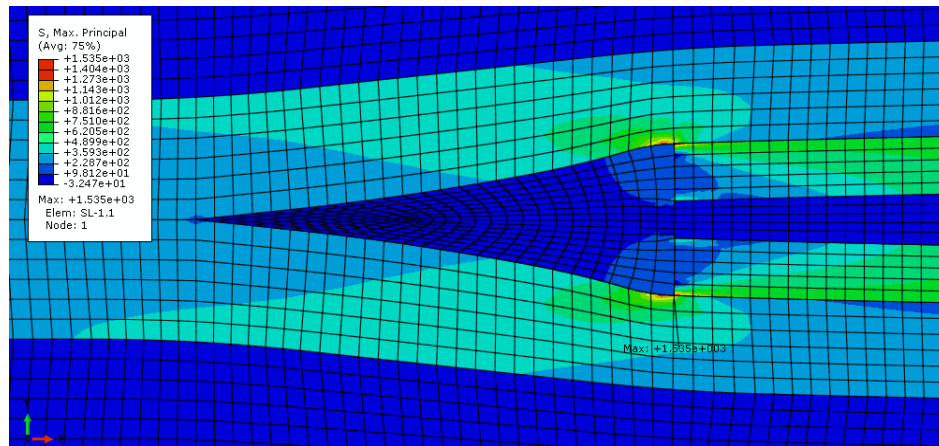
The locations for delamination onset were determined by comparing the results from three failure criteria: the Hashin-Rotem [3]; Puck [11] and the maximum principal stress transverse to the fiber direction [10].

The first two criteria allow to consider also fiber failure. However, only the matrix cracking margin of safety was considered at this time. According to the stress analysis, the critical locations for delamination onset for the embedded TFB were the CFRP/substrate interface and the substrate/sealant interface at the TFB edges transverse to the specimen longitudinal axis, Figure 7. These potential delaminations are identified as *CFRP/sl* and *sl/s-out* respectively. This result confirmed the findings of similar analyses conducted on ply drops by other authors. Experimental results available in literature showed that the delamination onset as a tension crack between the dropped ply and the adjacent resin pocket, at then the crack turns by 90 deg and becomes a delamination, whereas the vertex of the resin pocket is not critical [2]. For the externally bonded configuration, similar results were obtained and the resulting critical locations were the CFRP/adhesive film, the adhesive film/substrate and the substrate/sealant interfaces at the TFB ends. These potential delaminations are identified as *CFRP/af*, *af/sl* and *sl/s-out* respectively.

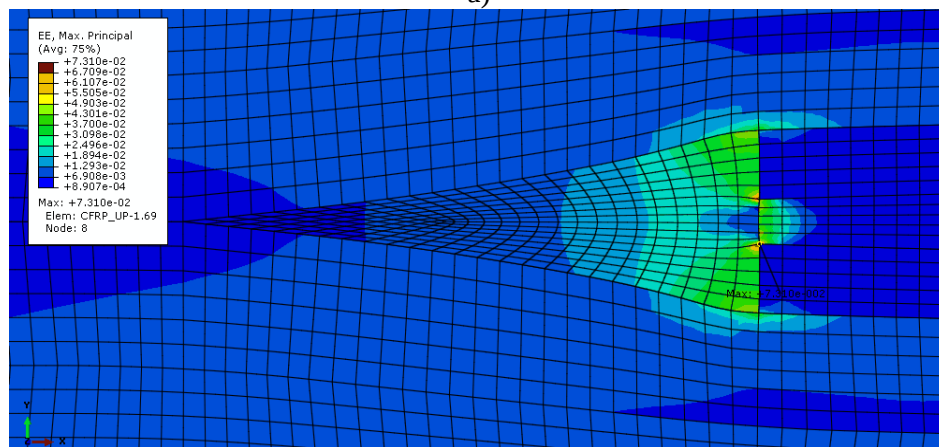
Bases on the experimental evidence of existing disbonded fronts at the edges of the active components, to critical location were added to the progressive failure analysis. These delaminations are identified as *sl/s-in* for the interface between the lower substrate and the sealant, and as *ac/s* for the interface between the active components and the sealant. The summary of all critical location is illustrated in Figure 8 for the embedded configuration. In the following section the delamination propagation analysis was applied at these locations.

The normal strain distribution for the embedded configuration is shown in Figure 9 for an applied far field strain of 6375 μ strain, which corresponded to the strain at electrical failure. The uniform applied strain field showed to be altered by the resin pocket and the TFB termination. The strain concentration was localized but significant. The normal x-strain ε_x peaked to a 6% strain in the resin pocket due to the low matrix modulus. Moreover, ε_x in the battery remained lower than the far field strain for about 1.5 mm from the TFB, before the section recovered strain uniformity. The strain ε_y was negative everywhere, as expected, except in the sealant in proximity of the TFB end. This observation is important for the delamination propagation analysis reported hereinafter because it affects the critical region for onset and

propagation and because the unexpected positive ε_x excited mode I fracture when the laminate was subjected to uniaxial tension.



a)



b)

Figure 7. Uniaxial tension. a) Maximum principal stress. b) Maximum principal strain.

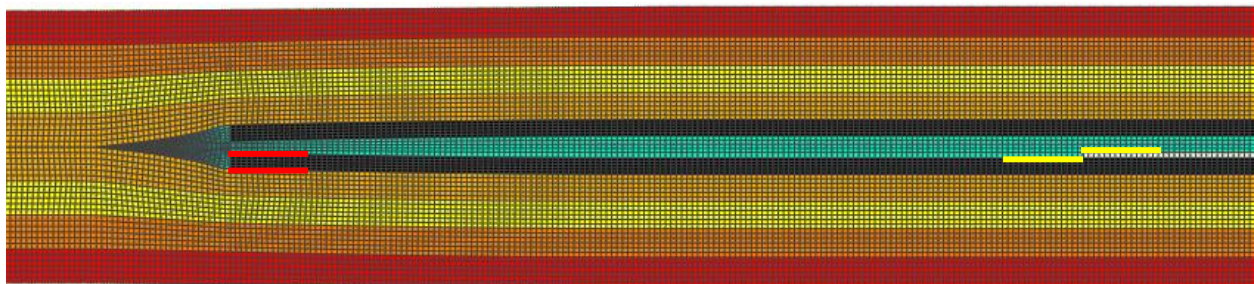
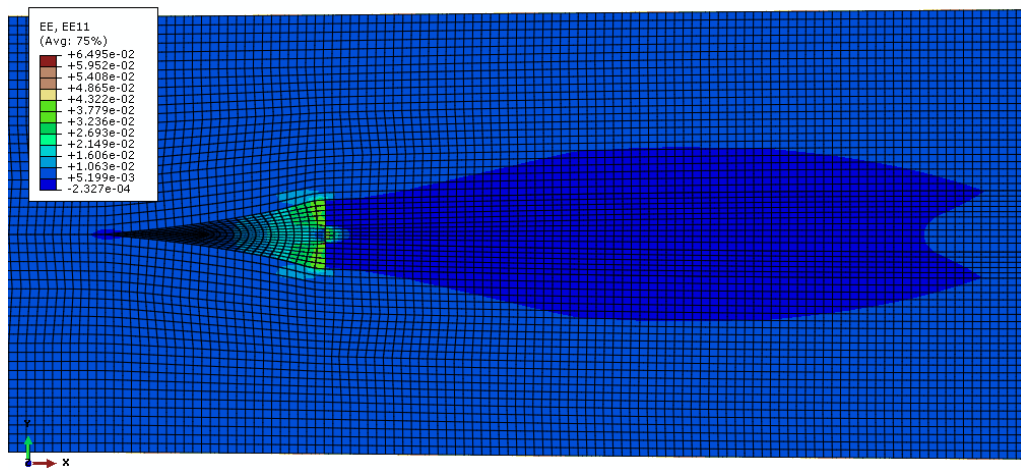


Figure 8. Embedded configuration. Critical delamination onset locations by stress analysis (red lines) and experimental evidence (yellow lines).

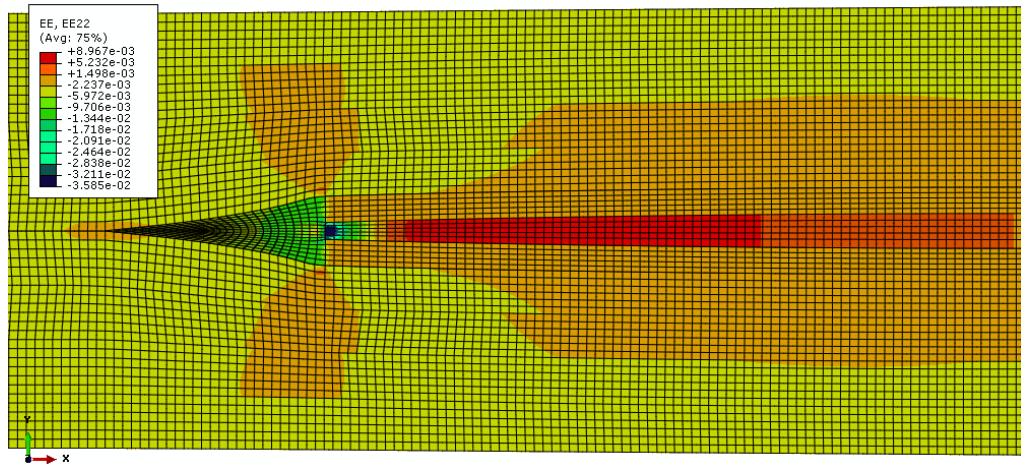
In order to develop a better understanding of the of the stress-strain field, let us define the battery coordinate system x_{TFB} and four transverse sections as illustrated in Figure 10. The displacement, strain and stress distributions at the four sections are shown in Figure 11 - Figure 15. The data were computed at node locations. Stress and strain corner values were averaged only for elements having the same material properties and for values that differed for less than 75%. Therefore at material interfaces the contribution relevant to different elements were not averaged and both values were plotted. Thus, at bi-material interfaces there are discontinuities in the stress and strain distributions due to the different derivatives of the shape functions at the element corners.

The uniform far field displacement was perturbed and non-uniform displacements were computed in the battery and in the CFRP laminate in proximity of the TFB termination. The section did not remain flat, as in pristine laminate, and the perturbation extended all through the thickness of the laminate and regained uniformity at about 2.4 mm away from the TFB termination, Figure 11. The local softening caused by the low modulus of the resin pocket and the sealant led the strain ε_x to drop dramatically in the substrates at the battery end, Figure 12. This caused a sharp perturbation in the strain distribution at section $a - a$. As a results the normal stress σ_x in the substrates was, on average, close to zero at the TFB end, but with peaks at the $CFRP/sl$ interface where load transfer between the CFRP plies and the packaging took place. The stress in the substrates increase progressively and reached a uniform distribution at about 2.4 mm from the TFB end. At this section, the stress σ_x recovered uniformity within all the layers, Figure 13. The load transfer from the CFRP plies to the battery took place by shear, as demonstrate by the peaks in the shear strain γ_{xy} at the $CFRP/sl$ and ls/s interfaces at the TFB end, Figure 14. The shear strain in concentrated in the 45° ply adjacent to the battery and in the substrates. Its distribution within these layers is not uniform and nonlinear, reaching peak values at the aforementioned interfaces. The shear stress reached the highest peak value of 411 MPa in the 45° CFRP ply at section $a - a$, Figure 15. Hence, the embedding of the TFB within the composite laminate led to a three-dimensional stress-strain field in proximity of the TFB ends, as described above. The main effect of the TFB termination was to

concentrate the load path at the center of the laminate, as shown by the peak in ε_x in the ply adjacent to the battery (Figure 16), in order to transfer the load to the TFB substrates. Thanks to the absence of a soft adhesive film and to the load transmission taking place through both the upper and lower substrate, the shear lag region was shorter than in the externally bonded battery configuration. In fact, the strain ε_x in the CFRP ply and in the substrate converged to the same value at about 3 mm distance from the TFB ends, Figure 16. Therefore the shear lag did not involve the active components.



a)



b)

Figure 9. a) Normal x-strain ε_x and b) normal y-strain ε_y . Applied far field strain $\varepsilon_x = 6375 \mu\text{strain}$. Uniaxial tension.

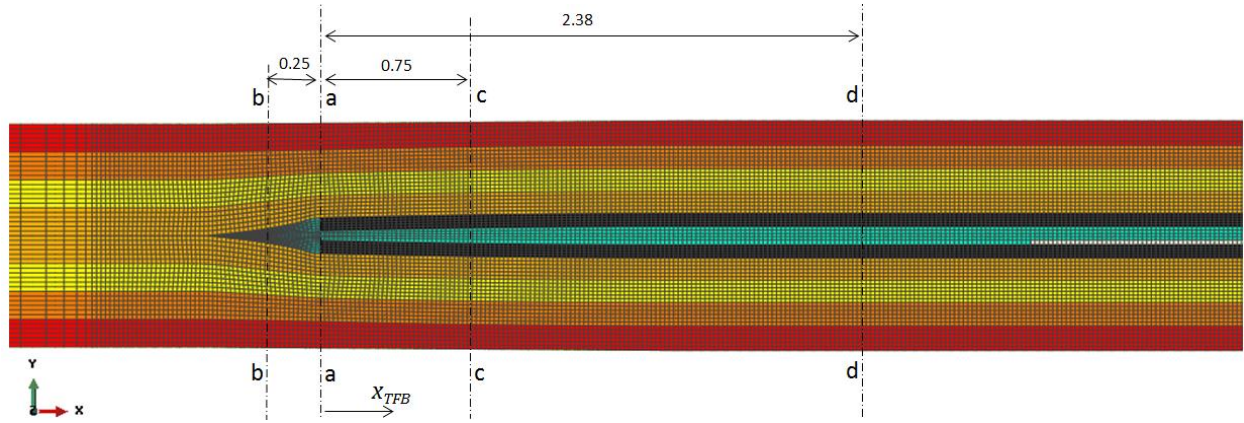


Figure 10. TFB coordinate system x_{TFB} and sections definition.

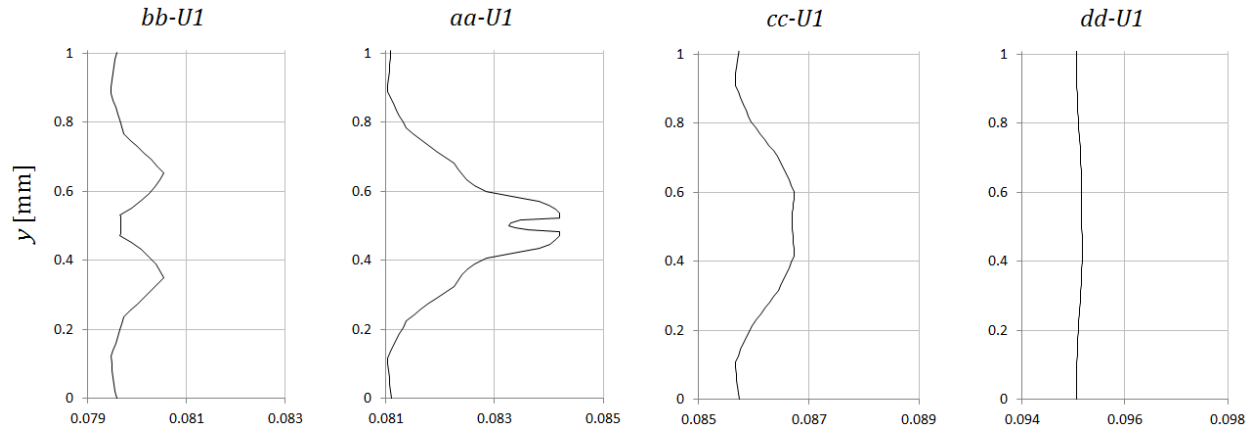


Figure 11. Nodal x -displacements $U1$ [mm] at transverse sections. Uniaxial tension.

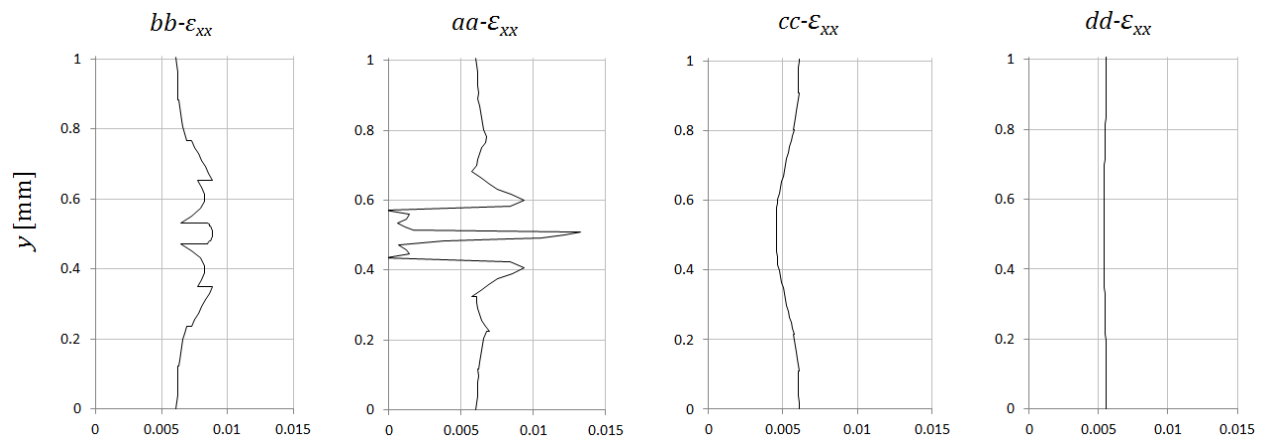


Figure 12. Strain ϵ_x at transverse section. Applied far field strain $\epsilon_x = 6375 \mu\text{strain}$. Uniaxial tension.

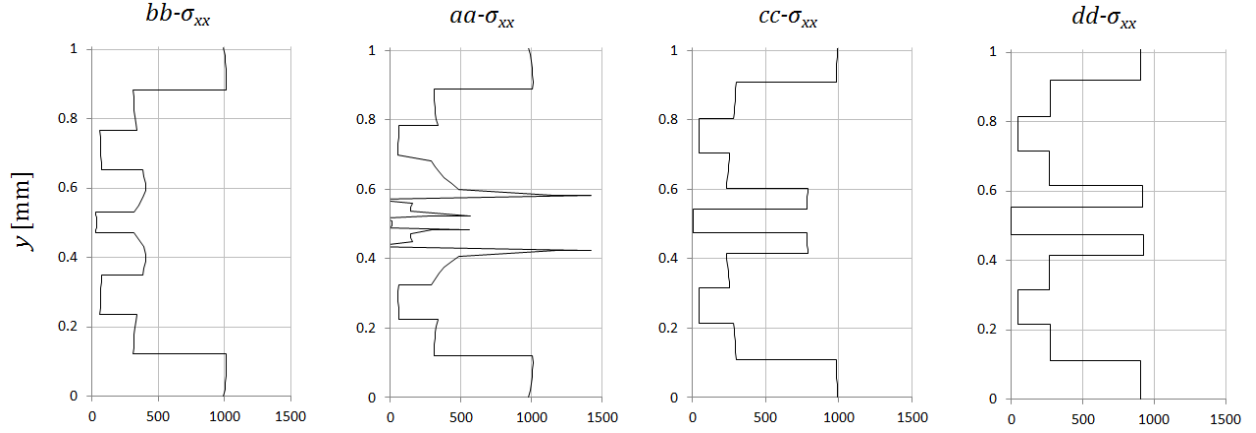


Figure 13. Stress σ_x at transverse sections. Applied far field strain $\epsilon_x = 6375\mu\text{strain}$. Uniaxial tension.

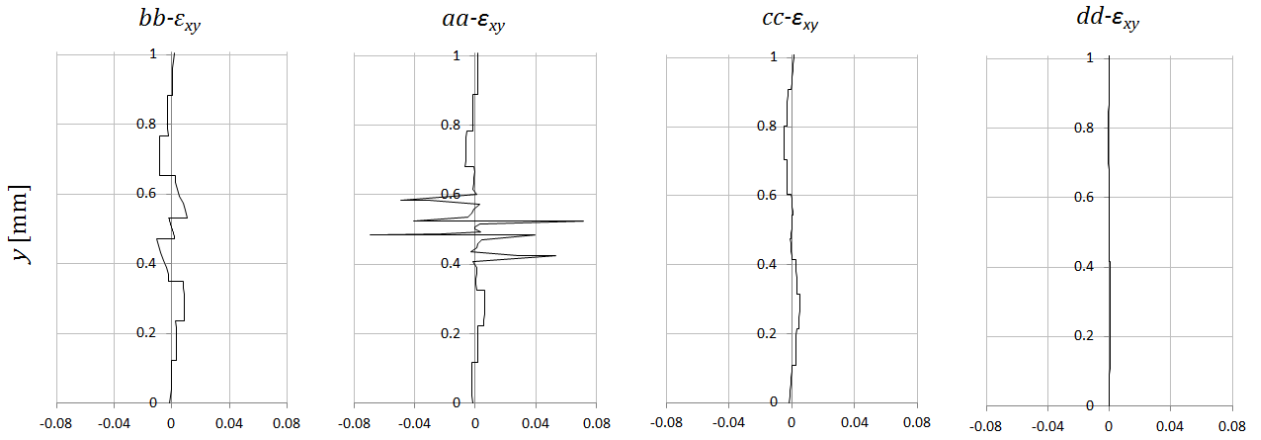


Figure 14. Shear strain γ_{xy} at transverse sections. Applied far field strain $\epsilon_x = 6375\mu\text{strain}$. Uniaxial tension.

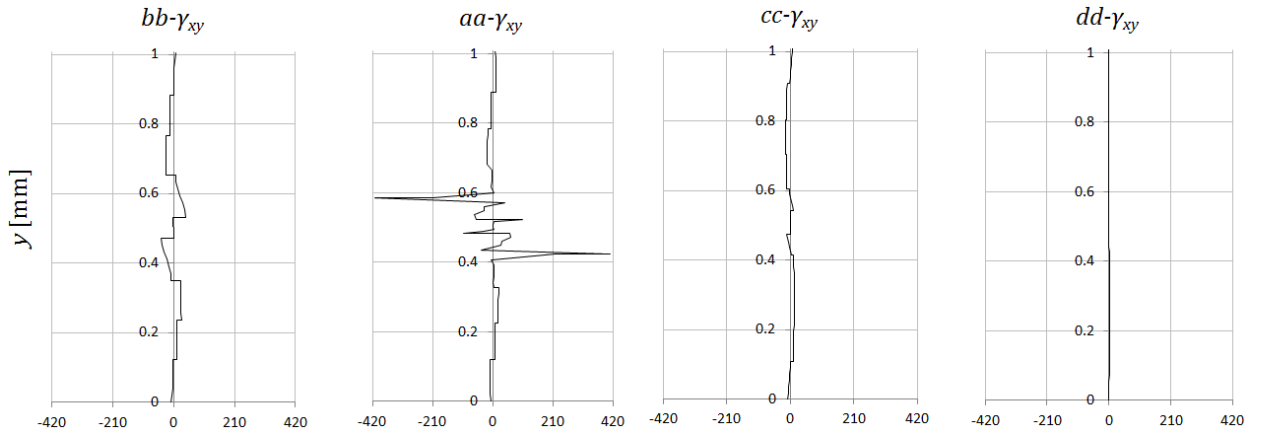


Figure 15. Shear stress τ_{xy} at transverse sections. Applied far field strain $\epsilon_x = 6375\mu\text{strain}$. Uniaxial tension.

Similar conclusions could be drawn for the externally bonded configuration under uniaxial tension, therefore the results were omitted. These results were in agreement with the shear lag elasticity solution described in chapter 2, showing extensive transverse shearing of the sections concentrated in soft layers (adhesive film and sealant). Moreover, the shear lag region extended well over the active components, reaching a distance of 13 mm from the TFB end.

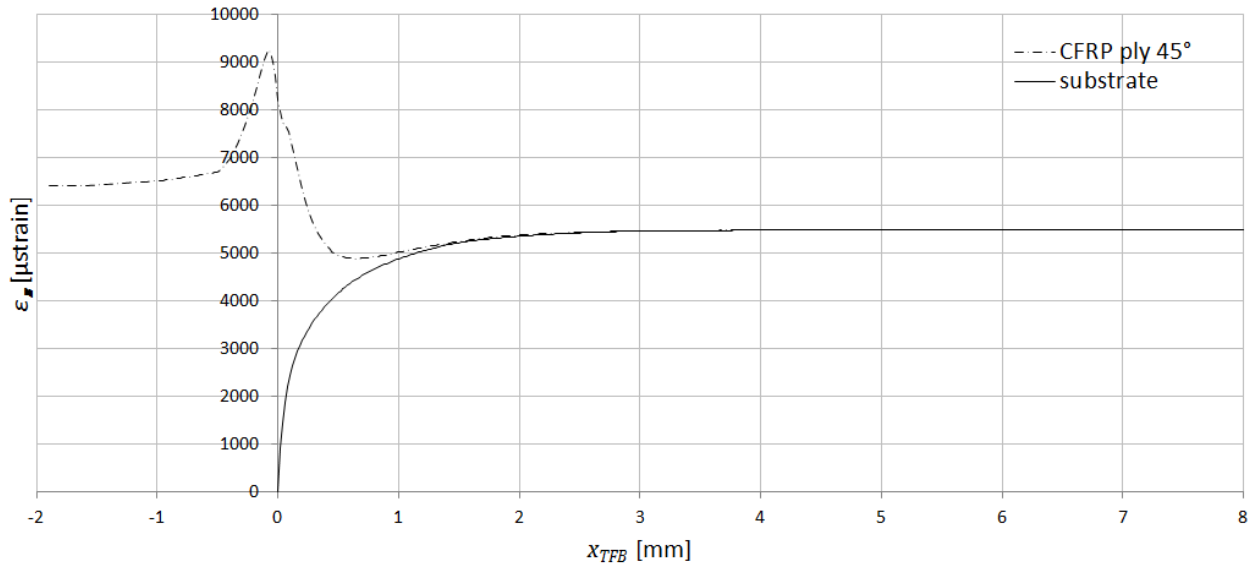


Figure 16. Average normal strain ε_x in the CFRP 45° ply adjacent to the TFB and in the substrate. Applied far field strain $\varepsilon_x = 6375 \mu\text{strain}$. Uniaxial tension of (0/45/90/-45/TFB/-45/90/45/0) laminate.

Similar conclusion could be drawn for the embedded configuration under bending, therefore the results were omitted.

Some peak values of strain above 5% were calculated in the sealant. However, these were strain concentrations the involved small areas, whereas the average sealant strain was below 4%. The Surlyn sealant is known for having a linear elastic response characteristic up to a strain of 4% and these progressively flattens out, becoming almost flat at 10%. The yield strength is reported at 8%. Therefore the use of a linear elastic material model was deemed close to correct.

4.5 Experimental match

The objective of modeling the delamination propagation under the same experimental loading conditions and material properties was to develop a better understanding of the critical failure mechanism that triggered the premature failures documented in chapter 2. The choice of focusing on delamination was driven by such experimental evidence. Additional goals included the determination of the propagation characteristics, critical loading conditions, onset locations and delamination lengths. These information were fundamental to narrow down the analysis effort for the subsequent parametric study. Finally, by matching the experimental failures it was possible to estimate the fracture of the muscovite substrate and of the *ac/s* interface, which were uncertain and unknown respectively.

A loading condition and onset location were defined critical if propagation with associated mechanical or electrical failure occurred before the desired ultimate load. The ultimate load was defined as the mechanical failure load of the baseline laminate, i.e. the CFRP laminate without the TFB. An electrical failure was assumed if the sealing provided by the packaging was compromised by a *ls/s – in* or a *ls/s – in* delamination propagated all the way through the battery peripheral seal, or if *ac/s* delamination occurred, with or without buckling of the packaging. Moreover, a failure was classified as critical only if it was the most critical for the interface and for the laminate configuration (externally bonded or embedded) under configuration.

Outside from the above critical conditions, a delamination round out was considered acceptable for preliminary design. Therefore stable crack growth and limited propagation (without the risk of TFB contamination) at the laminate ultimate load was allowed.

Ultimate loads were experimentally determined by testing baseline CFRP laminates and the results are listed in Table 2. The specimen dimensions and test methods were the same as the corresponding tests performed for the TFB-CFRP laminates as described in chapter 2.

	Stacking sequence	p_c [kN]	u_c [mm]
Uniaxial tension	(0/45/90/-45) _S	52.2	2.12
Four point bend	(0/45/90/-45) _{3S}	3.9	15.92

Table 2. Critical failure load p_c and displacement u_c . Mean values of 3 repetitions.

The delamination propagation analysis was performed on the same laminate configurations and loading conditions considered for the stress analysis, with the addition of the battery thickness variation during charge/discharge, Table 3. Also the crack onset locations were the same. The propagation analysis was performed for each delamination independently, therefore no interaction between them was considered. The boundary conditions and specimen dimensions for the finite element models were the same as the stress analysis, but since delaminations were simulated only on one end of the TFB, the problem was not symmetric and the specimens were therefore modeled entirely.

For a given interface, the lower value of fracture toughness of the adherents was used, Table 1.

The assumption for the VCCT is the energy release rate required to extend the crack is equal to the energy required to close the crack by the same length. This condition is verified only if the crack extension is small compared to the current length and if the growth occurs in a self-similar fashion. This ensures that the stress-strain field at the crack tip does not change significantly before and after a finite extension, which corresponds to an element length. This could lead to incorrect results if the initial delamination length is short compared to the mesh size. The shortest initial delamination length considered was 0.5 mm, whereas the mesh size adjacent to the crack tip was 0.02 mm. This mesh size was selected after mesh convergence studies that allowed to obtain consistent ERR distributions throughout the whole range of delamination lengths analyzed. All the initial delamination lengths were 0.5 mm except otherwise specified for specific cases in the following sections.

In addition to the crack length control scheme method, an automatic crack extension scheme was applied to propagate the delamination under constant load. The method consisted in a sequence of growth sub-increments as they are described in chapter 3. Artificial stabilization techniques and automatic increment size control were also applied according to the scheme illustrated in chapter 3. The objective of this

method was to trace the G_I , G_{II} and G_C curves at varying delamination length and under constant loading. Since the ERR is derivative of a potential energy, the G_T curve is referred to as driving force curve, whereas the G_C is the resistance curve or R-curve. This analysis is independent from the toughness of the interface and therefore could be performed when the material properties G_{IC} , G_{IIC} and η were not available. This was the case of the ac/s interface, since the fracture toughness of the active components was unknown.

Test description	Lay-ups	Configuration
TFB thickness variation during charge/discharge	(0/45/90/-45/TFB/-45/90/45/0)	embedded
Uniaxial mechanical tension test	(0/45/90/-45/TFB/-45/90/45/0)	embedded
	[(0/45/90/-45) _s /AF/TFB]	ext. bonded
Four point bending test	[(0/45/90/-45) ₃ /TFB/(-45/90/45/0) ₃]	embedded
	[(0/45/90/-45) _{3s} /AF/TFB]	ext. bonded
	[TFB/AF/(-45/90/45/0) _{3s}]	ext. bonded

Table 3. Analyzed loading conditions and laminate configuration for the experimental match.

4.5.1 Externally bonded configuration

4.5.1.1 Uniaxial tension

The uniaxial tension was simulated for the [(0/45/90/-45)_s/AF/TFB] laminate.

The af/l_s delamination modeled with the nominal muscovite toughness reported in Table 1 showed propagation at a much lower applied load than the experimental results. The actual fracture toughness was apparently higher than the theoretical value. This was attributed to the additional resistance provided by the resin ramp all around the battery. Therefore the automatic crack extension scheme was employed for tracing ERR distribution at the experimental failure load of the substrate in order to determine a more representative fracture toughness. The result is shown in Figure 17. The flat driving force curve is associated to a limit condition for stable delamination propagation. In fact the conditions for stable crack growth in case of constant G_C are

$$G = G_C \quad (1)$$

$$\frac{dG}{da} \leq 0 \quad (2)$$

This result confirmed the sudden delamination propagation documented in chapter 2 from the experiments.

The flat R-curve (i.e. almost constant G_C with crack length) was caused by the almost constant mode mixity under self-similar propagation.

In order to simulate this event with the crack length control scheme under variable load, a new value of fracture toughness was calculated by assuming that the mode II fracture toughness was twice the mode I fracture toughness. Within this assumption the BK law becomes

$$G_C = G_{IC} + G_{IC} \left(\frac{G_{II}}{G_T} \right)^\eta \quad (3)$$

Solving for G_{IC} we obtain

$$G_{IC} = \frac{G_C}{1 + \left(\frac{G_{II}}{G_T} \right)^\eta} \quad (4)$$

and

$$G_{IIC} = \frac{2G_C}{1 + \left(\frac{G_{II}}{G_T} \right)^\eta} \quad (5)$$

The mode I and II fracture toughness were calculated by substituting into equations (4) and (5) the maximum computed value of G_C . The resulting values were 89.5 J/m² and 178.9 J/m² respectively. The crack length control scheme simulation confirmed the unstable propagation with an almost vertical drop in the equilibrium path, Figure 18. This failure mode was classified as critical based on experimental evidence. The experimentally determined load-displacement curve of the baseline laminate with included post-failure response is superimposed to the calculated response in the figure to highlight that propagation occurred well before the desired ultimate load.

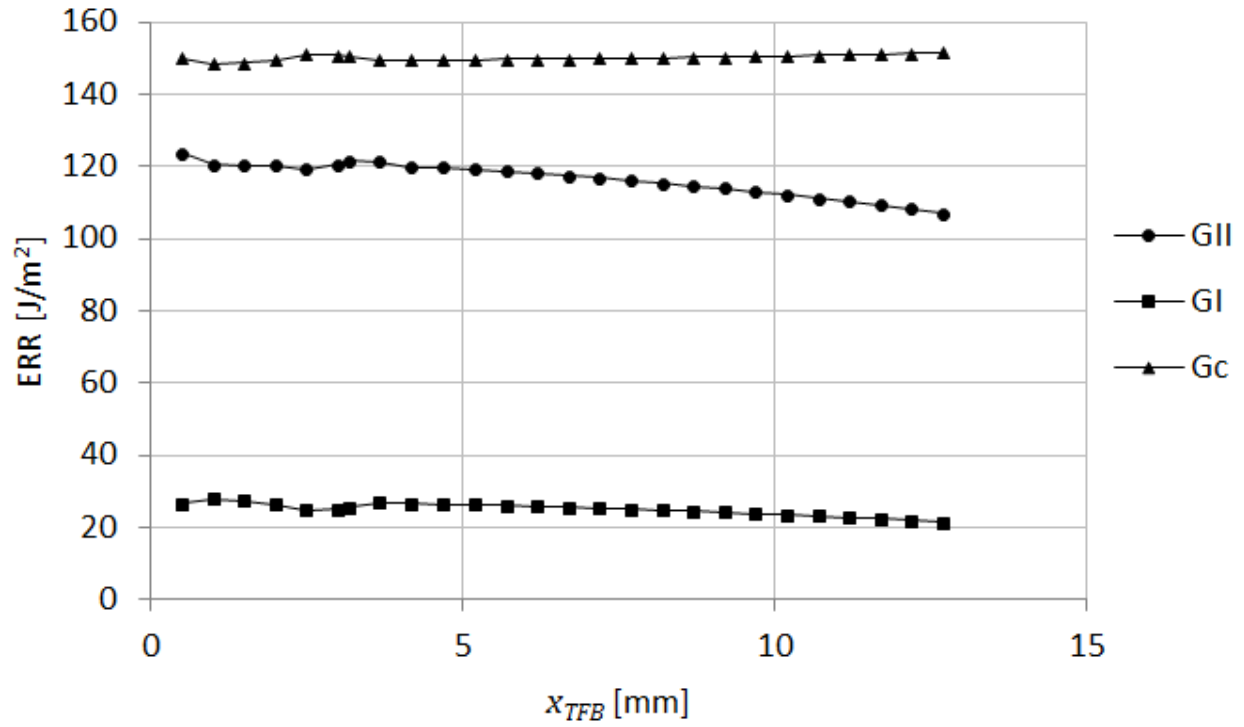


Figure 17. Uniaxial tension of [(0/45/90/-45)_s/AF/TFB]. Strain energy release rates at *af/sl*. Applied far field strain $\epsilon_x = 4898 \mu\text{strain}$.

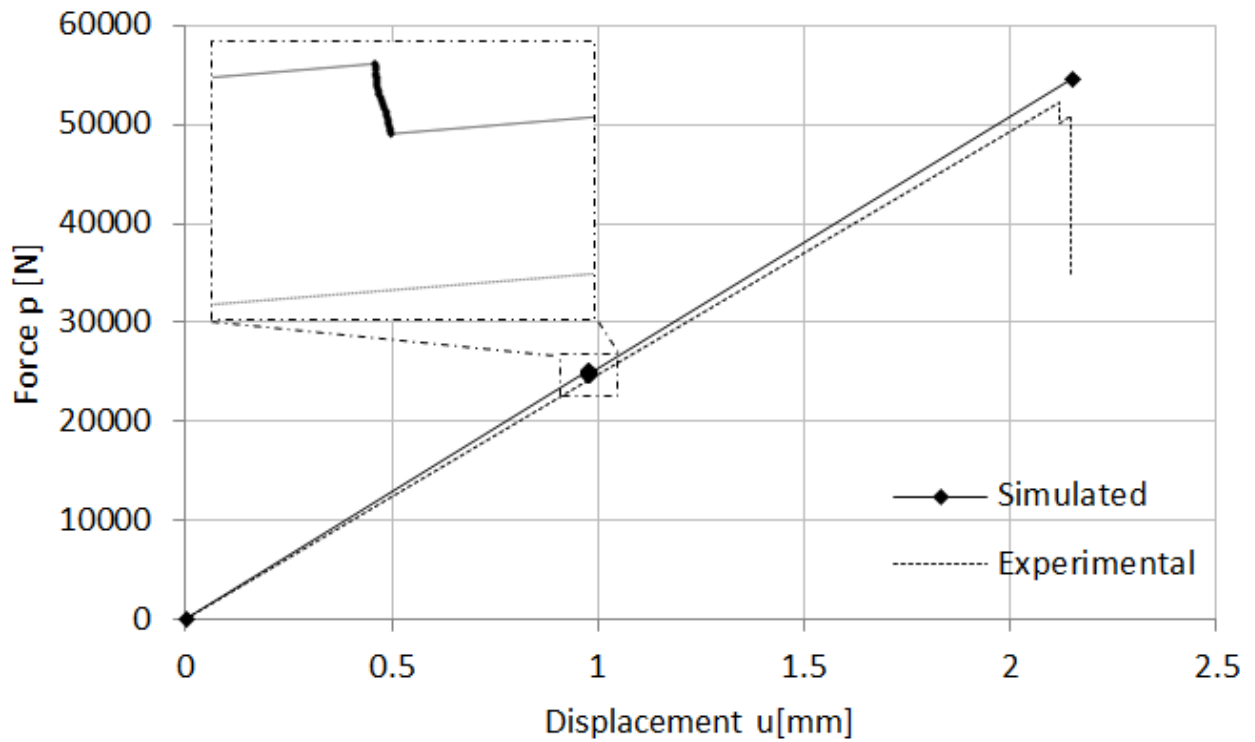


Figure 18. Uniaxial tension of [(0/45/90/-45)_s/AF/TFB]. Load-displacement curve for the *af/sl* delamination.

The $CFRP/af$ delamination was simulated using the fracture toughness of the CFRP material IM7/977-3, Table 1. The calculated load-displacement response, Figure 19, shows a sudden delamination propagation throughout the entire TFB length associated with a vertical load drop, similar to the af/sl delamination. This catastrophic failure led to the complete TFB disbonding from the laminate. The failure load was higher than the critical failure load for the af/sl delamination.

The driving force curve traced under the applied far field strain of 4898 μ strain, which corresponded to the critical load level for the experimental af/sl failure, confirms that the $CFRP/af$ was less critical. In fact, the driving force curve lies entirely below the R-curve, Figure 20.

The G_I was unexpectedly high for a uniaxial tension case. The mode I ERR was even larger than the mode II. To capture this behavior a nonlinear analysis was necessary for computing the correct stress-strain distribution. This failure mode was classified as critical.

The remaining delamination ac/s , $ls/s - in$ and $ls/s - out$ resulted not critical because covered by other loading conditions.

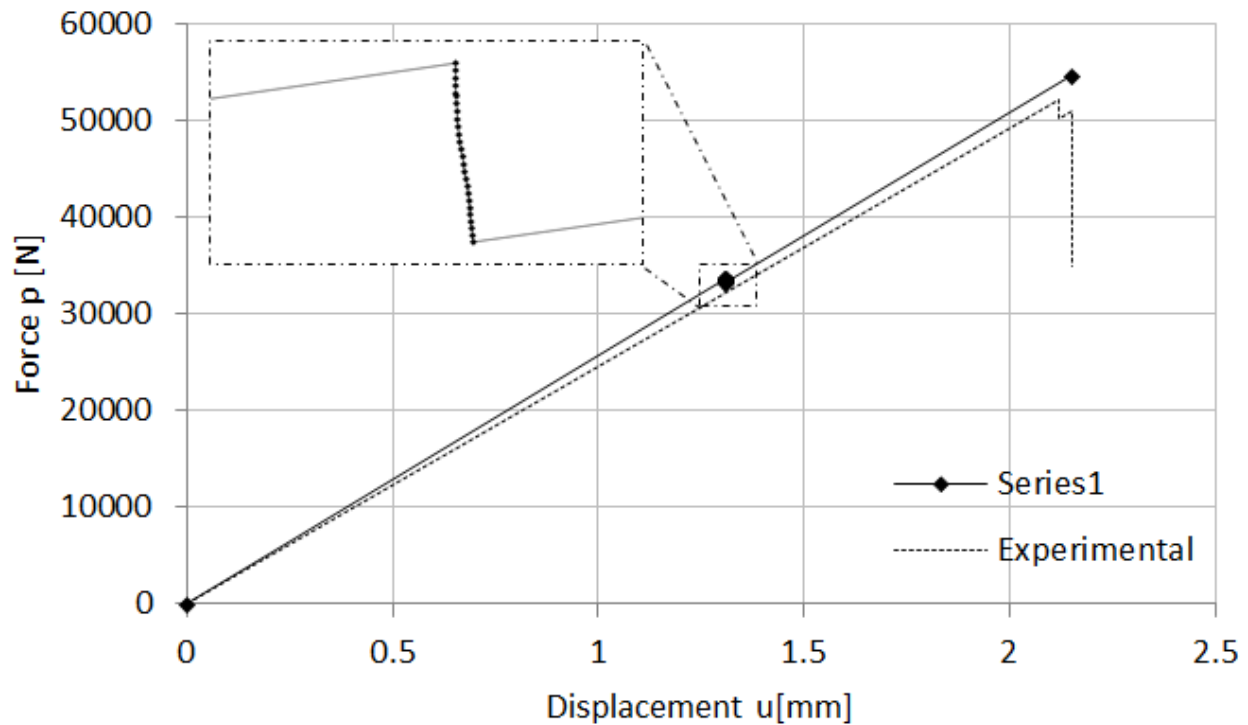


Figure 19. Uniaxial tension of $[(0/45/90/-45)_s/AF/TFB]$. Load-displacement curve for the $CFRP/af$ delamination.

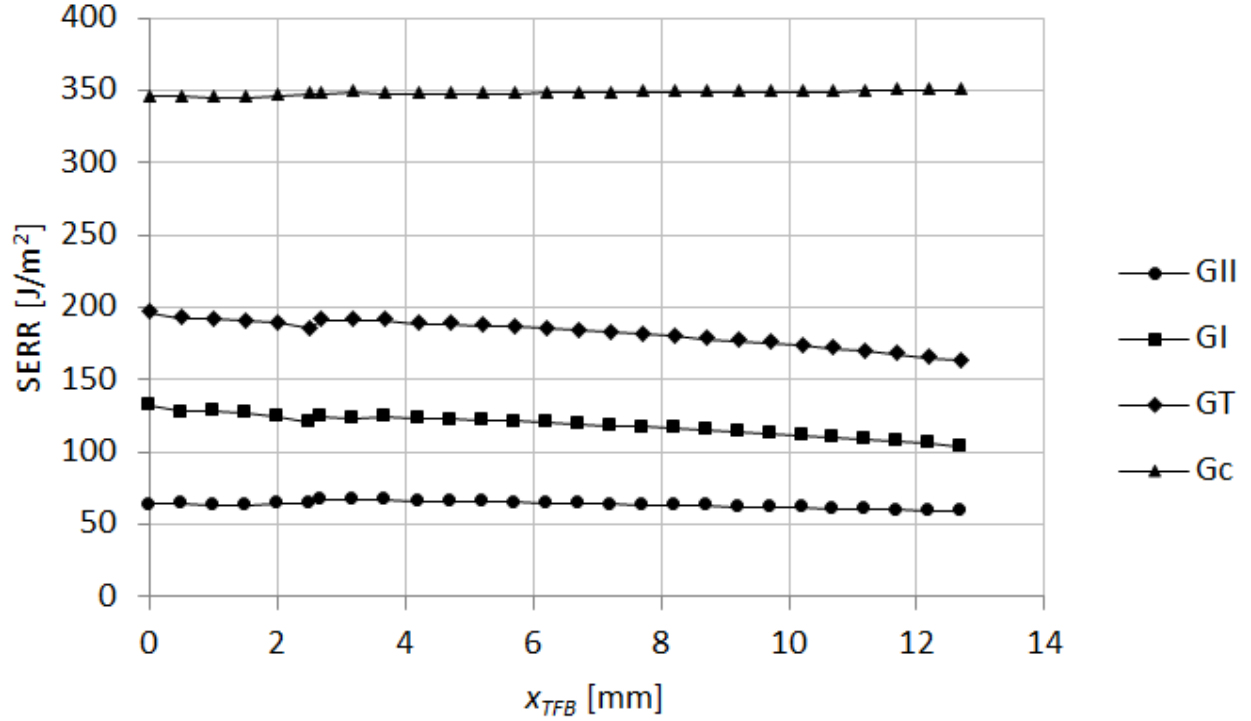


Figure 20. Uniaxial tension of [(0/45/90/-45)_s/AF/TFB]. Strain energy release rates at *CFRP/af*. Applied far field strain $\epsilon_x = 4898 \mu\text{strain}$.

4.5.1.2 Four point bending

The simultaneous occurrence of curvature and compression was simulated with the [(0/45/90/-45)_{3s}/AF/TFB] laminate loaded in four point bending. The finite element model is shown in Figure 21. This loading condition was expected from the experimental evidence to be particularly critical because of the delamination buckling at the *ac/s* interface.

The *CFRP/af* delamination was modeled with an initial 0.5 mm delamination the interface between the CFRP 0° ply and the adhesive film. In addition the vertical interface between the TFB and the resin ramp was modeled pre-cracked through its full length to allow the TFB to slide through the resin ramp. This incompatibility was introduced assuming the resin ramp would in reality break off.

The calculated load-displacement curve featured a sharp snap back instability due to the unstable delamination propagation and complete disbonding of the TFB. Such failure, however, occurred at an applied load that was higher than the desired ultimate load for any initial delamination length. In fact the

whole softening branch of the equilibrium path occurred after the experimentally determined failure of the baseline laminate, whose experimental curve is included in Figure 22. This failure mode was therefore not critical. The driving force curve, Figure 23, had a positive tangent in the first part, denoting unstable growth, and then the slope became negative, denoting a small portion of stable growth, as seen in the load displacement curve.

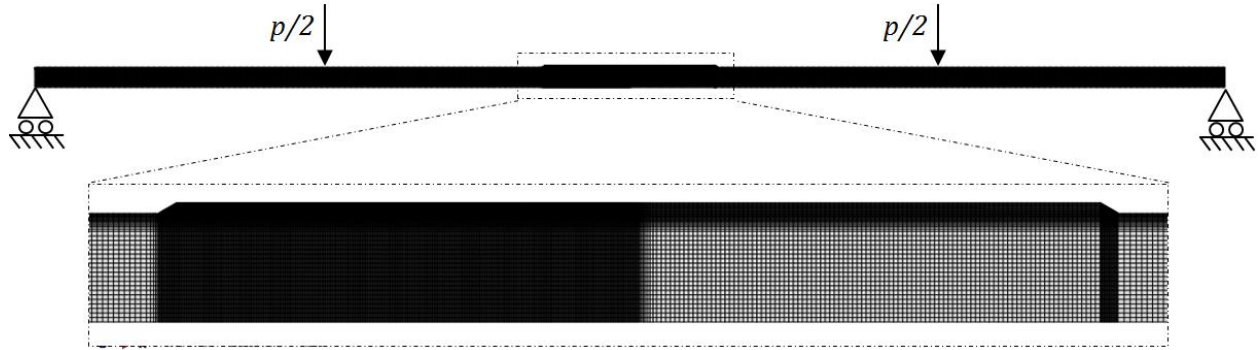


Figure 21. FEM of $[(0/45/90/-45)_{3S}/AF/TFB]$ laminate subjected to four point bending.

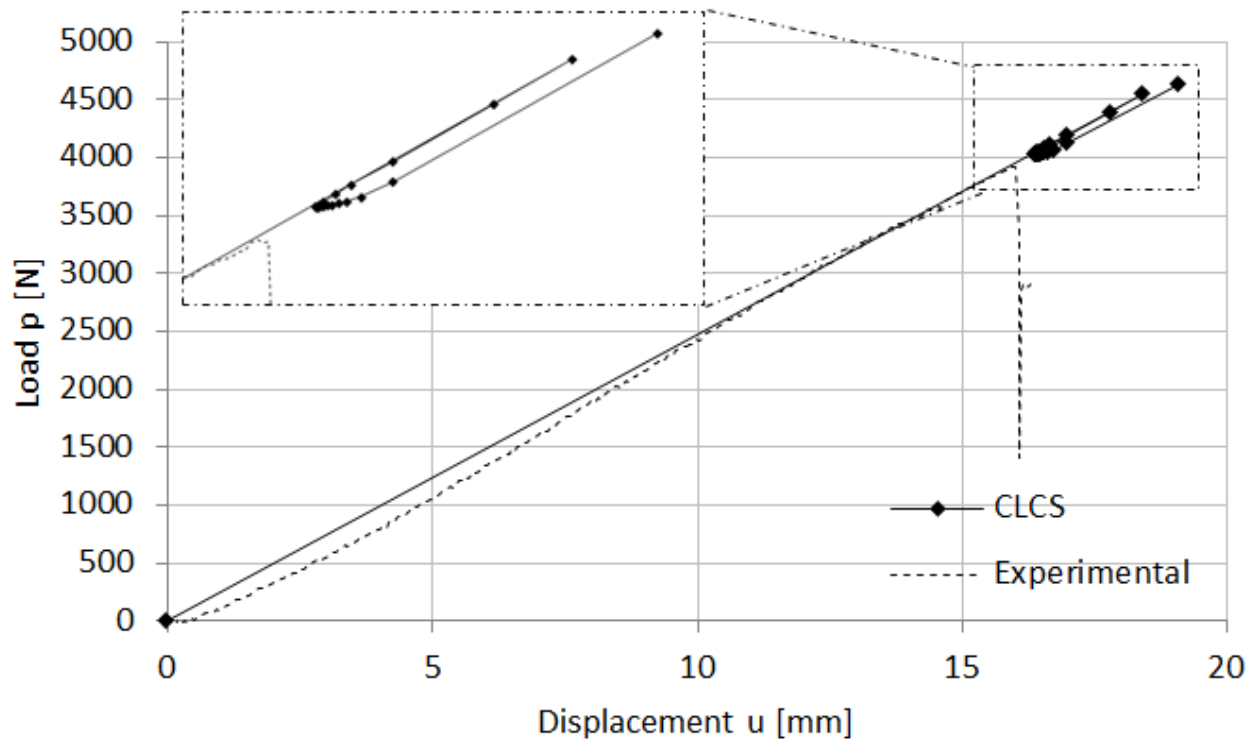


Figure 22. Four point bending of $[(0/45/90/-45)_{3S}/AF/TFB]$. Load-displacement curve for the *CFRP/af* delamination.

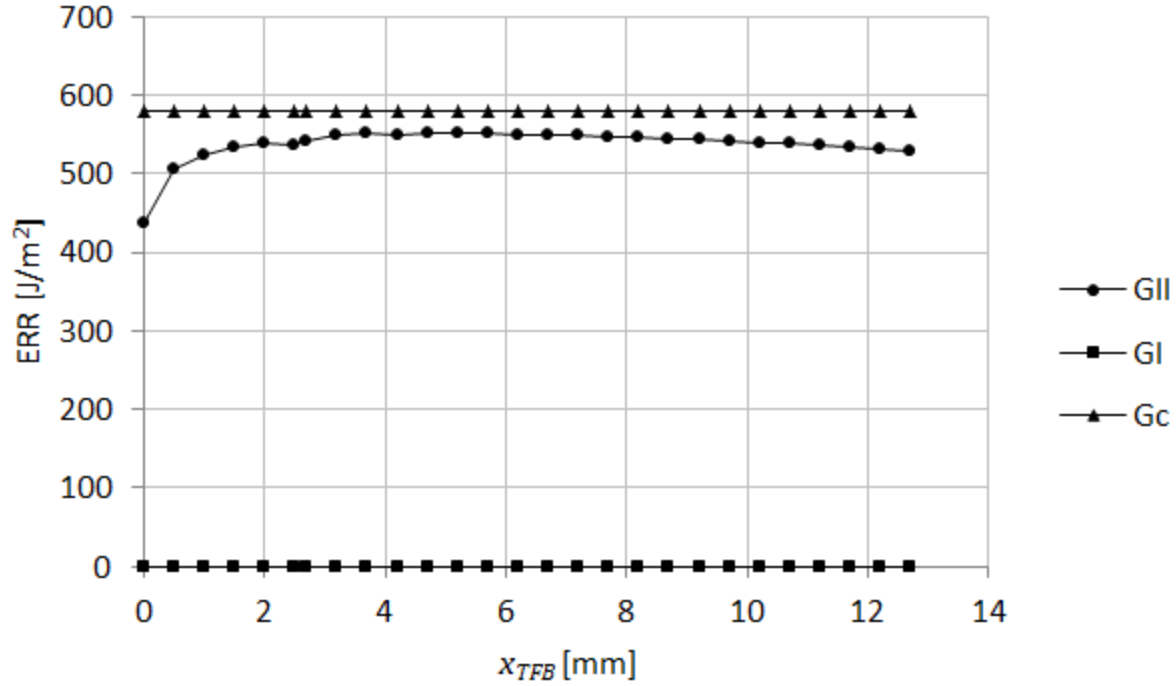


Figure 23. Four point bending of $[(0/45/90/-45)_{3S}/AF/TFB]$. Strain energy release rates at $CFRP/af$ under ultimate load.

The pre-cracking of the resin ramp was also applied to the simulation of the af/sl delamination. The resulting incompatible displacement is shown in Figure 24.

The crack length control scheme analysis revealed a snap back instability with complete TFB disbonding at a lower load than experimental strength of baseline laminate, Figure 25. This load case, however, was not classified critical because covered by the uniaxial tension loading failure at the af/sl interface discussed earlier.

The ERR distribution traced at the baseline ultimate load confirmed the high levels of G_{II} , Figure 26. The average value of G_{II} for the af/sl delamination was twice as much the value for the $CFRP/af$ delamination under the same loading condition. The difference can be hardly explained by the slightly increased distance from the neutral axis. More likely a higher stress-strain concentration at the af/sl interface was the most important cause. Considering the similarity and vicinity of the two delaminations, as well as the remarkable difference of the energy release rates, it seemed that the high material

inhomogeneity led the dependence of the delamination characteristics to the stacking sequence to an extreme.

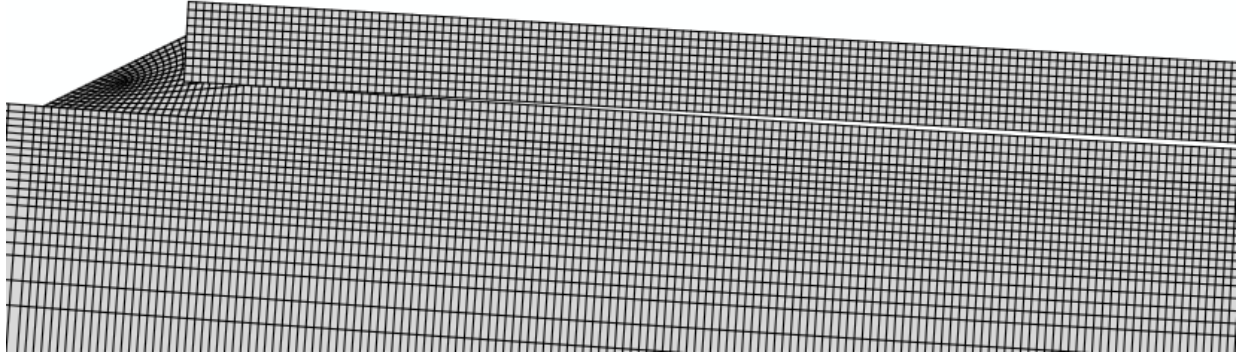


Figure 24. Incompatible displacements due to resin ramp pre-cracking.

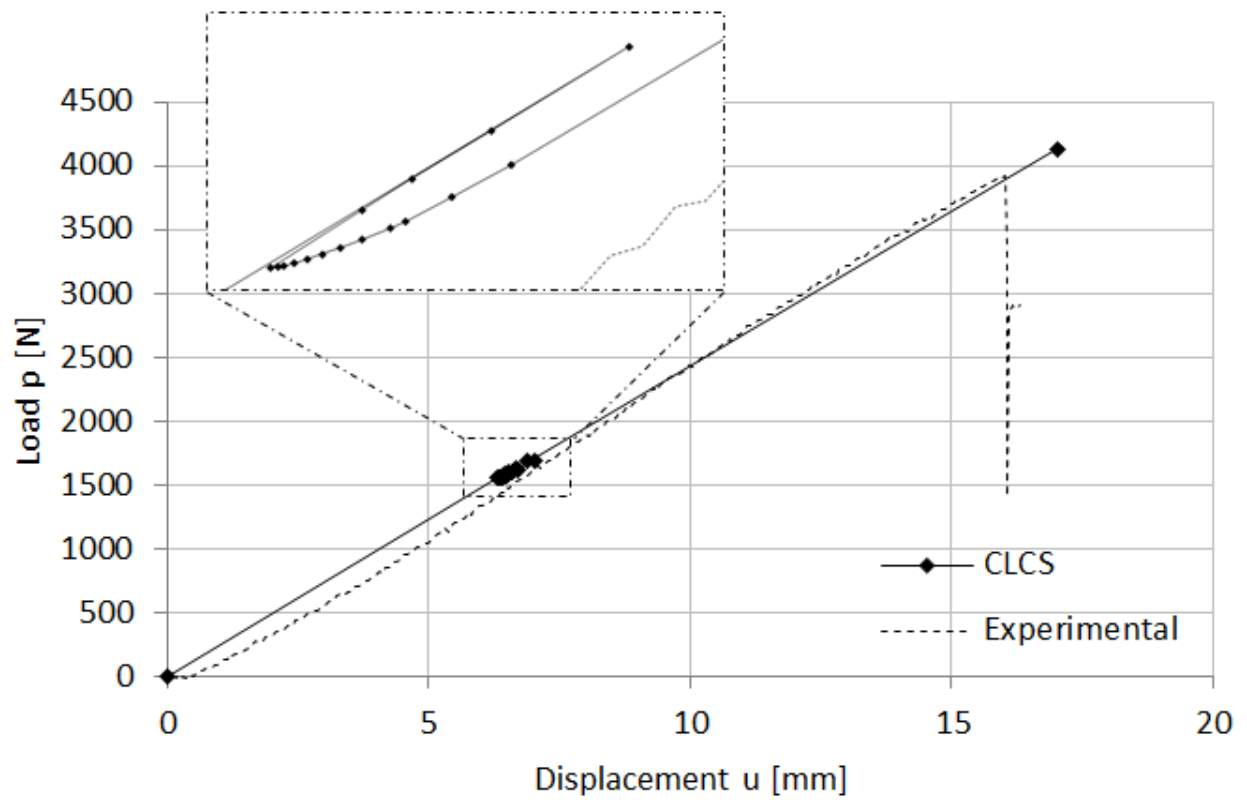


Figure 25. Four point bending of $[(0/45/90/-45)_{3S}/AF/TFB]$. Load-displacement curve for the *af/sl* delamination.

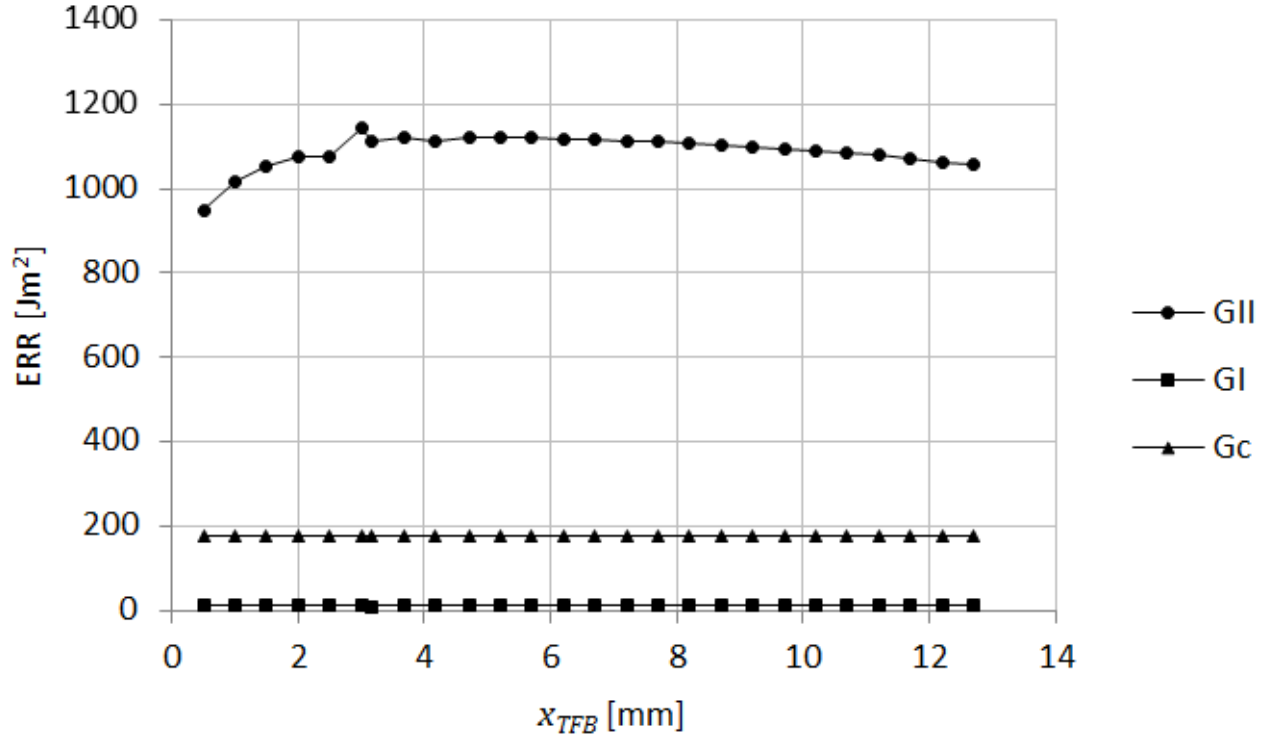


Figure 26. Four point bending of $[(0/45/90/-45)_{3S}/AF/TFB]$. Strain energy release rates at af/sl and ultimate baseline load.

The ac/s delamination was modeled by inhibiting propagation of the left crack tip, which was located at the ls/s interface at $x_{TFB} = 2.675$ mm. Therefore only the crack tip at the right end of the delamination was allowed to propagate through the ac/s interface. The initial crack length was 1mm, which included 0.5 mm through the ls/s interface.

The automated propagation analysis with the crack length control scheme could not be performed because the fracture toughness of the active components was unknown. The driving force curve traced at the delamination-buckling failure load detected by the experiment (displ. 5.1mm, -2832 μ strain upper surface strain, radius of curvature 531mm, see chapter 2) confirmed the buckling of the upper substrate, Figure 27. The elastic instability of the upper TFB substrate led to the sharp increase of G_{II} , which was decreasing with delamination propagation before the instability occurred. The G_{II} reached a peak value of about 40 J/m². The G_I , which was negligible before buckling, was excited by the crack opening caused by the substrate instability and reached a peak value of about 11 J/m², Figure 28. Therefore the

occurrence of the local buckling turned the stable delamination growth into unstable. After reaching the peak, both the G_I and G_{II} decreased monotonically.

This analysis suggested that the fracture toughness at the interface between the active components and the sealant was lower than the peak values quoted above. These values were relatively high for multifunctional electrode and electrolyte materials. This suggested that the possibility of lowering this requirement, which would probably be prohibitive to achieve even with the next generation of nanostructured materials, should be explored by functionally grading the moduli of the packaging materials. This subject is discussed in the following section. Based on these considerations, this failure mode was classified as critical.

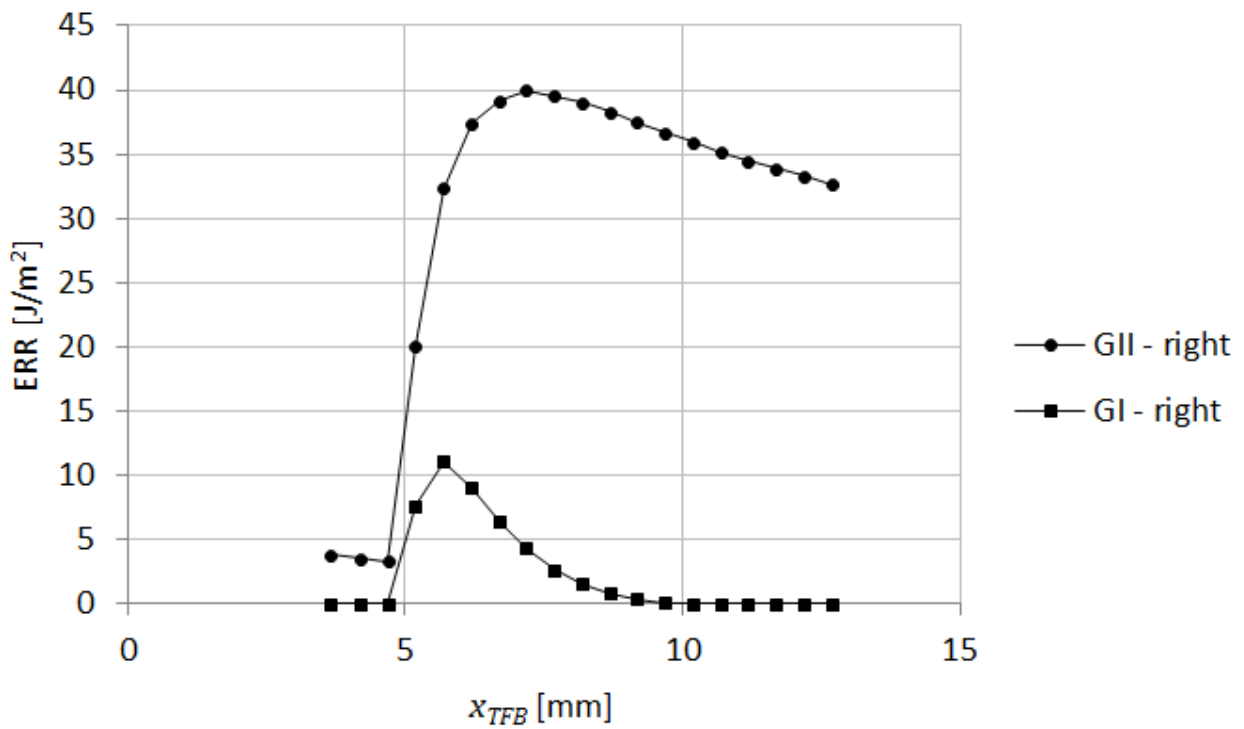


Figure 27. Four point bending of $[(0/45/90/-45)_{3S}/AF/TFB]$. Strain energy release rates at ac/s under critical delamination-buckling load determined experimentally (see chapter 2).

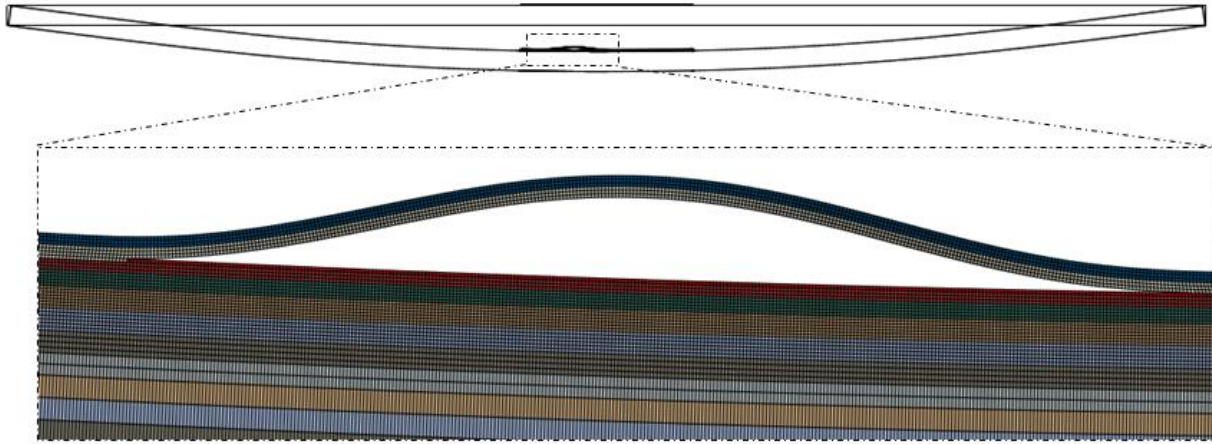


Figure 28. Close-up at local delamination-buckling collapse of the externally bonded TFB.

The fracture toughness calculated for the *af/sl* delamination under uniaxial tension was used to simulate the *sl/s – in* delamination. This choice was based on the assumption that both delamination would consist in the cohesive failure of the muscovite substrate, which also features a lower toughness than the Surlyn sealant. The crack length control scheme simulation calculated an unstable delamination growth of the initial 1 mm long delamination that started at the edge of the active components and propagated all the way to the TFB end, thereby opening a path for air and moisture contamination. This was associated to a snap back instability in the load displacement response, Figure 29. Because of the small change in specimen stiffness upon propagation, the softening branch was about coincident to the elastic branch. The automatic loading/unloading procedure was stopped when the delamination reached the TFB end, otherwise loading would have taken place along a stiffening branch with about the same slope as the elastic one. By observing the equilibrium path and the superimpose experimental response of the baseline laminate we could conclude that this failure mode was critical for an initial delamination length longer than 2 mm. Moreover, a parametric study of varying fracture toughness and elastic moduli should be carried out in order to exclude a more critical delamination buckling that could occur for certain combinations of material properties.

The driving force curve was traced at the critical delamination buckling load (displ. 5.1mm, 2832 ustrain, radius of curvature 531mm, see chapter 2) even if this failure mode was not seen experimentally. The

exponentially increasing mode I and II ERR as the delamination tip approached the TFB end indicated that the propagation was unstable throughout the whole delamination path. The increase in the ERR towards the TFB end was due to the stress concentration located at the battery end. The high value of G_I was probably due to the rotation and lifting of the upper substrate enforced by the resin ramp.

On the other hand, the G_I was negligible if the delamination propagated at the same interface but in the other direction, that is from the outside in. This was the case of the *sl/s – out* delamination. For the *sl/s – out* the vertical interface of the resin ramp was pre-cracked as described before. The calculated structural response showed a similar but less deep snap-back instability than the *sl/s – in* delamination, Figure 31. The softening branch, in fact, was shorter and turned into a stiffening branch towards the end of the propagation, indicating stable growth following the initially unstable propagation. The equilibrium path showed that growth could occur before the baseline ultimate load for an initial delamination length larger than 1.5 mm. This was confirmed by the intersection of the R-curve and the driving force curve shown in Figure 32. The *sl/s – in* and the *sl/s – out* delaminations seemed to be equally critical because the critical initial delamination length is about the same. The second tends to stabilize growth before reaching the active components and causing contamination, showing more damage tolerance than the first. However, the *sl/s – out* delamination under this loading condition was not classified critical because covered by a more critical *sl/s – out* delamination under simultaneous curvature and tension.

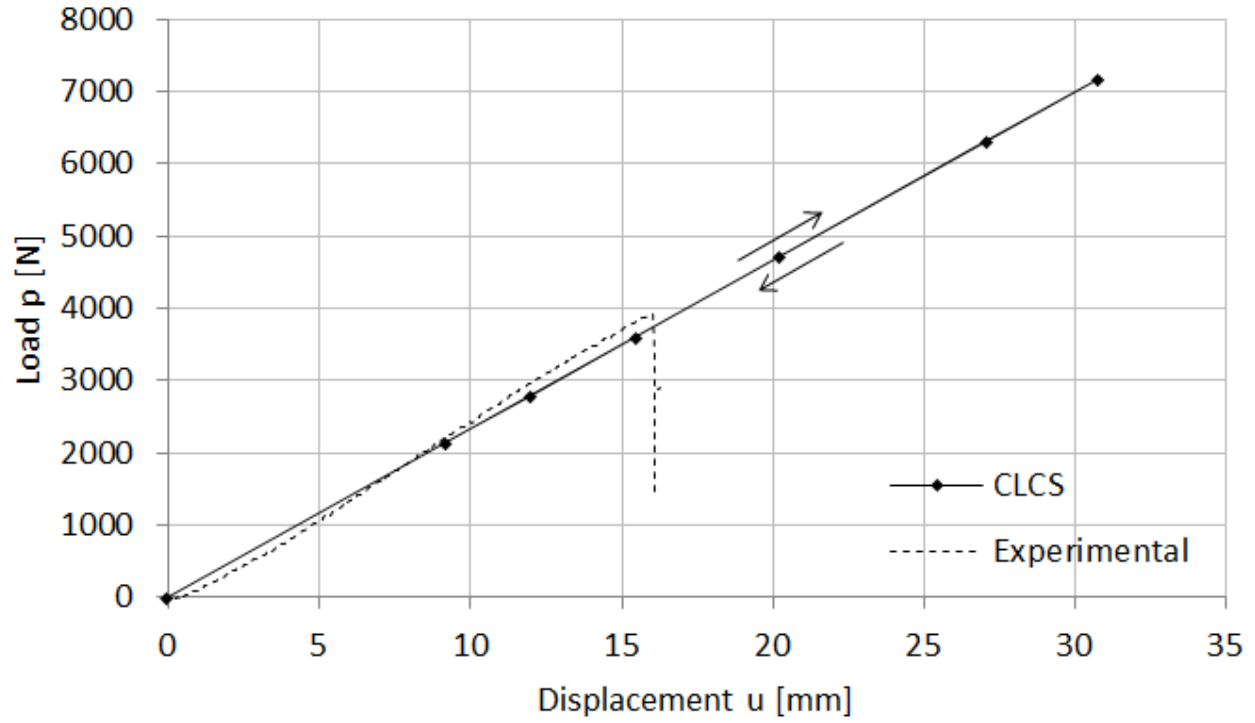


Figure 29. Four point bending of $[(0/45/90/-45)_{3S}/AF/TFB]$. Load-displacement curve for the $sl/s - in$ delamination.

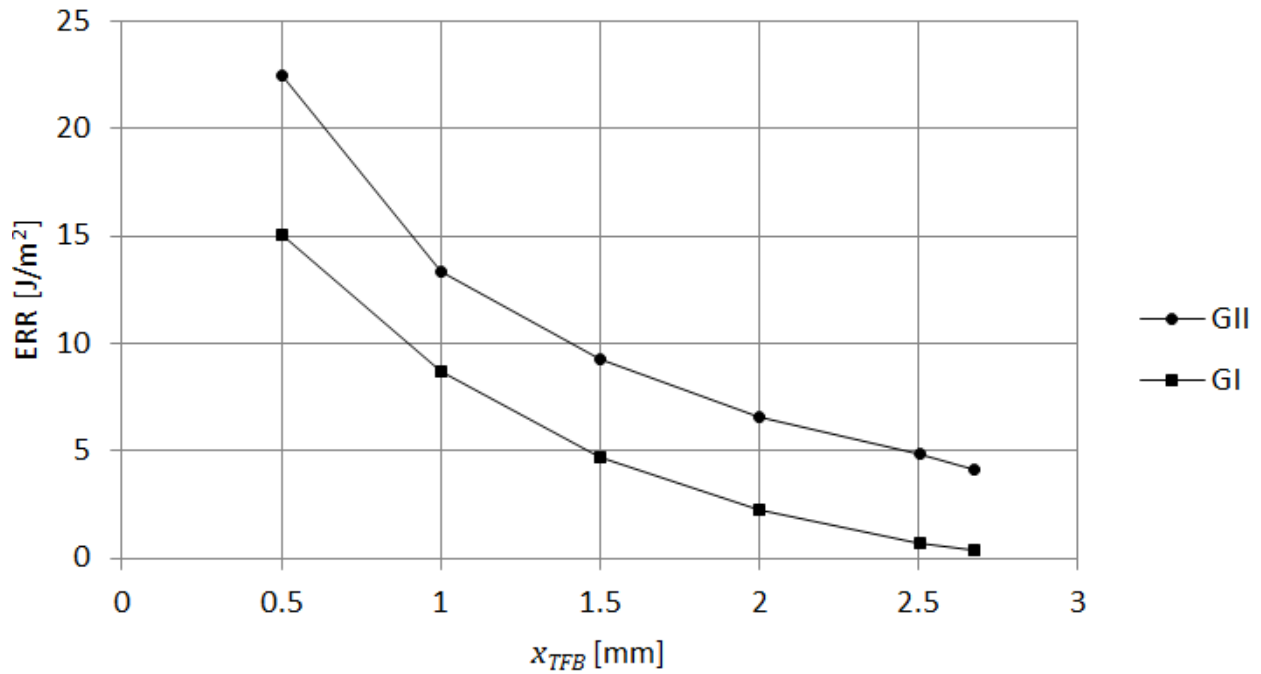


Figure 30. Four point bending of $[(0/45/90/-45)_{3S}/AF/TFB]$. Strain energy release rates at $sl/s - in$ under critical delamination-buckling load determined experimentally (see chapter 2).

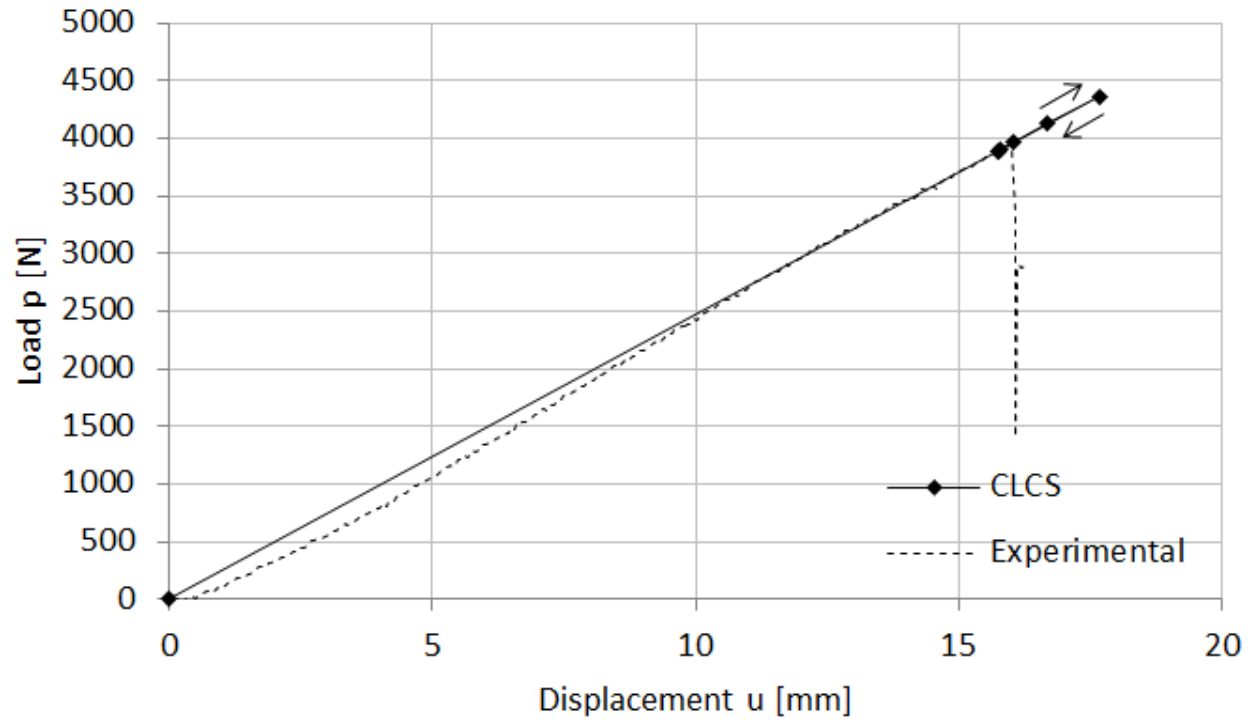


Figure 31. Four point bending of $[(0/45/90/-45)_{3S}/AF/TFB]$. Load-displacement curve for the *sl/s - out* delamination.

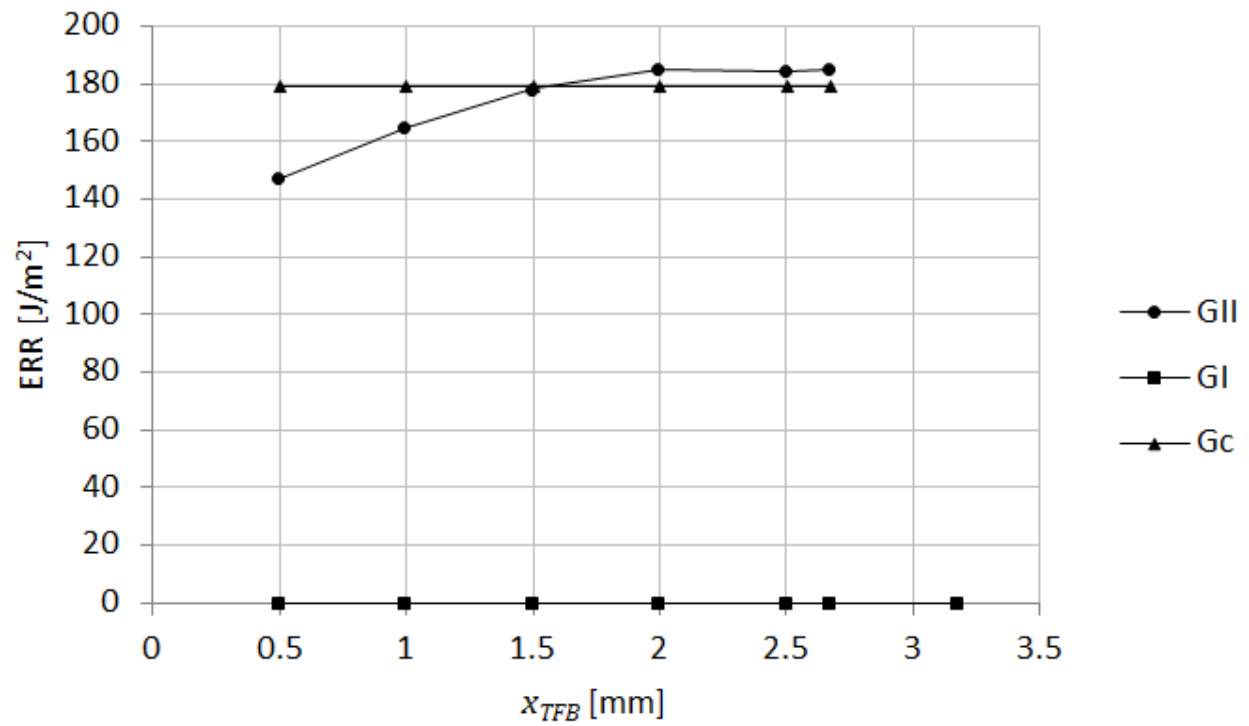


Figure 32. Four point bending of $[(0/45/90/-45)_{3S}/AF/TFB]$. Strain energy release rates at *sl/s - out* under ultimate load.

The simultaneous occurrence of curvature and positive in-plane strain was simulated by modeling the four point bending test of the [TFB/AF/(-45/90/45/0)_{3S}] laminate, where the TFB was bonded onto the lower surface of the CFRP specimen.

The *CFRP/af* delamination showed stable propagation, if we exclude a small initial snap-back due to a numerical discontinuity in the value of G_{II} . However, the propagation throughout the full TFB length occurred within a small load increase of less than 100 N. As shown in Figure 33 this failure mode was critical and it was classified as the *CFRP/af* delamination occurring under uniaxial tension. In both cases the fracture was mode I dominated with a similar mode mixity ratio.

The gentle negative slope of the G_I distribution confirmed the stable but fast propagation nature of this failure, Figure 34. The high values of ERR dissipated at ultimate load suggest that this is a critical failure mode. However this is expected to be critical for any structural secondary bonding.

This loading condition was similar to a single leg bending test (SLB) as evident from the deformed configuration shown in Figure 35. As in a SLB test, a mixed mode fracture was found. On the other hand, mode I only was present for the simultaneous occurrence of curvature and compression. This was the reason why the curvature plus compression case was more critical.

The propagation behavior of the *af/sl* delamination was similar to the *CFRP/af* delamination, but more critical. In fact it occurred at a lower applied load, Figure 36. This is in agreement with the experimental failure. However, even if the loading and the location of the delamination was similar to the *CFRP/af* case, G_I decrease notably as G_{II} increased, confirming the strong effect of stacking sequence on delamination characteristics for these inhomogeneous laminates. Also for this failure more the vertical interface of the resin ramp was pre-cracked to allow for the upper substrate to separate from the CFRP sub-laminate, Figure 38. This loading condition was not classified critical for the *af/sl* delamination because the curvature plus compression case was slightly more critical.

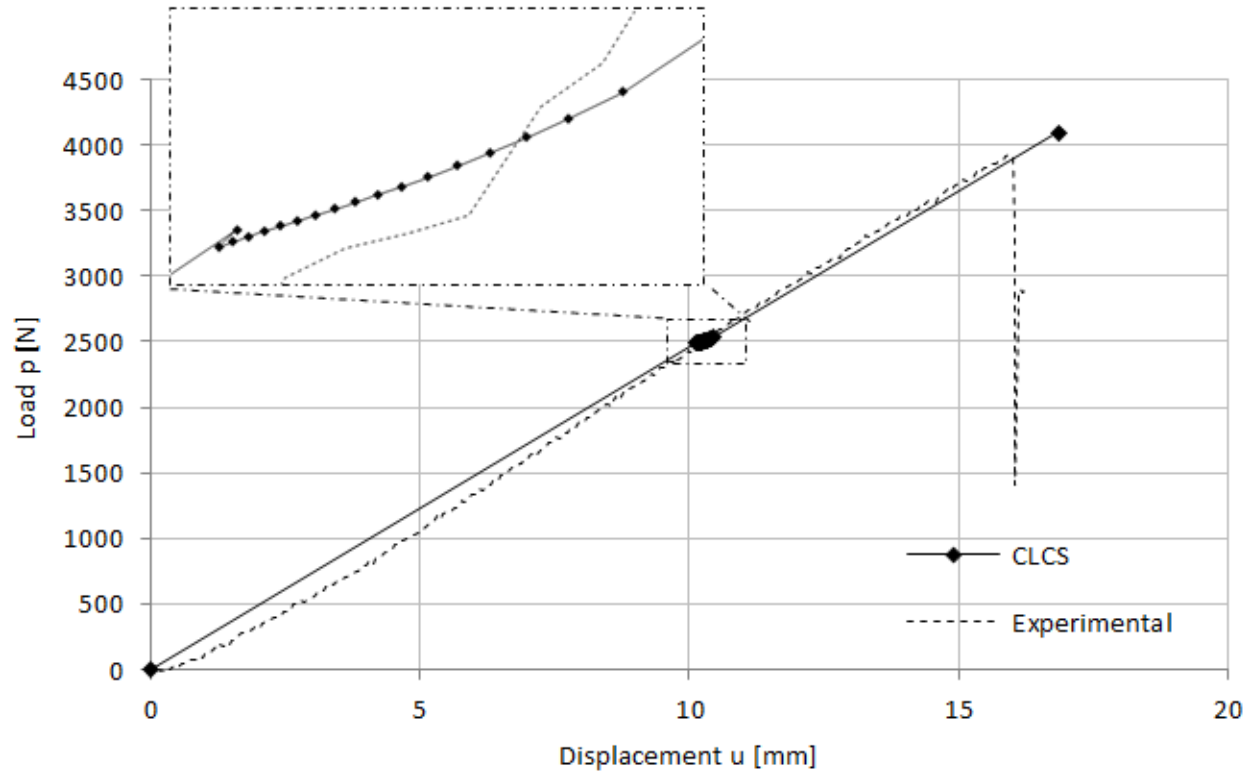


Figure 33. Four point bending of $[TFB/AF/(-45/90/45/0)_{3S}]$. Load-displacement curve for the $CFRP/af$ delamination.

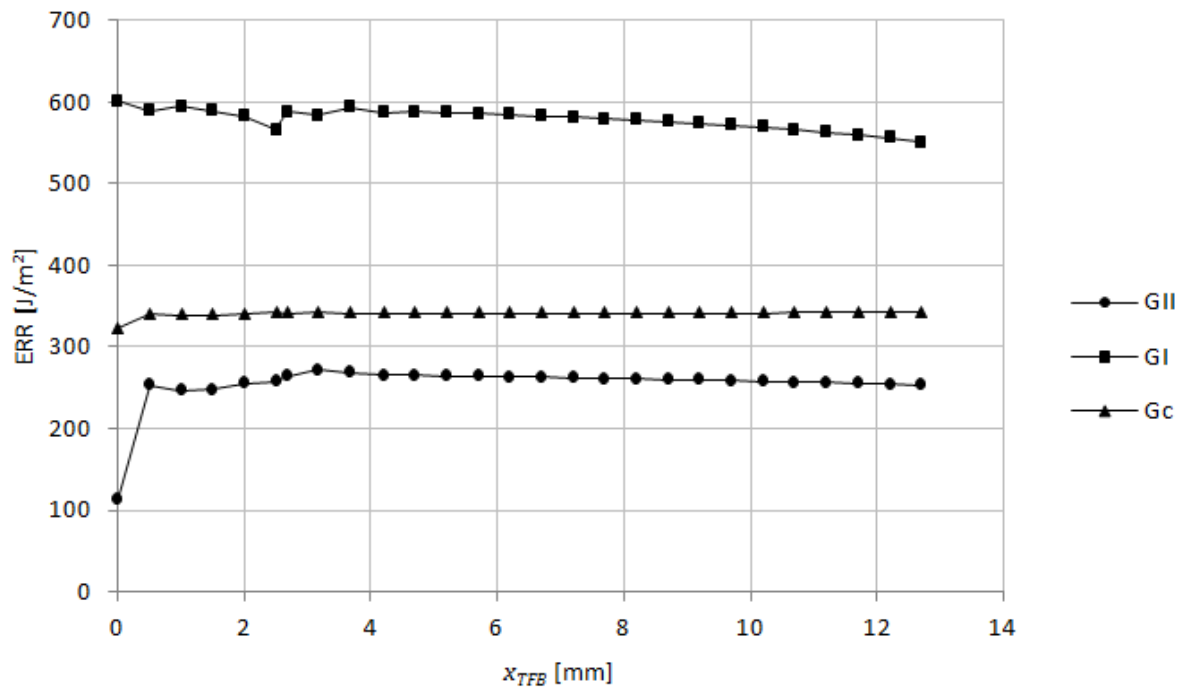


Figure 34. Four point bending of $[TFB/AF/(-45/90/45/0)_{3S}]$. Strain energy release rates at $CFRP/af$ under ultimate load.

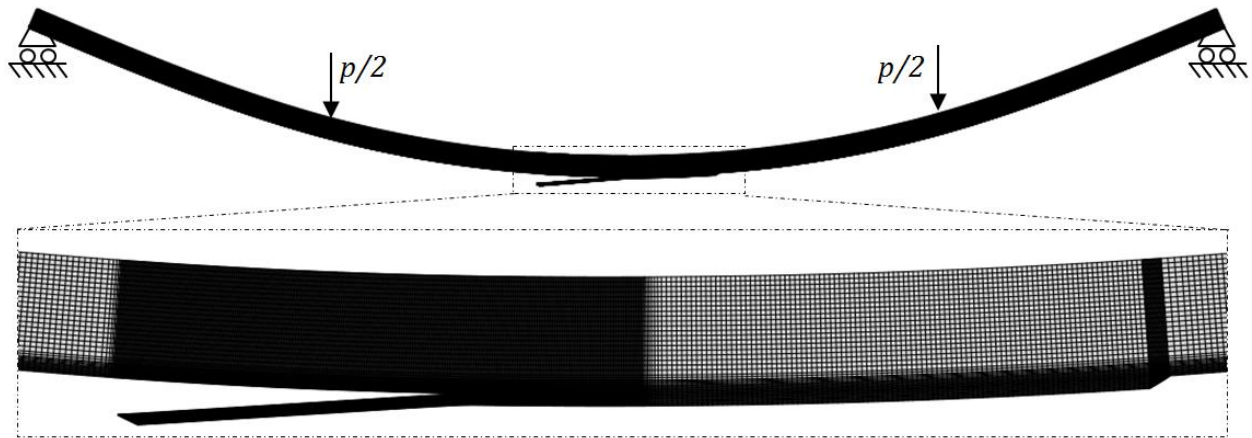


Figure 35. FEM of four point bending of [TFB/AF/(-45/90/45/0)_{3S}].

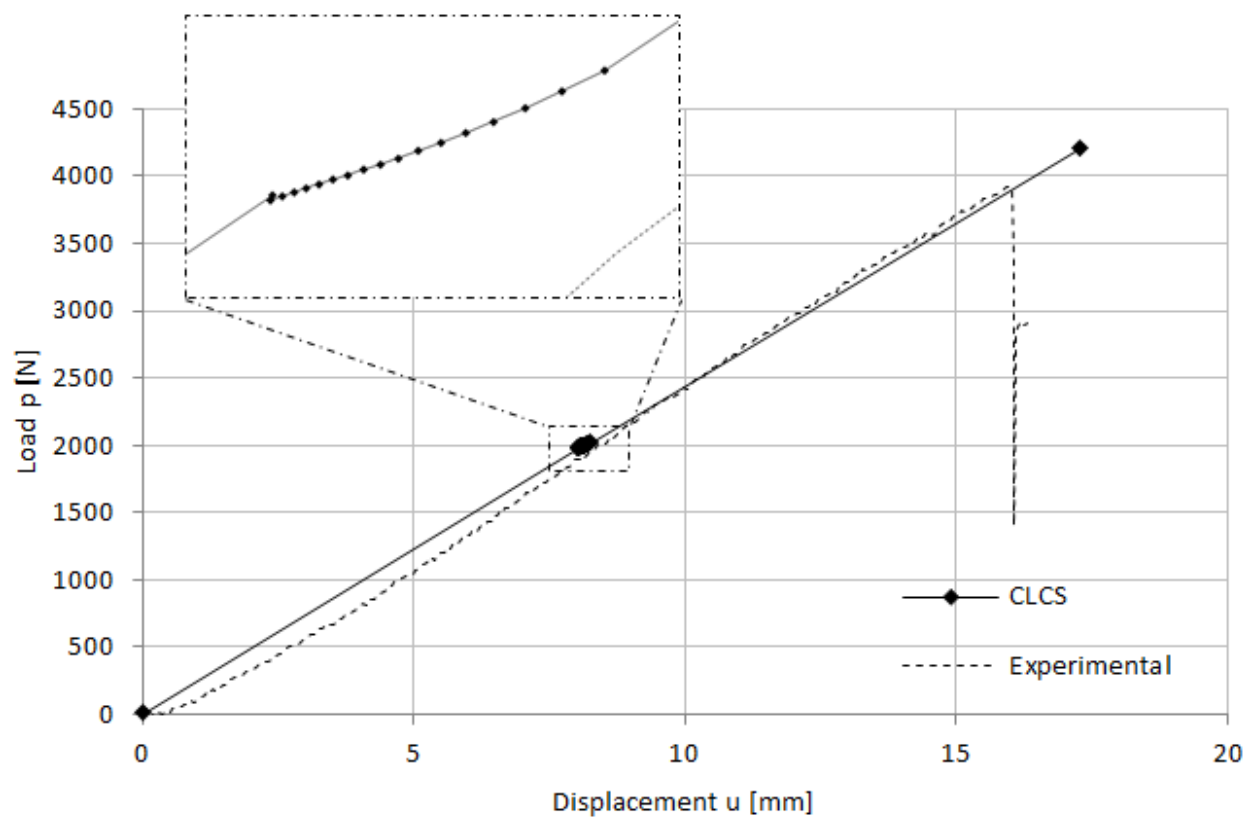


Figure 36. Four point bending of [TFB/AF/(-45/90/45/0)_{3S}]. Load-displacement curve for the *af/sl* delamination under ultimate load.

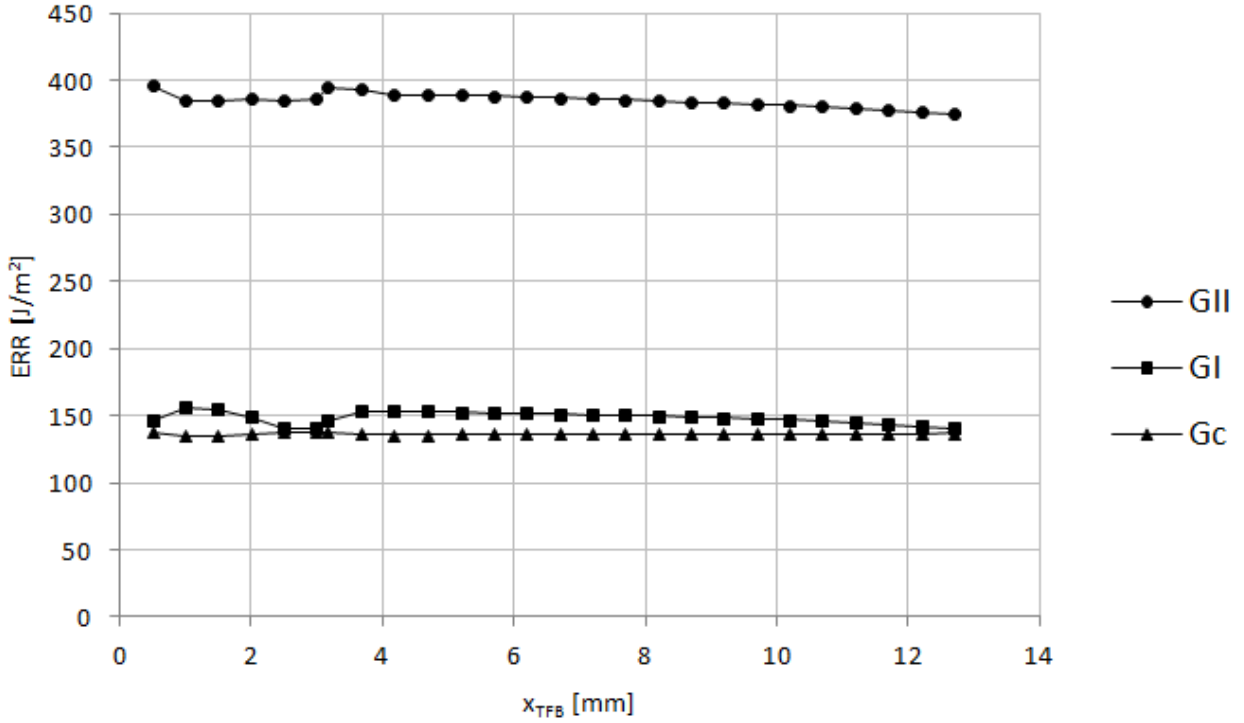


Figure 37. Four point bending [TFB/AF/(-45/90/45/0)_{3S}]. Strain energy release rates at af/sl .

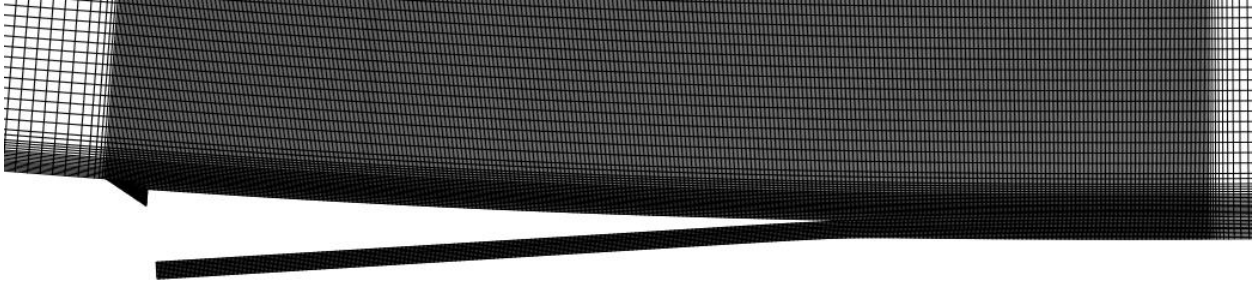


Figure 38. Detail of deformed FEM of [TFB/AF/(-45/90/45/0)_{3S}] under four point bending.

The ac/s delamination was simulated with an initial delamination length of 1 mm as described for the same delamination tested under the previous loading conditions. The calculated driving force curve was decreasing with delamination length, showing a stable propagation, Figure 39. The mode II delamination was dominant. The dissipated ERR reached values that can be classified as “structural” and were deemed critical for electrode and electrolyte materials. Moreover, the failure mode of the ac/s delamination under this loading condition was different from the compression case, where buckling was present. Therefore,

the simultaneous occurrence of curvature and tension was classified as a critical loading condition together with the compression one. Both of them were considered for the parametric study presented in the next section.

The *sl/s – in* propagated with an unstable fashion from the active components to the TFB outer edge, thereby opening an air and moisture path for the TFB contamination failure. The snap-back response was similar to the compression case, but less critical, Figure 40. Tension was less critical than compression also because unstable propagation occurred for an initial delamination of 3 mm or longer, Figure 41.

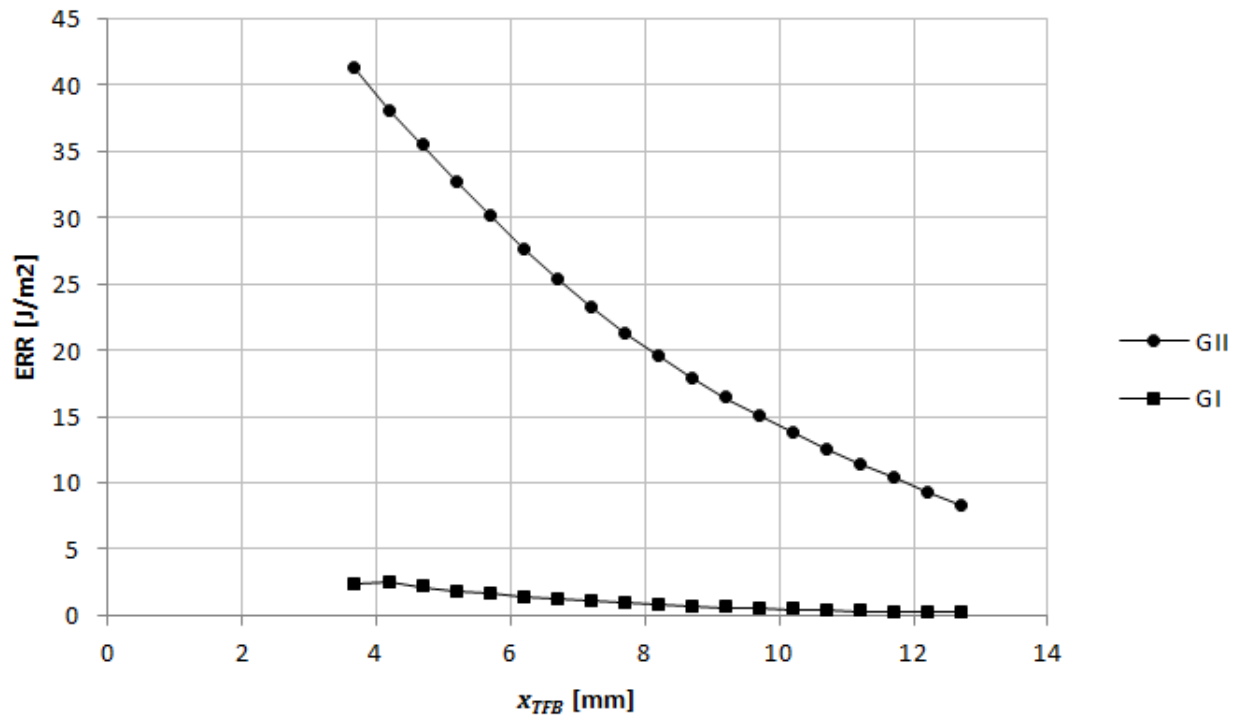


Figure 39. Four point bending [TFB/AF/(-45/90/45/0)_{3S}]. Strain energy release rates at ac/s.

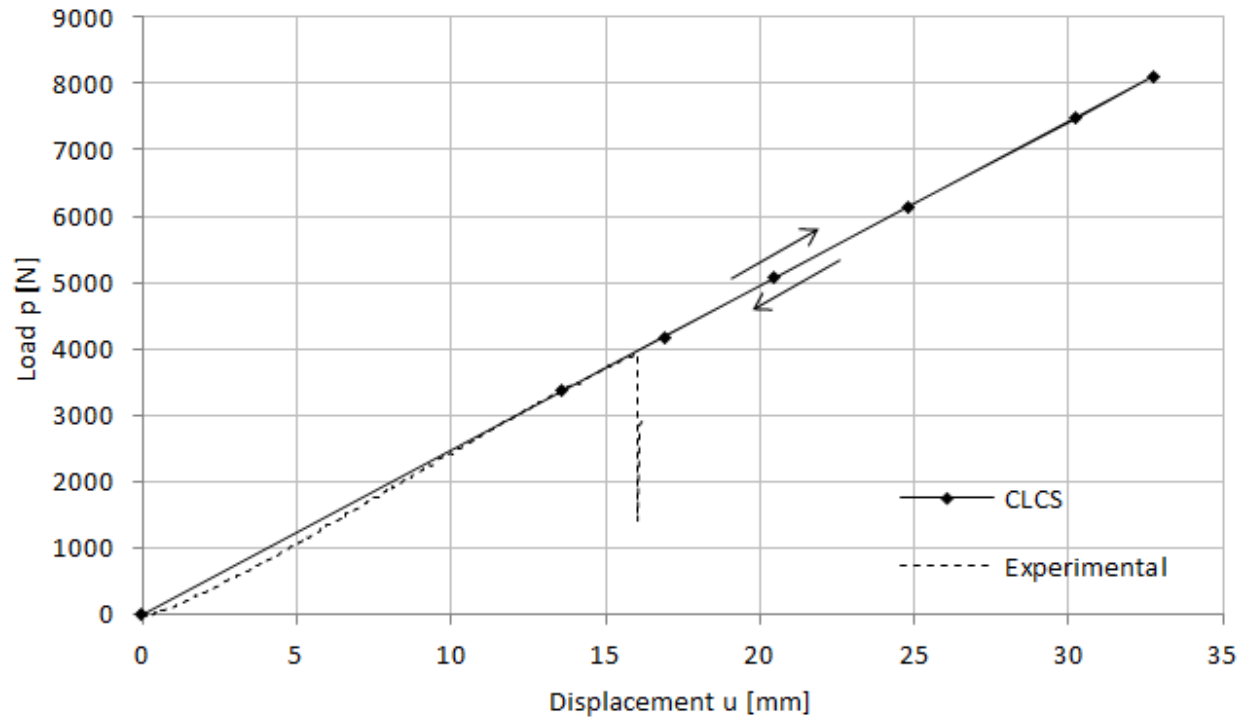


Figure 40. Four point bending of $[TFB/AF/(-45/90/45/0)_{3S}]$. Load-displacement curve for the $sl/s - in$ delamination.

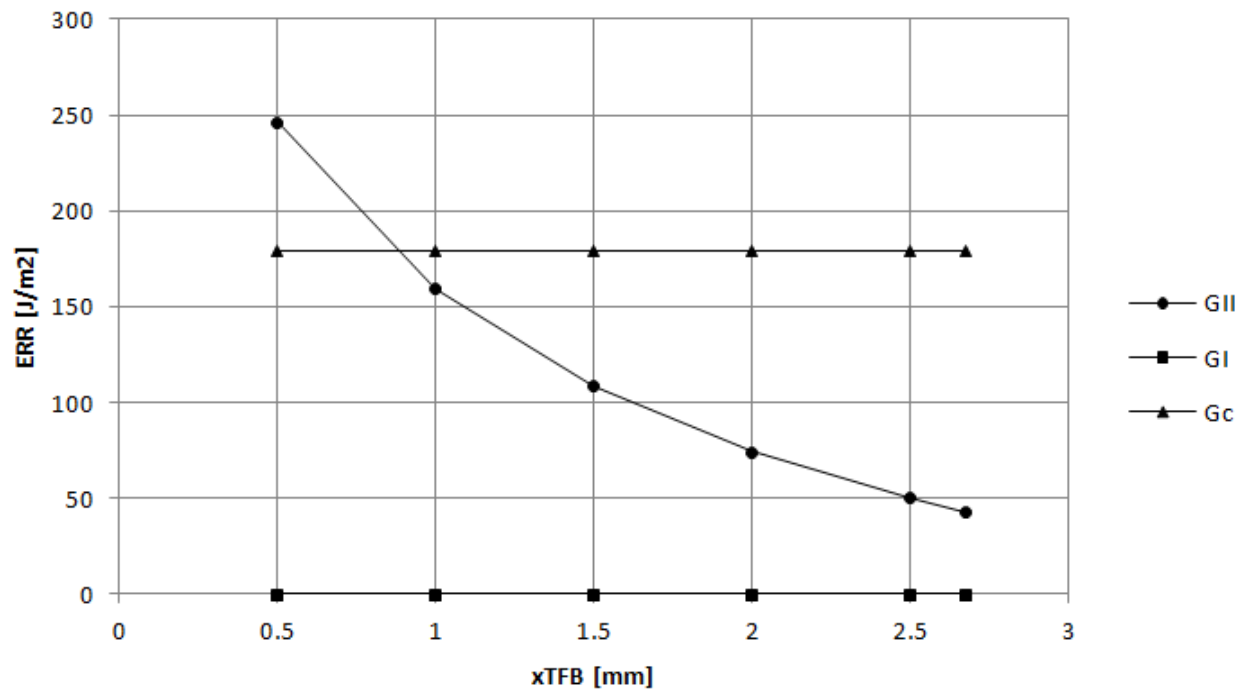


Figure 41. Four point bending $[TFB/AF/(-45/90/45/0)_{3S}]$. Strain energy release rates at $sl/s - in$ under ultimate load.

The crack length control scheme simulation of the $sl/s - out$ delamination computed a critical unstable delamination from the TFB outer edge to the active components, Figure 42. The driving force curve resulted similar to the compression case for G_{II} , but there was also a large component of G_I , which was absent in the compression case. Therefore this tension case classified as critical.

A summary of all the failure modes for the externally bonded TFB configuration is shown in Table 4.

	$CFRP/af$	af/sl	ac/s	$sl/s - in$	$sl/sn - out$
Curv.+comp.	not critical	not critical	critical	critical	not critical
Tens.	critical	critical	not critical	not critical	not critical
Curv.+tens.	critical	not critical	critical	not critical	critical

Table 4. Summary of the critical failure modes for the externally bonded TFB configuration.

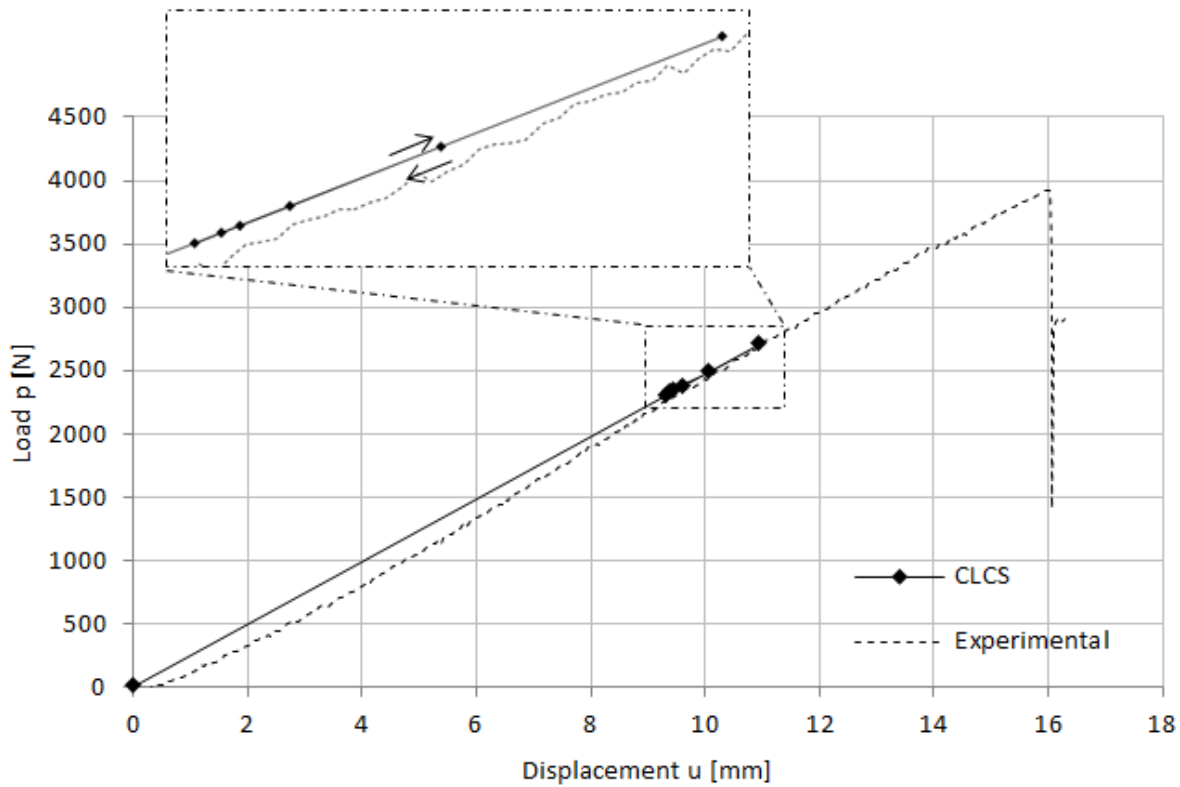


Figure 42. Four point bending of $[TFB/AF/(-45/90/45/0)_{3S}]$. Load-displacement curve for the $sl/s - out$ delamination.

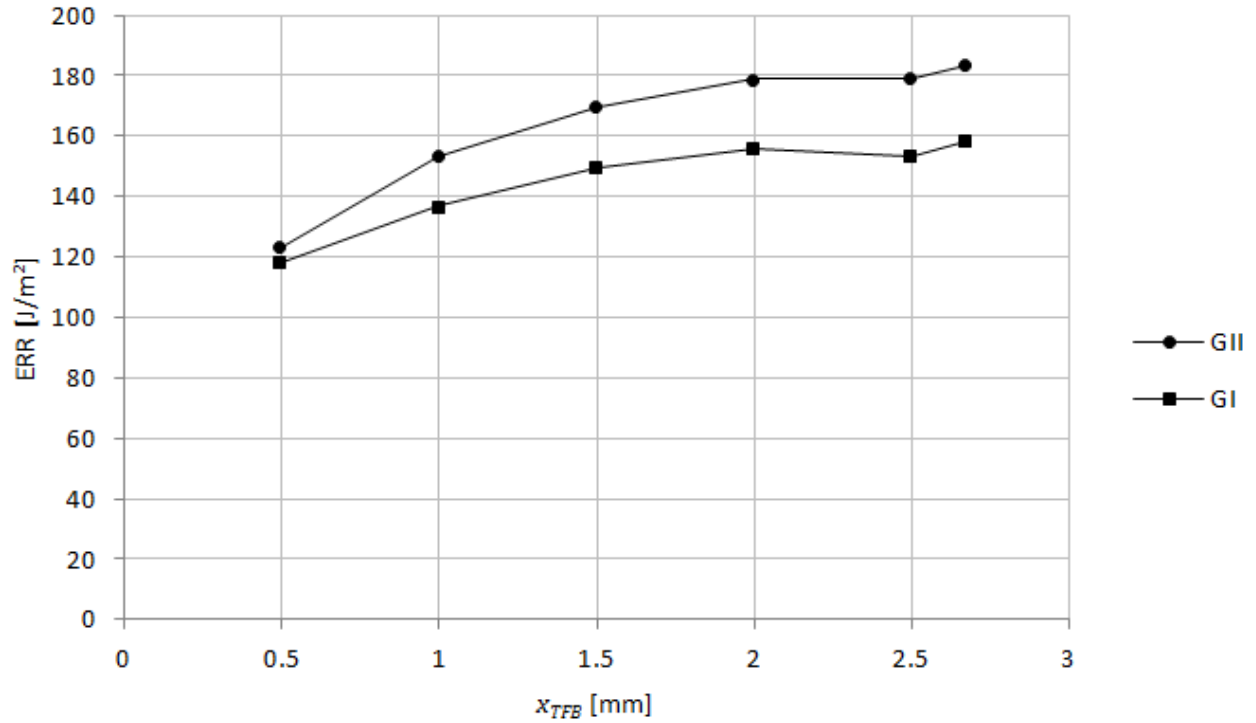


Figure 43. Four point bending [TFB/AF/(-45/90/45/0)_{3S}]. Strain energy release rates at *sl/s – out*.

4.5.2 Embedded configuration

4.5.2.1 Uniaxial tension

The progressive failure of the embedded battery configuration under uniaxial tension was assessed by simulating the tension test of the (0/45/90/-45/TFB/-45/90/45/0) laminate. A close-up of the finite element model at TFB termination is shown in Figure 44. The experiment reported in chapter 2 showed a critical electrical failure at an applied far field strain of 6375 μ strain. The causes of the failure were unknown.

The *CFRP/sl* delamination propagation resulted stable and not critical, Figure 45. The ERR at the critical experimental load level decreased rapidly with delamination length. Therefore this failure mode was not critical.

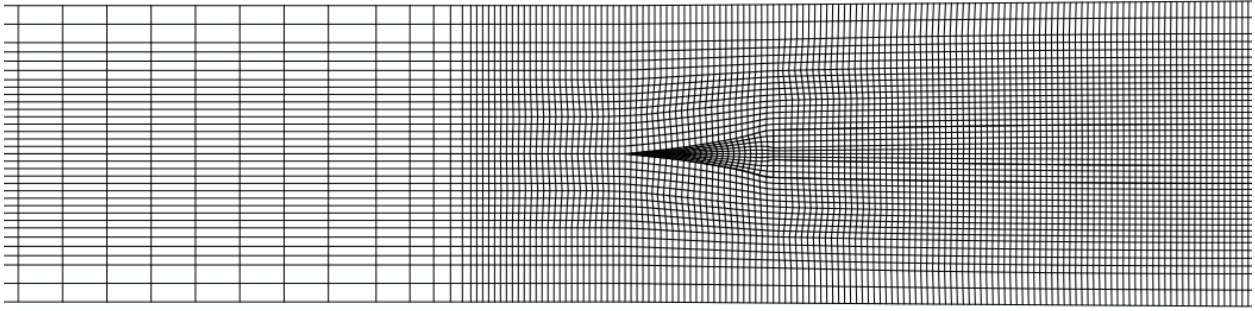


Figure 44. Close-up of uniaxial tension specimen FEM at TFB termination.

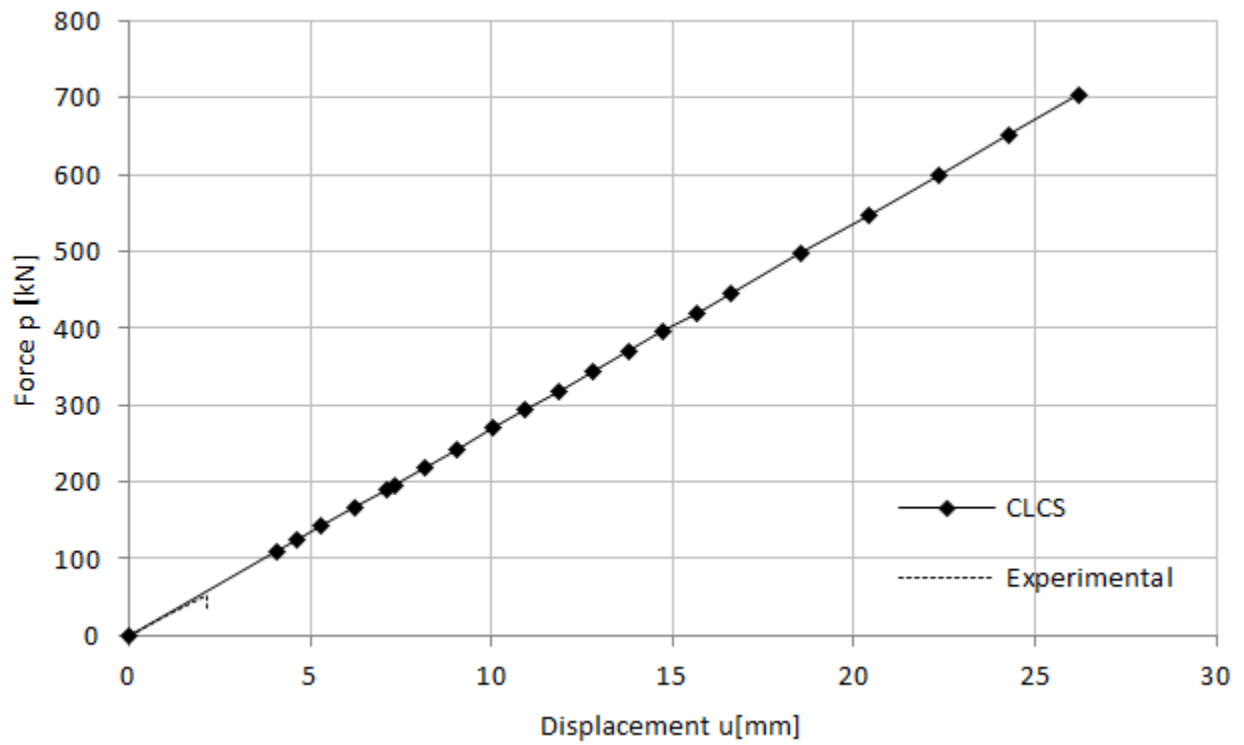


Figure 45. Uniaxial tension of (0/45/90/-45/TFB/-45/90/45/0). Load-displacement curve for the *CFRP/af* delamination.

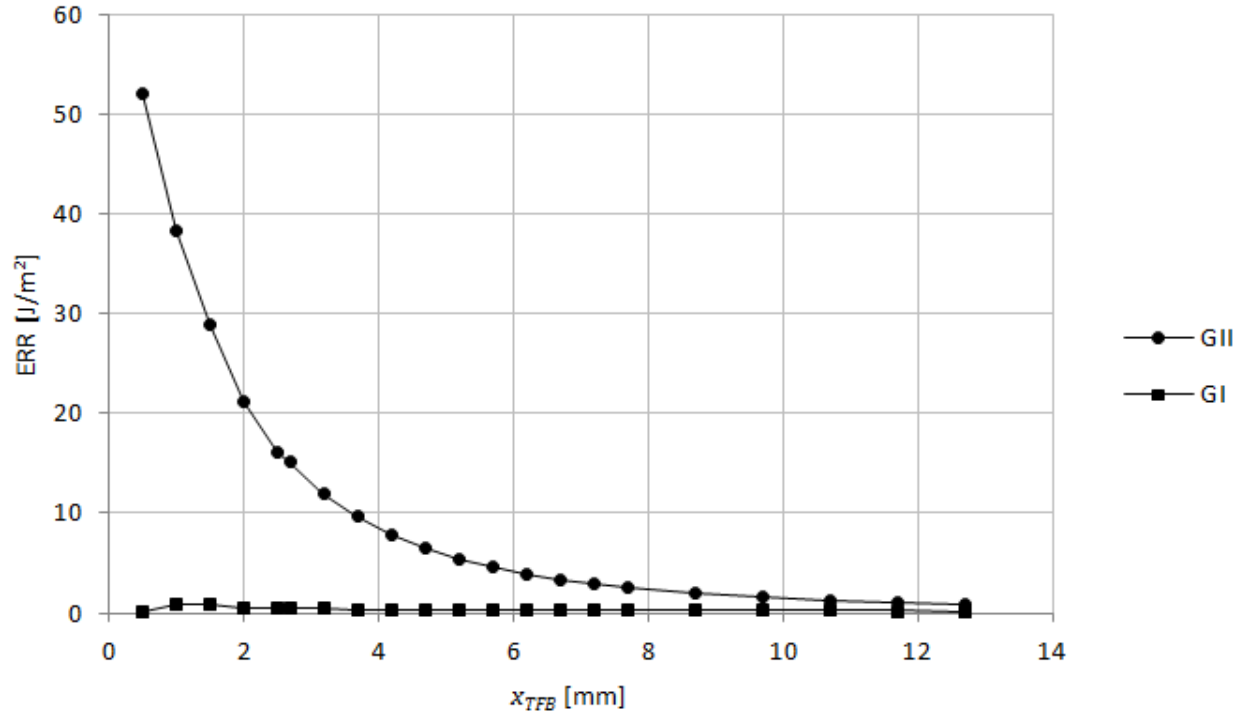


Figure 46. Uniaxial tension of (0/45/90/-45/TFB/-45/90/45/0). Strain energy release rates at $CFRP/af$. Applied far field strain $\varepsilon_x = 6375 \mu\text{strain}$.

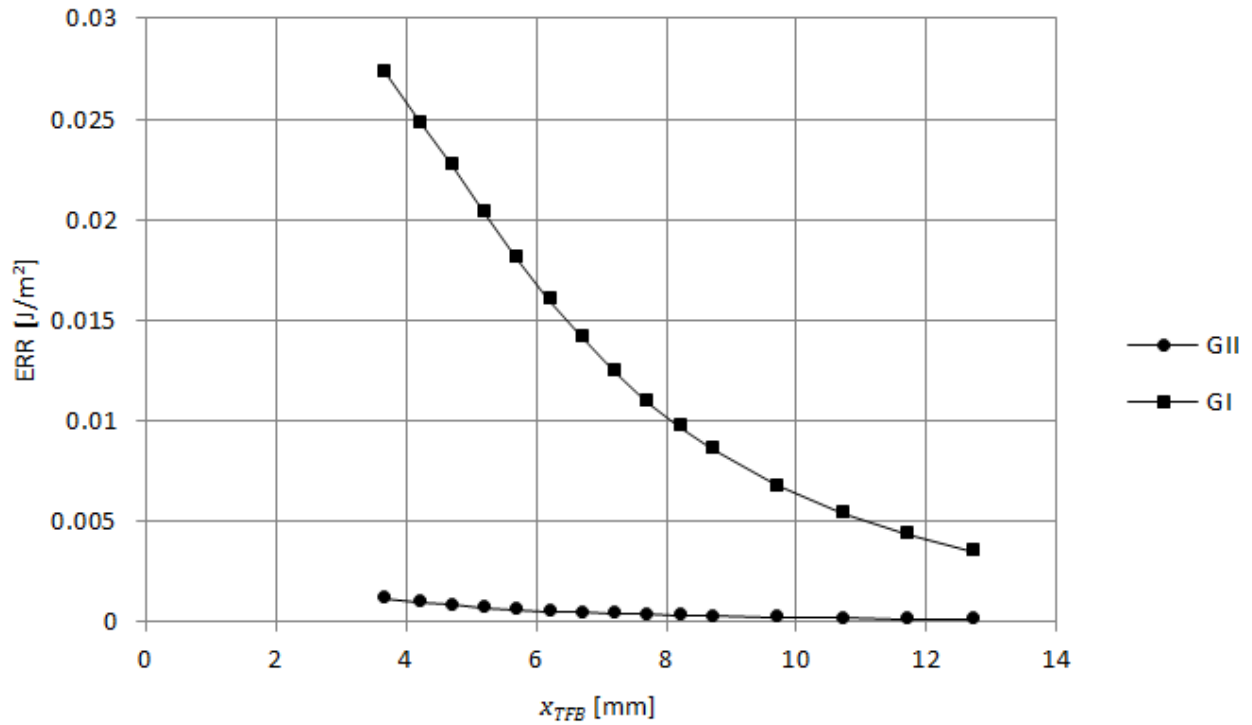


Figure 47. Uniaxial tension of (0/45/90/-45/TFB/-45/90/45/0). Strain energy release rates at ac/s . Applied far field strain $\varepsilon_x = 6375 \mu\text{strain}$.

Also the *ac/s* delamination, modeled with the usual initial length of 1 mm, showed stable propagation. The mode I dominate driving force curve showed that a fracture toughness of 0.03 J/m^2 was necessary to avoid propagation. This ERR value was the highest at the active components among the embedded configuration loading conditions and it was therefore classified as critical. Although the value was modes compared to structural materials, we cannot exclude that the fracture toughness of the *ac/s* interface was lower and the electrical failure was caused by this delamination. However this event was deemed unlikely because the propagation would have taken place for a short length due to the rapidly decreasing G_I .

The *sl/s – in* delamination was modeled in the same way described earlier and with an initial length of 1 mm. It propagated with an unstable fashion, but at a not critical load level, Figure 48. Due to the small variation in the specimen stiffness upon propagation, the softening and elastic branch are almost coincident. The sharply increasing G_I as the delamination approaches the TFB end, is associated with the stress concentration at TFB end and to the positive ε_y strain documented by the stress analysis, Figure 49. This failure mode was deemed critical for the *sl/s – in* delamination in the embedded TFB configuration. This classification was based on the low fracture toughness of the muscovite substrate. However of a more structural substrate material this failure mode could be considered not critical.

The *ls/s – out* showed a stable propagation and the ERR values were lower than for the *ls/s – in* delamination. This failure mode was therefore classified not critical because it was covered by the *ls/s – in* delamination occurring at the same interface.

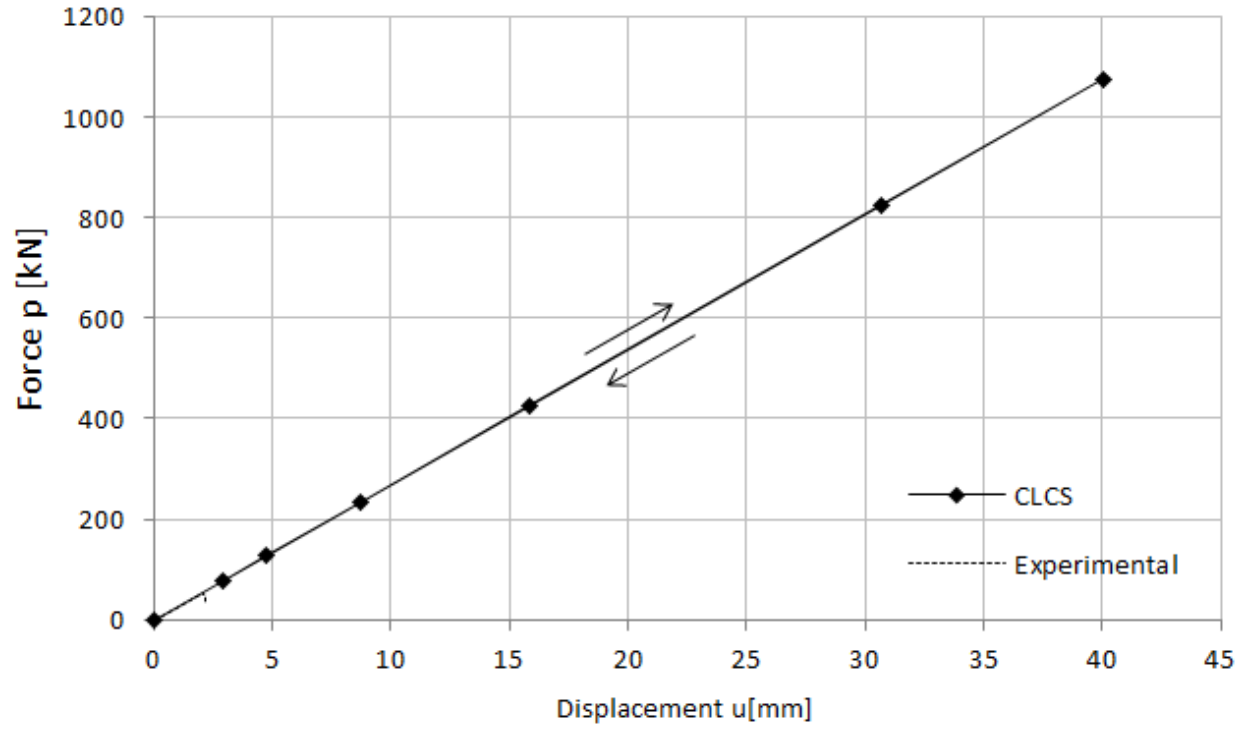


Figure 48. Uniaxial tension of (0/45/90/-45/TFB/-45/90/45/0). Strain energy release rates at $ls/s - in.$

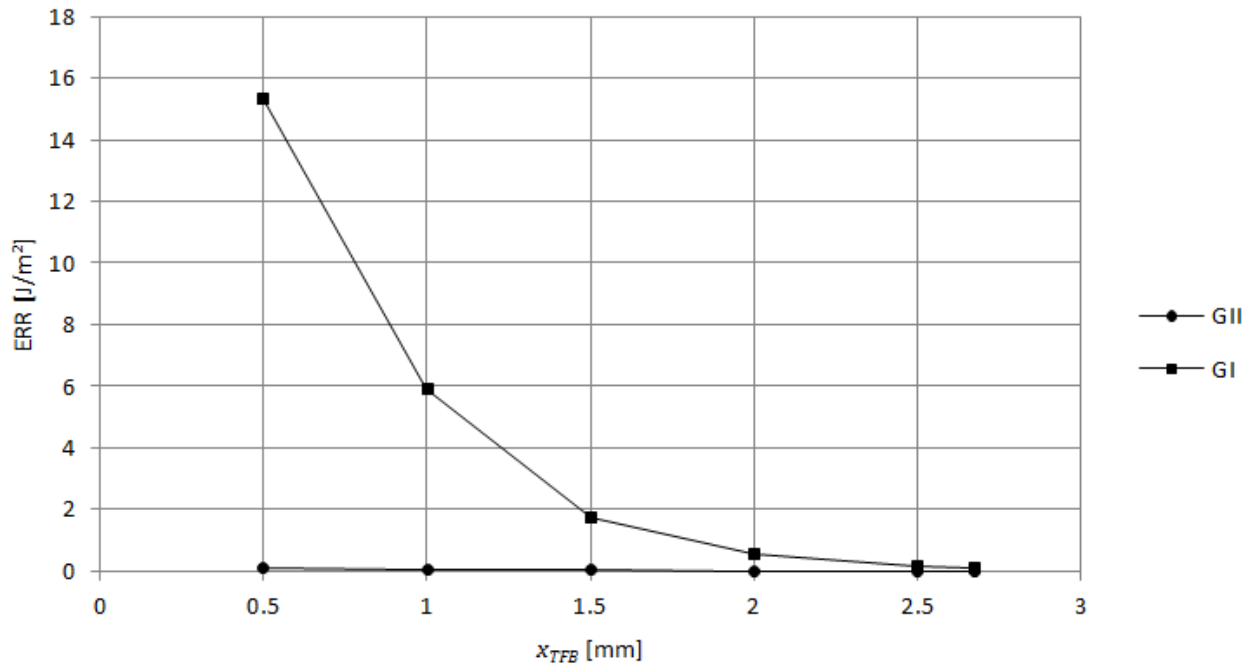


Figure 49. Uniaxial tension of (0/45/90/-45/TFB/-45/90/45/0). Strain energy release rates at $ls/s - in.$ Applied far field strain $\varepsilon_x = 6375 \mu strain.$

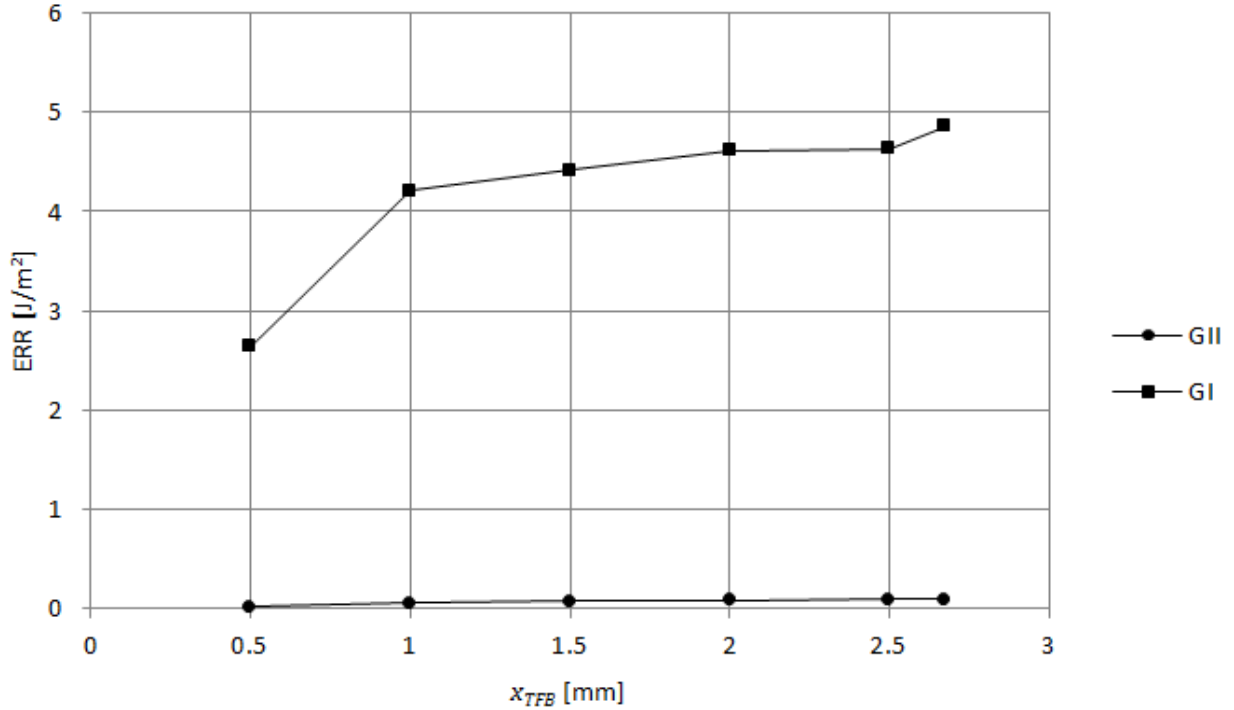


Figure 50. Uniaxial tension of (0/45/90/-45/TFB/-45/90/45/0). Strain energy release rates at *ls/s – out*. Applied far field strain $\varepsilon_x = 6375 \mu\text{strain}$.

4.5.2.2 Four point bending

The progressive delamination failure of the embedded TFB configuration under pure curvature was assessed by simulating the four point bending test of the $[(0/45/90/-45)_3/\text{TFB}/(-45/90/45/0)_3]$ laminate.

The calculated *CFRP – sl* delamination propagation characteristics were not critical, Figure 51.

The *ac/s* delamination, modeled in the usual way, showed stable propagation at negligible ERR values. This failure was classified not critical because less critical than the *ac/s* delamination occurring under uniaxial tension.

The *sl/s – in* was modeled in the usual way, with an initial delamination length of 1mm. This delamination resulted not critical as shown by the driving force curve, Figure 53. The delamination propagated with an unstable fashion, as seen in previous loading conditions for this interface, but the ERR values were negligible.

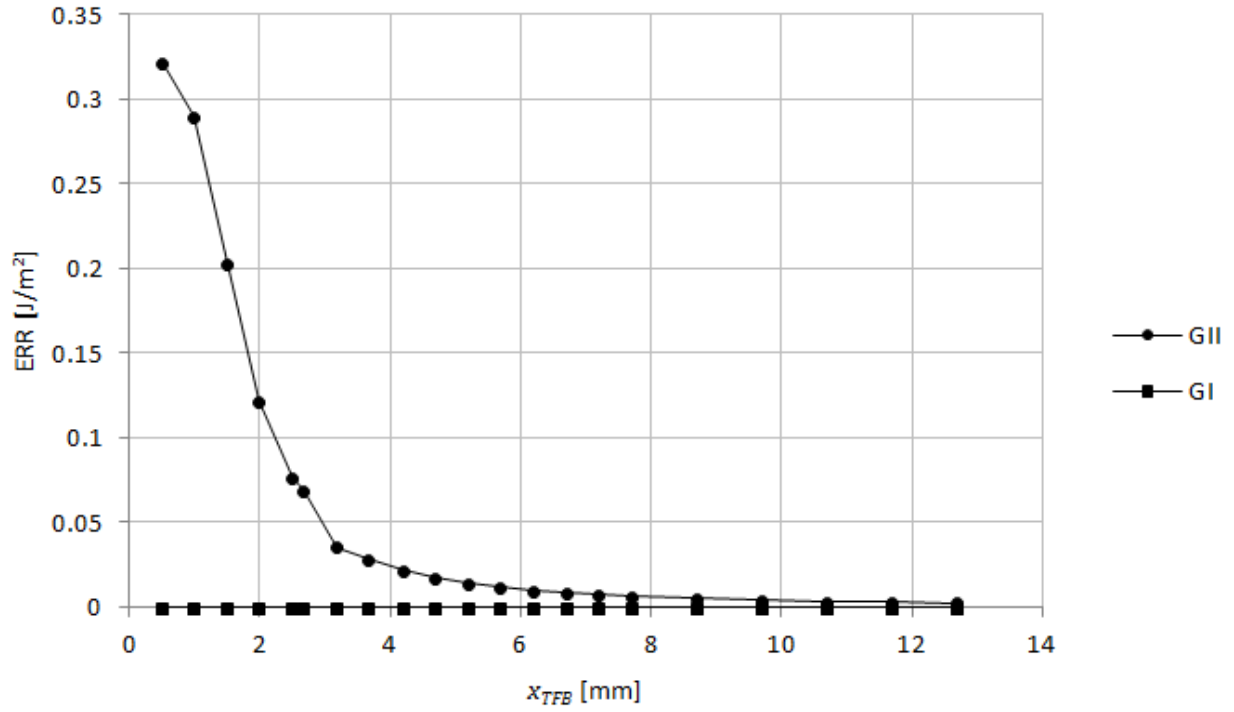


Figure 51. Four point bending of $[(0/45/90/-45)_3/TFB/(-45/90/45/0)_3]$. Strain energy release rates at $CFRP/sl$ under ultimate load.

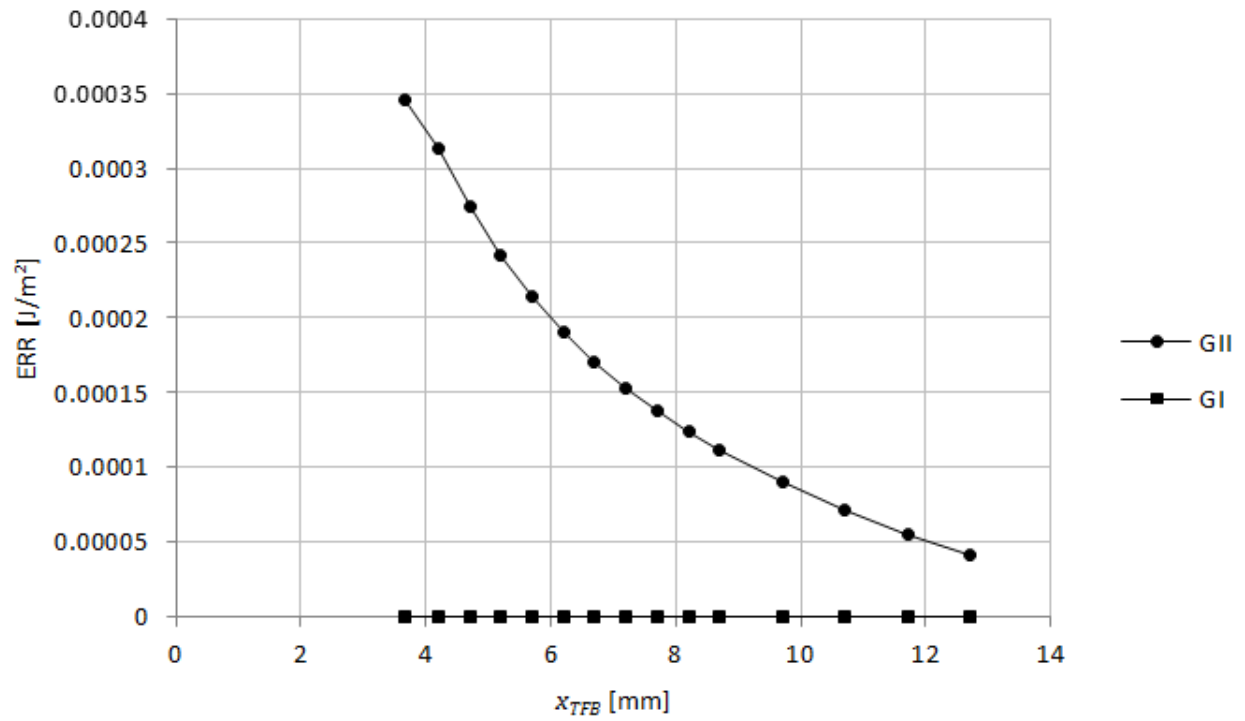


Figure 52. Four point bending of $[(0/45/90/-45)_3/TFB/(-45/90/45/0)_3]$. Strain energy release rates at ac/s .

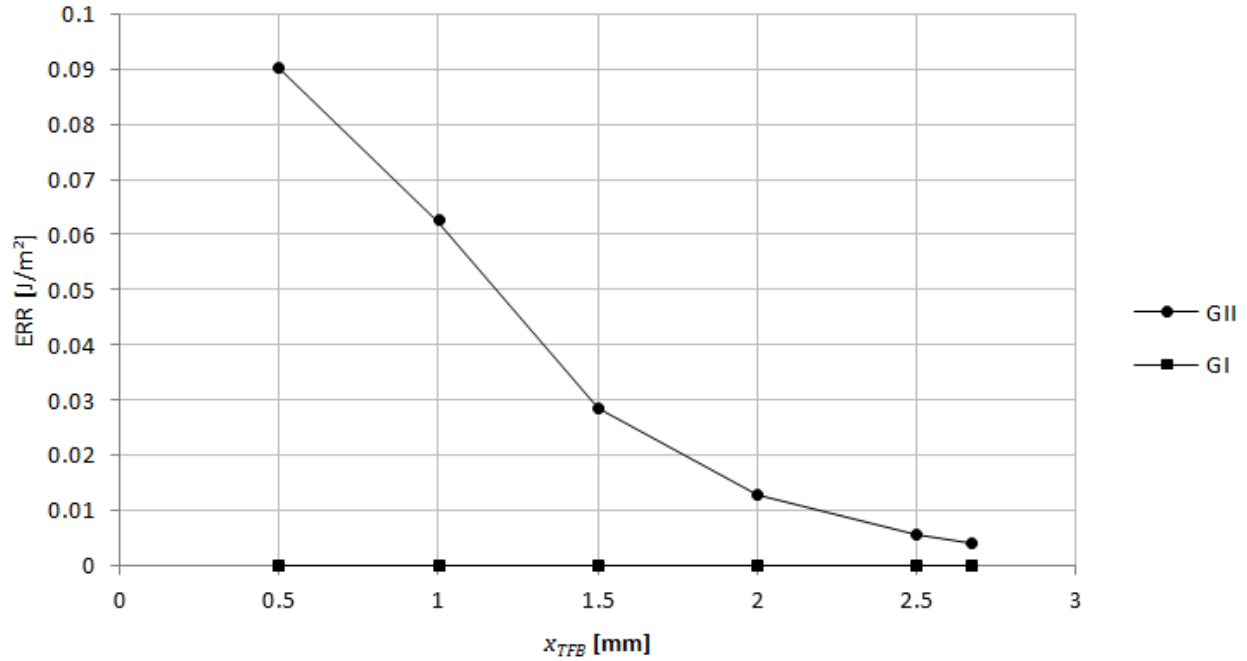


Figure 53. Four point bending of $[(0/45/90/-45)_3/TFB/(-45/90/45/0)_3]$. Strain energy release rates at $sl/s - in$.

The $ls/s - out$ delamination was not critical because of the negligible ERR dissipated. The G_{II} distribution decreased as the delamination progressed, as opposed to the externally bonded configuration for the same interface.

All the results confirmed that curvature for the embedded configuration was not critical, in agreement with the experimental evidence.

A summary of the failure modes for the embedded TFB configuration is shown in Table 5.

	<i>CFRP/sl</i>	<i>ac/s</i>	<i>sl/s - in</i>	<i>sl/s - out</i>
Curvature	not critical	not critical	not critical	not critical
Tension	not critical	critical	critical	not critical

Table 5. Summary of failure modes for the embedded TFB configuration.

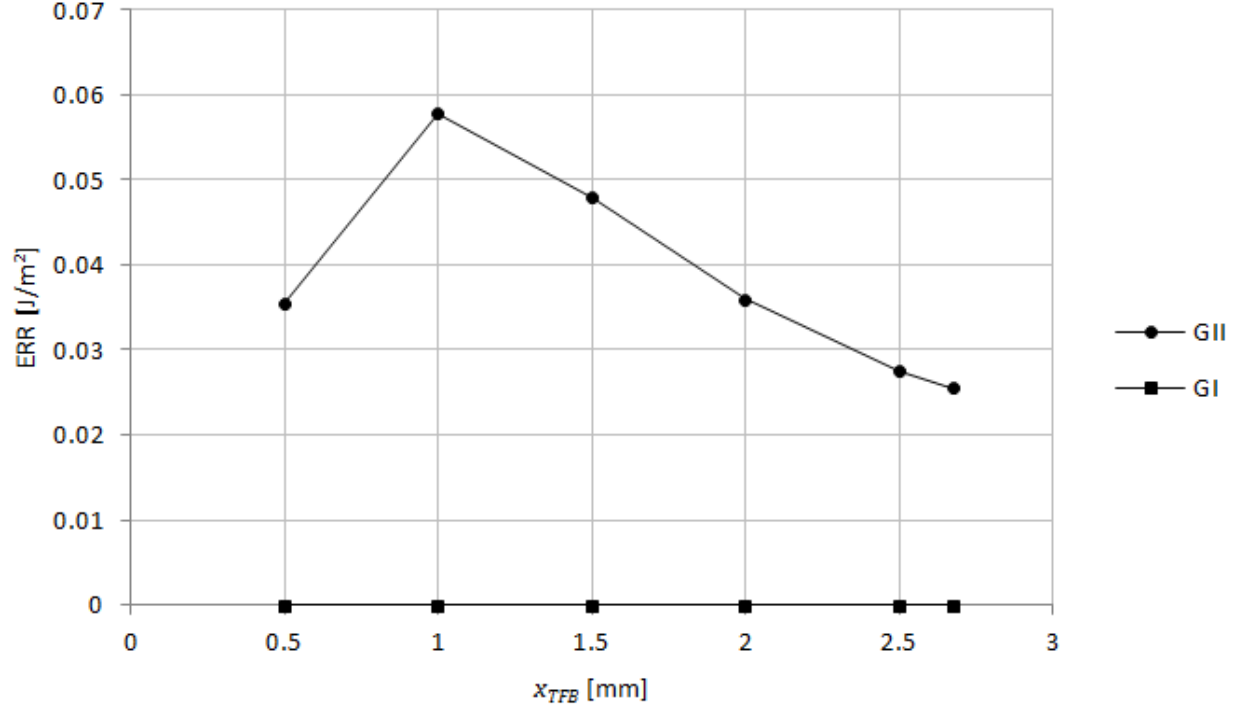


Figure 54. Four point bending of $[(0/45/90/-45)_3/TFB/(-45/90/45/0)_3]$. Strain energy release rates at $sl/s - out$.

4.6 Parametric study

The objective of the parametric study was to determine the requirement for fracture toughness at the TFB active components as a function of the substrate and sealant moduli of elasticity. The choice of focusing on one interface, the ac/s as defined in the previous section, was dictated but the observation that the requirement for fracture toughness, established by the above analysis, would be the most difficult to satisfy, even with the next generation of nanostructured electrode and solid electrolyte materials. The critical fracture toughness of the active components associated with the current TFB materials was determined by the progressive delamination analysis to be 51 J/m^2 ($G_I + G_{II}$). This value was required by the most critical failure mode of the ac/s interface, which was the delamination buckling under the simultaneous occurrence of curvature and compression.

A typical fracture toughness value for the current Li-ion batteries electrode materials is lower than 10 J/m^2 . As an example, an amorphous silicon anode on a copper substrate reaches 7.7 J/m^2 [12]. More studies are being performed on nanomaterials, such as graphene [13] for example, which could potentially

increase the toughness. However available data do not allow to predict a toughness level for the next generation materials. Therefore determining the optimum packaging materials that minimize the toughness requirement is an enabling design step.

The parametric study was performed by considering the critical loading conditions for the *ac/s* delamination. These conditions are listed in Table 4 for the externally bonded configuration and in Table 5 for the embedded configuration. Two separate parametric studies were performed in order to compare the effectiveness of the two opposite laminate configurations.

The automatic crack extension scheme analysis was iterated for varying sealant elastic modulus E_{seal} and substrates elastic modulus E_{sub} . The applied loads were the ultimate loads reported in Table 2. For every parametric iteration (p) a different material set was analyzed. At iteration (p) the total strain energy release rate distribution G_T was computed from the resulting distributions of G_I and G_{II} . The required fracture toughness for the particular material set was computed as the maximum G_T value

$$G_C^{(p)} = \max[G_T(x_{TFB})] \quad (6)$$

The $G_C^{(p)}$ values were then used to generate performance plots of G_C as a function of E_{seal} and E_{sub} .

For the externally bonded configuration this procedure was applied separately to the two critical loading condition, which were the four point bending of the [TFB/AF/(-45/90/45/0)_{3s}] laminate and the [(0/45/90/-45)_s/AF/TFB] laminate (Table 4). Thereafter the two performance plots were merged into a final plot that enveloped the two failure modes. The same procedure was also applied to the embedded TFB configuration. In this case there was only one critical loading condition, namely the uniaxial tension of the [(0/45/90/-45)₃/TFB/(-45/90/45/0)₃] laminate (Table 5), therefore only one performance plot was created. For the other interfaces, analogous studies could be carried out. However the study presented in present work was limited to the *ac/s* interface.

Therefore a performance plot was defined for a given geometry and failure mode. The geometry was kept constant and equal to the TFB-CFRP laminates experimentally characterized in chapter 2. For a different geometry, which could consist in a simple thickness variation for example, the parametric study should be

repeated with modified finite element models. The substrate and sealant materials were isotropic, linear elastic materials with a Poisson's ratio equal to 0.3.

Since the values of fracture toughness were computed from the energy release rate dissipate upon delamination propagation, the curves shown in the following figure should be considered the maximum value of fracture toughness at which failure occurs. Therefore the fracture toughness required to avoid propagation should be higher than G_C . In other words the design should be above the relevant curve of the performance plot.

The performance plot for the $[(0/45/90/-45)_S/AF/TFB]$ laminate loaded in four point bending is shown in Figure 55. The G_C was a strong function of E_{sub} and E_{seal} . The sharp increase in G_C was caused by elastic instability of the upper TFB substrate. For low elastic modulus of the substrate, a small fracture toughness is necessary to avoid propagation, and therefore buckling. By increasing E_{sub} the instability can be avoided by increasing the sealant modulus. This can be explained by two beneficial effects that a higher E_{seal} produced. The first consist in increasing the critical buckling load. The second is related to the stress-strain field and to the fact that a higher modulus sealant reduced the stress level, and therefore the energy release rate, at the edge of the TFB active components where the ac/s delamination was located.

A smoother performance plot was obtained for the $[TFB/AF/(-45/90/45/0)_{3S}]$ laminate loaded in four point bending, Figure 56. The result confirmed the beneficial effect of a higher E_{seal} to reduce the required fracture toughness. The two plots were merged by enveloping the G_C curves, thus obtaining the final plot for the ac/s delamination, Figure 57. The x-axis was rescaled to highlight the optimum region for design. In fact, by selecting the appropriate values for E_{seal} and E_{sub} , the required fracture toughness of the active components could be reduced to a minimum. The dominant failure mode that highly affects material selection is delamination buckling. For a given sealant modulus, the substrate modulus should not be too high, otherwise buckling would occur. Therefore the higher the sealant modulus the

more structural the battery can be designed by increasing the substrate modulus. But also the strength of the TFB would be higher because a lower fracture toughness would be required.

The performance plot for the embedded TFB configuration is shown in Figure 58. In this case the required fracture toughness was less a strong function of the packaging materials. More importantly, the required fracture toughness at optimum design was one order of magnitude lower than the externally bonded configuration.

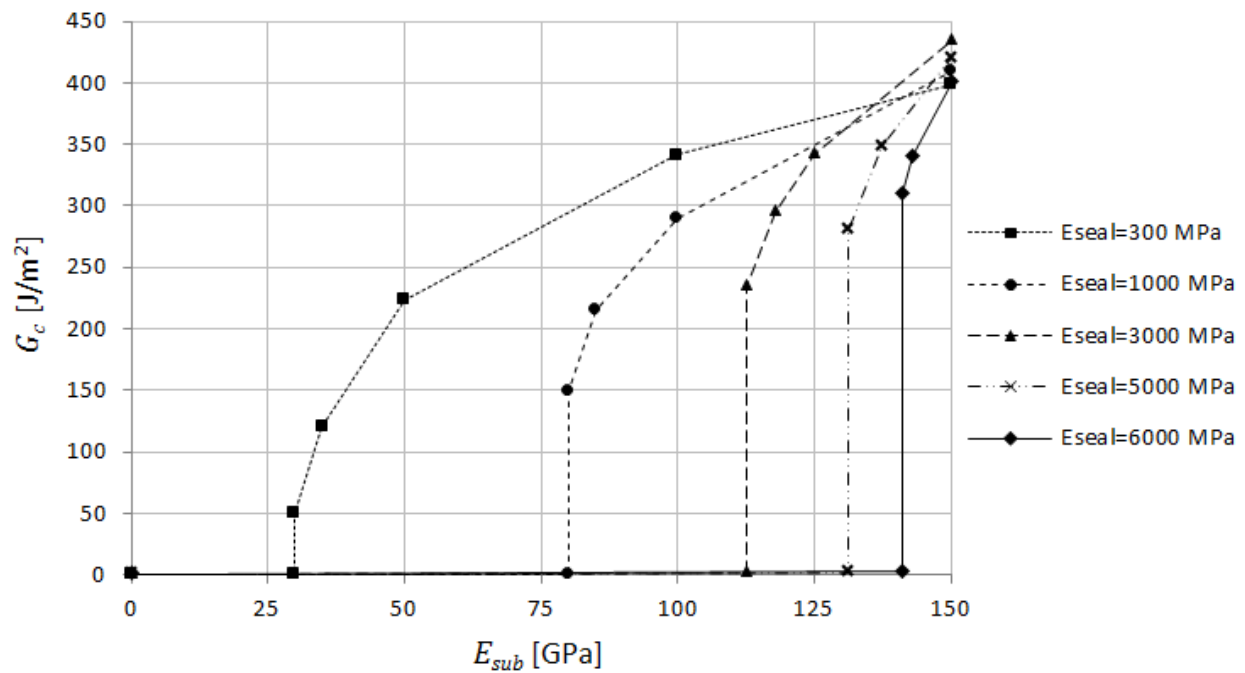


Figure 55. Performance plot for four point bending test of [(0/45/90/-45)_s/AF/TFB]. Delamination ac/s .

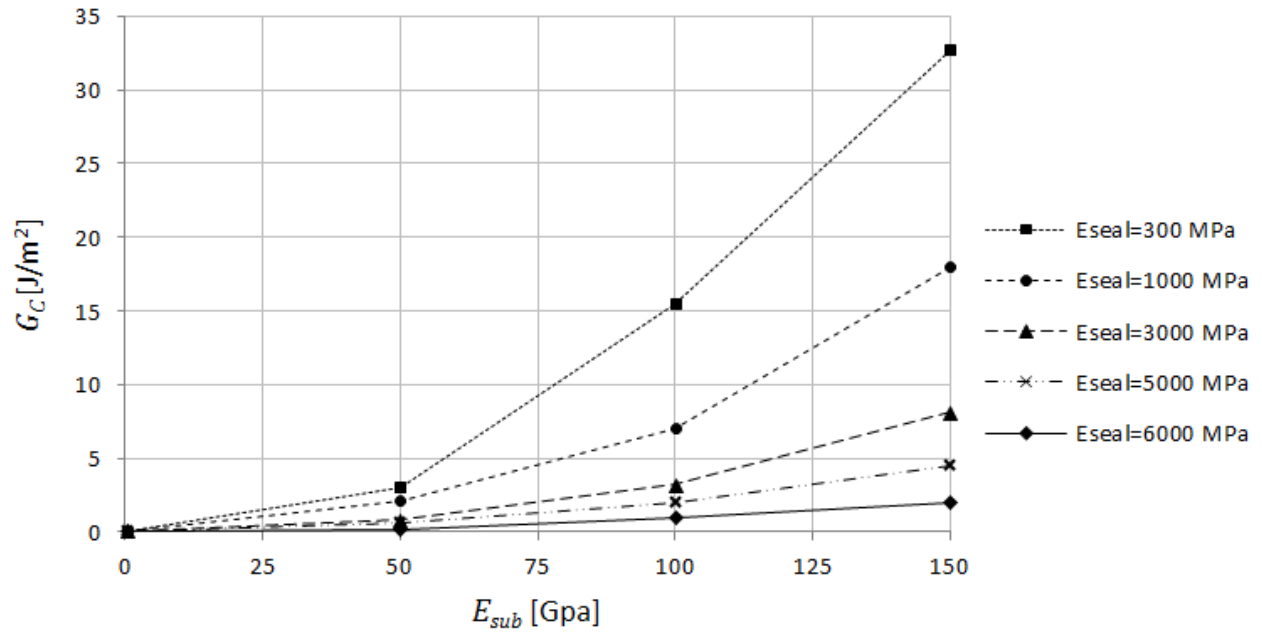


Figure 56. Performance plot for four point bending test of [TFB/AF/(-45/90/45/0)_{3s}]. Delamination ac/s .

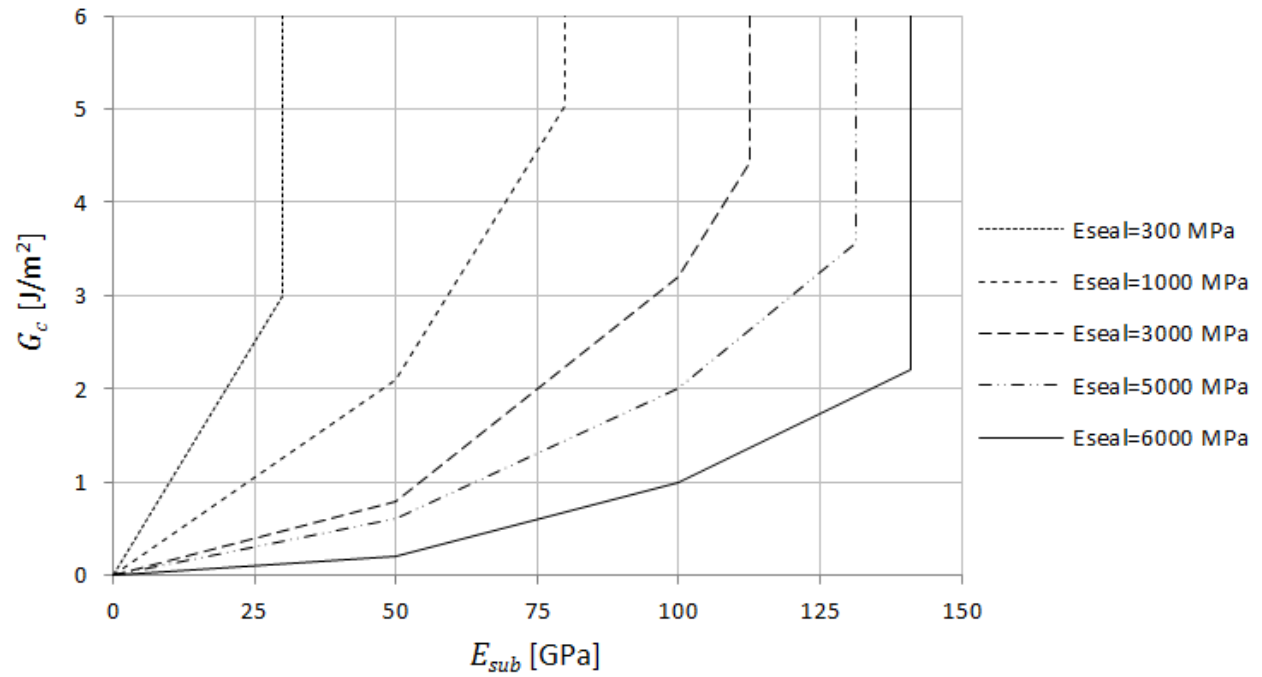


Figure 57. Performance plot for the externally bonded configuration. Delamination ac/s .

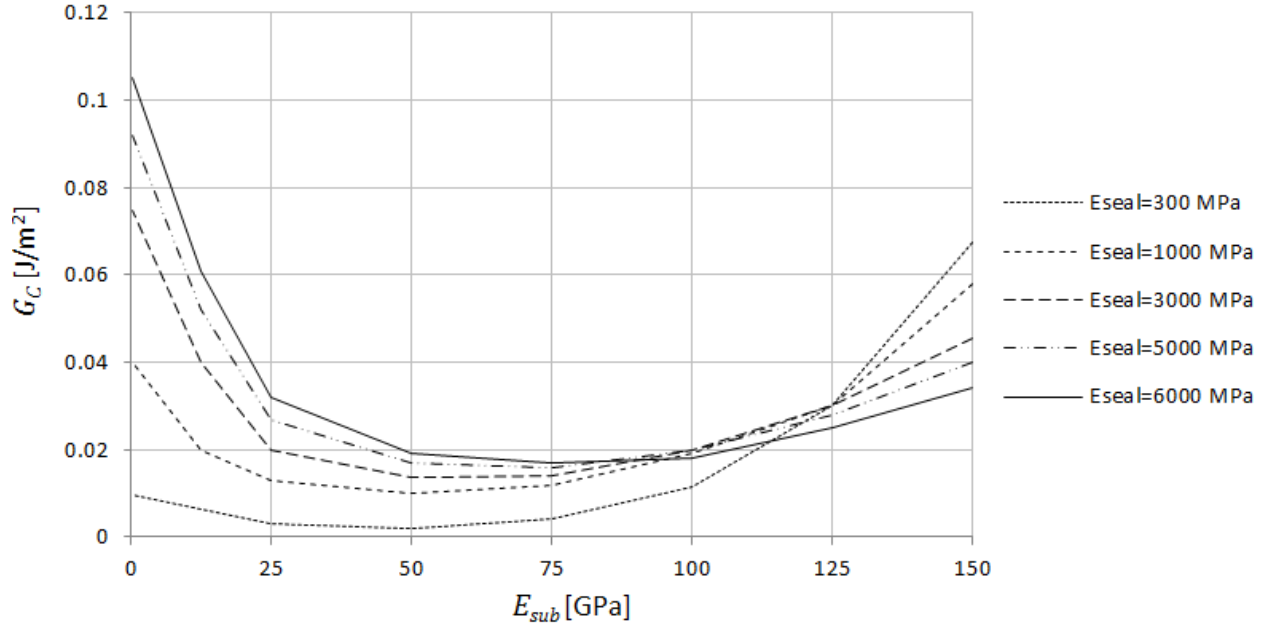


Figure 58. Performance plot for the embedded configuration. Delamination ac/s .

4.7 Conclusions

The study demonstrated the effectiveness of the crack length control scheme in the analysis of the TFB-CFRP laminates. These inhomogeneous laminates were particularly prone to sharp snap-back mechanical responses because of the low fracture toughness at some interfaces and of the short initial delamination length. This behavior was also amplified by elastic instabilities such as delamination buckling of the externally bonded TFB configuration.

The progressive delamination analysis showed a reasonable agreement with the experimental failures and allowed a better understanding of the underlying failure mechanisms. The results showed, for example, that the premature electrical failure under uniaxial tension documented by the experimental test campaign was not caused by any of the delaminations considered. That failure could be therefore attributed to the cracking of one of the electrode or electrolyte materials that reached its critical strain to failure. To corroborate this hypothesis it is useful to note that the electrolyte was a ceramic material with a low expected strain to failure. However, more test repetitions would be necessary to assess the variability of the critical load. Furthermore, the unstable and catastrophic delamination at the interface between the

adhesive film and the TFB substrate (af/sl) were confirmed by the analysis. Finally, the matching with the experimental results allowed to estimate the current fracture toughness of the active component/sealant interface (ac/s) to be certainly lower than 40 J/m^2 for mode II and 11 J/m^2 for mode I. The two considered configurations, embedded and externally bonded, represented the two extremes of several possible intermediate scenarios. The embedded configuration showed larger margins of safety and less critical failure modes than the externally bonded configuration. Since the analysis of pure compression was not performed, this observation is limited to the tension dominated loading conditions considered. Embedding seemed to be particularly advantageous for the low fracture toughness required by the active components. However, the embedded configuration requires a more complex manufacturing process.

The analysis determined the propagation characteristics, critical loading conditions, onset locations and critical initial delamination lengths for all the failure modes identified by the experimental test campaign and the stress analysis. The recurrent disbonding of the TFB packaging noted at the edge of the active components resulted critical for the integrity of the battery. In fact, it showed unstable propagation throughout the full length of the peripheral TFB seal ($ls/s - in$), creating a path for contaminants such as air and moisture. It showed also potentially unstable propagation between the active components and sealant interface in case of compressive applied loading. This unstable propagation was caused by the elastic instability of the TFB substrate for the externally bonded configuration. This failure mode appeared particularly critical since the active components are, in general, characterized by particularly low fracture toughness. The delamination buckling, as for failure of the peripheral seal, exposes the highly reactive anode and electrolyte materials to lethal contaminations.

The critical delamination lengths for the TFB packaging, either at the substrate/sealant or at the active components/sealant interfaces, ranged between 1.5 and 3 mm for the current packaging materials. Therefore the manufacturing process of the structural batteries should be able to exclude defects of this size.

The delaminations that originate outside the battery, at the interface between the TFB and the CFRP plies, are also characterized by unstable propagation. However, these failure modes seemed to be less critical for the design of the next generation of TFB-CFRP laminates because they can be managed by well-established structural composite materials.

The parametric analysis showed that an optimized packaging design could greatly improve the strength and structural efficiency of the TFB-CFRP laminate. The optimum combination of sealant and substrate modulus of elasticity could reduce the fracture toughness requirement for the active components to 3 J/m^2 for the externally bonded configuration and to 0.1 J/m^2 for the embedded configuration. However, the higher is the substrate modulus to achieve more structural performance, the higher should be the sealant modulus in order to keep the toughness requirement at a reasonably low level. A high modulus substrate material such as Titanium, for example, would require a sealant with a modulus of at least 3000 MPa or higher. Structural epoxies are in that range, but their compatibility to the TFB manufacturing process needs to be proved.

These results were referred to the given TFB geometry utilized for the present study. Moreover, the study did not consider pure compression loading. Therefore the general results and the performance plots presented herein should be taken as a preliminary design study to assess the feasibility of TFB-CFRP laminates for structural members loaded mostly in tension. For more specific purposes, the analysis method proposed in this work should be applied to the specific design and loading conditions under consideration.

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5. Conclusions and future work

5.1 Conclusions

The compatibility of state-of-the-art thin film Li-ion batteries with the curing environment of carbon/epoxy composite materials was experimentally assessed through environmental testing. The study, which investigated for the first time the effect of the exposure of quiescent batteries to the environmental conditions of composites manufacturing, gave evidence that temperature is the most influential parameter for battery survivability. Batteries could be successfully cured up to 149°C preserving long- and short-term capacity. If this temperature is exceeded, different types of failures caused by the physiochemical degradation of the anode and the electrolyte occur. The failures appeared to be triggered by the melting of the battery thermoplastic sealant, which is therefore the most critical limiting factor from a processing standpoint.

The experimental characterization of the multifunctional laminates showed that the electrochemical performance is unaffected by the applied mechanical loading up to a mechanical failure. Therefore the analysis and design could exclusively focus on mechanics. The elastic response of the laminates is characterized by a three-dimensional stress-strain field which could not be predicted with the classical laminate theory, which is based on the plane stress assumption. In terms of failure, the low fracture toughness and the inhomogeneity of the material properties of the current battery packaging design led to critical unstable delaminations. These failures occurred at about 4900 μ strain of applied uniaxial strain in tension, and -2800 μ strain in compression. These strain values are less than half of the desired strain to failure. The critical failure mode under applied tension strain consisted in the unstable delamination propagation within the battery muscovite substrate. Therefore a high-temperature battery substrate material with increased fracture toughness is required for the next generation of structural thin film batteries. The simultaneous occurrence of elastic instability and unstable delamination propagation, namely delamination buckling, was the critical failure mode under applied compression.

Hence, in order to gain insight in the failure mechanisms, a comprehensive finite element method to combine three-dimensional stress-strain field and unstable delamination propagation analysis was necessary. The proposed load-displacement-constraint method, the *crack-length control scheme*, proved to be effective in capturing sharp snap-back instability that characterizes the brittle delamination failure of structural composites and in particular the finely meshed multifunctional laminates under consideration. The method provides the envelope of stable and unstable delamination growth and delamination buckling collapse responses by tracing the complete equilibrium path of the structure with continuation through singular points.

Although it does not guarantee convergence, the *crack-length control scheme* showed improved convergence rate and robustness with respect to the conventional VCCT implementation in the Newton-Raphson method. The enhanced computational efficiency was attributed to three reasons. The first was the capability of advancing the delamination by multiple element lengths. The second consisted in controlling the load increment size by means of the energy release rate, thereby avoiding unduly expensive load cutbacks. The last was the automatic selection of the largest increment size that ensured the computation of the structural response with the desired resolution. The use of the delamination length as a constrained variable, instead of the nodal displacements used by the arc-length method, was the main reason for the robustness of the method. Since the delamination length is monotonically increasing with time, the algorithm never fails in selecting the correct equilibrium path direction, even in presence of arbitrarily sharp singularities in the structural response. The user can also reasonably establish a priori, based on the geometry of the problem under consideration, an upper and lower limit to the incremental variation (Δa) of the delamination length. These limit values can be utilized in place of the values computed by the constraint equations to overcome inconsistencies in the *crack length control scheme* that could arise in case of a highly nonlinear structural response such as delamination buckling. Moreover, by extending the delamination under constant applied displacement intervals, the unstable propagation of the delamination can be computed without the use of an artificial stabilization technique.

The drawback of the *crack length control scheme* consists in the dependency of the constraint equation and convergence criteria to the delamination topology, i.e. to the element connectivity. As a result, the delamination has to be tracked constantly during the solution process, leading to a cumbersome addition to a finite element code. This requirement seems to be the main limitation for the extension of the method to three-dimensional problems. In addition, at this stage, the method is not compatible to multi-delamination problems and the possibility to implement this functionality was not assessed.

The computational efficiency and robustness of the analysis method allowed to match the experimental results and to perform a parametric study on the battery packaging design variables. The analysis showed that the elastic modulus mismatch between the battery substrate and sealant was the main cause for the premature delamination buckling failure. Moreover, it demonstrated that the packaging design is a determining factor for the integrity of the multifunctional laminates. More specifically, with an appropriate selection of the substrate and sealant moduli, the fracture toughness requirement for the battery active components, which is a critical design variable, can be minimized.

The study provided a design space for improving the limits of utilization for the next generation of structural thin film batteries. The use of this technology seems to be possible for low stress applications and secondary structures with short lifecycle, provided that the next generation of electrode and electrolyte materials have a fracture toughness of at least 5 J/m^2 . For structural applications (i.e. substrate Young's modulus greater than 70 Gpa) a structural sealant with Young's modulus greater than 1000 Mpa is required.

5.2 Future work

The next step in the development of the battery packaging should include the manufacturing of a second generation structural thin film battery. The novel design should feature new substrate and sealant materials, with values for elastic moduli as close as possible to the optimum values calculated in chapter 4. The development should begin by assessing the compatibility of potential structural substrate materials such as aluminum or titanium to the thin film battery manufacturing process. Recent advances in high

temperature carbon fiber reinforced structural polymer composites should also be assessed [1, 2]. The sealant materials should then be selected according to the performance plot provided in the previous chapter and their compatibility to the battery manufacturing process should be also experimentally assesses. Thereafter electromechanical tests should be performed to confirm the increase in the strain to failure of the multifunctional laminates.

From the analysis standpoint, the crack length control scheme should be implemented into a multi-scale finite element model to study the electrochemical-mechanical interaction at the macro-scale (i.e. at the laminate scale). The multi-scale approach should capture the charge/discharge Li-ions kinetics and electrochemical-mechanical phenomena within a particle of active material, as in recently developed battery models [3], as well as the chemical reactions and swelling that occur during thermal runaway. Homogenization techniques can relate the micro-scale particle model to the macro-scale model describing the thickness variation caused by Li-ion insertion/removal and associated electric potentials, temperature and volume increase due to thermal runaway, laminate elastic and failure response.

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