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Establishment of the Cohesive Law of an Epoxy Adhesive Using the Direct Method and the Effect of the Substrate Material

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Abstract
The direct method was applied to characterize the fracture behavior of a structural adhesive obtaining the cohesive law for Mode I. Additionally, the effect of the substrate material in the results was analyzed. The Double Cantilever Beam (DCB) tests were applied to achieve the fracture toughness in Mode I and the J-integral approach was used to measure the critical energy release rate, J_{Ic}. The results were then compared and validated using the Compliance Based Beam Method (CBBM). By differentiation of the J-integral values with respect to the crack tip opening displacement (CTOD), the cohesive law was obtained. Moreover, a numerical validation was performed, by comparing the load-displacement experimental results with the numerical data obtained with the Abaqus software.
Furthermore, Digital Image Correlation (DIC) analysis was applied resorting to a Python script aiming to optimize the process.
The obtained results indicate that the material substrate does not influence the value of the fracture toughness but could positively influence the precision of the obtained cohesive law. Moreover, it was observed that the direct method could predict the fracture behavior of the epoxy adhesive.

1. Introduction

Adhesive bonding has been extensively used in the aeronautical and automotive industries (Sekiguchi, Hayashi, and Sato 2020; Ciardiello et al. 2020). When compared to other joining processes of materials, it has proven to be a cost-effective and light joining process, with the ability to bond different materials, reducing the overall emissions whilst providing an advantageous strength to weight ratio(Costa et al. 2018).

There are numerous joint configurations that have been discussed in the literature, but the single-lap, double-lap, and scarf joints are the most frequent. Therefore, the availability of simple and reliable predictive techniques is essential for the secure design of bonded joints(Carvalho and Campilho 2016). There are a variety of predictive methods for bonded joints that range from analytical to numerical, using various criteria to predict the beginning of material degradation, damage, or even total failure (Constante, Campilho, and Moura 2015). The Finite Element Method (FEM), which calculates stress and displacement fields and combines them with appropriate failure criteria, is the technique that is most widely used to predict the numerical strength of these joints (Campilho et al. 2013). Cohesive Zone Models (CZMs) have been widely used to analyze the fracture of adhesive joints and composite laminates by applying traction-separation laws (TSLs) to describe the mechanical properties across the failure surfaces(Fengzhen Sun, Zhang, and Blackman 2021). Cohesive elements can be used to implement the method between adherends in 2-D or 3-D problems. This method's wide acceptance is largely due to its functionality on the most popular commercial Finite Element (FE) platforms and its ability to work with complex geometries (Tserpes et al. 2021). The prediction of adhesive joint strength is highly dependent on the CZM parameters, so even though CZM produces results that are extremely accurate, a thorough determination of the cohesive law's parameters is required (Lélias et al. 2019). The cohesive laws for Mode I relate the tensile tractions (t_n) to the tensile relative displacements (δ_n) between homologous nodes of the cohesive element. The main cohesive laws parameters that will be used in numerical models are t_n^0 (cohesive strength in tension, which provides the peak traction value) and the critical value of tensile strain energy release rate (J_{Ic}) (Carvalho and Campilho 2017). Currently, a range of methods, including the property identification technique, the direct method, and the inverse method, are available for the definition of the cohesive parameters. The first consists of performing various tests that individually can provide the different cohesive law parameters needed. On the other hand, in the inverse method such parameters are estimated through iteration, combining the Finite Element Analysis (FEA) along with experimental data, until an accurate representation is obtained. Both methods start off by assuming a CZM shape to simulate a particular material, which closely resembles it in terms of post-elastic behavior. The last, the direct method, shows to be advantageous to determine the fracture properties of adhesives, being able to determine the precise shape of the cohesive law, which is directly obtained from the experimental data of fracture tests, like Double Cantilever Beam (DCB) for Mode I (Carvalho and Campilho 2016).

The DCB tests are usually used to obtain the fracture toughness, J_{Ic}. Different classical data reduction methods based on Linear Elastic Fracture Mechanics (LEFM) are used to measure this property, such as the Compliance Calibration Method (CCM), the Direct Beam Theory (DBT) and the Corrected Beam Theory (CBT) (de Moura, Campilho, and Gonçalves 2008; F. Sun and Blackman 2020). The previous approaches can only be applied if the fracture process zone (FPZ) is small, when compared to the length of the component. The Compliance Based Beam Method (CBBM) is an alternative that differs from other approaches because it does not require the measurement of the crack length during propagation, which is difficult to perform accurately. This method uses the concept of a crack equivalent length having the advantage of considering the effect of Fracture Process Zone (FPZ). The critical energy release rate is given by:

$$G_{Ic} = \frac{6P^2}{b^2 h}\left(\frac{2a_{eq}^2}{h^2 E_f} + \frac{1}{5G_f}\right) \quad (1)$$

where a_{eq}^2 is the equivalent crack length, E_f is the corrected flexural modulus and G_f is the shear modulus of the substrate(de Moura, Campilho, and Gonçalves 2008).

Until now, there are several proposed methods to characterize non-linear FPZ, based on the J-integral approach. This approach, which is not based in LEFM, can be calculated considering a Mode I loading, along the exterior boundary of the DCB specimen by:

$$J_1 = \frac{P}{b} \cdot (\theta_{up} - \theta_{low}) \quad (2)$$

where P is the applied load, b is the width of the specimen and θ_{up} and θ_{low} are the rotations of the upper and lower substrates, respectively.

By differentiating the J-integral with respect to the normal opening displacement of the crack tip, δ_n, the cohesive law is given by (Rice 1968):

$$\frac{\partial J_1}{\partial \delta_n} = \sigma(\delta_n) \quad (3)$$

2. Materials and experimental procedures

The epoxy-based adhesive AV138-M1was used in the performed tests. This adhesive is a two component, high strength adhesive that, when fully cured, has as excellent performance at high temperatures as well as a high chemical resistance.

The first performed DCB tests using steel substrates proved to have displacement values of very low magnitude and, thus, were hard to accurately measure. The approach to overcome this issue was to modify the material used for the substrates that was changed from steel to aluminum. In the present work, two steel blocks were bonded to the substrates in order to improve Digital Image Correlation (DIC) analysis, as shown in Figure 1.

The substrates were grit blasted and degreased with acetone immediately before the application of the adhesive. The DCB specimen geometry is present in Figure 1. There is an initial region without adhesive, which is the pre crack length. The initial pre-crack was created in the adhesive by inserting a sharp razor blade with 0.1 mm of thickness at exactly the midpoint of the adhesive layer. This layer had a thickness of 0.2 mm, constant along the length of the specimen, which was guaranteed using calibrated tape. The final procedure consisted of coating the specimens with white matte paint and speckling them with black dots to perform the DIC analysis.

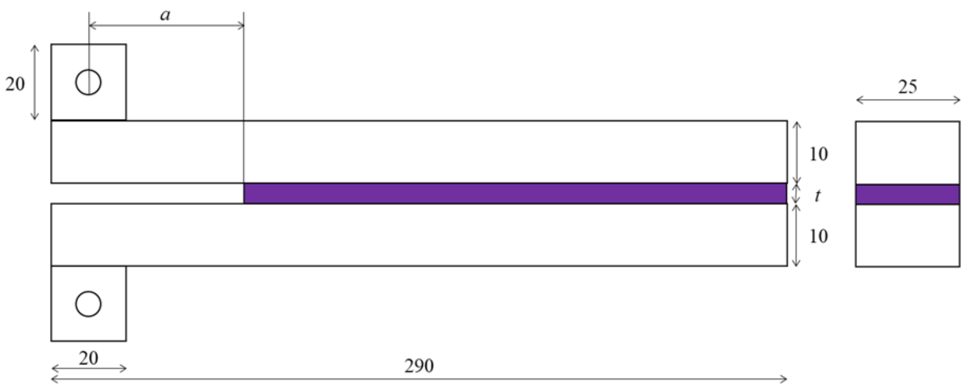

Figure 1: DCB specimen geometry, in mm.

For the DCB tests the universal testing machine INSTRON® 3367, equipped with a load cell of 30 kN was used. All the tests were performed at a speed of 0.2 mm/min. In the universal testing machine, the DCB specimens were mounted by connecting the blocks to the machine attachment elements with stiff steel pins. During the test it was necessary to ensure that the monitoring of the applied load was synchronized with the displacement field of the important areas. This was achieved by ensuring that the data acquisition of the universal testing machine was synchronized with the images acquired by a Nikon D5300, where a Nikon AF-P NIKKOR 18-55 mm F/3.5-5.6 lens was attached, required for the DIC analysis. This task is mandatory whilst applying the direct method so that the Mode I fracture toughness and later the traction-separation law could be determined. The displacements were registered and submitted to posterior DIC analysis, and the load-displacement curve was registered by the test machine. A small pre-load was applied to the DCB specimens to ensure a stable crack propagation.

The 2D analysis using DIC requires that the lens used to capture the images is perpendicular to the substrate. To ensure this perpendicularity between the DCB specimens and the camera, a laser method was used. A 3D printed part was placed parallel to an axis along the camera lens, and a laser was pointed onto the face of this object. When the laser beam appeared perfectly defined, final position was obtained. **Figure 2** depicts this method and presents the correct alignment of the beam.

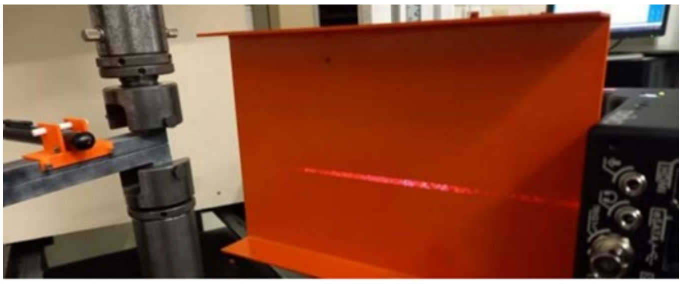

Figure 2: Laser beam in alignment with the camera lens.

The GOM Correlate software was used to process the images registered during the test. Both the substrate displacements and the crack tip opening were processed, using a facet size of 12 pixels and a pixel in the image corresponded to 0.101 mm on the measured surface.

Figure 3 the method for measuring the loading point rotations. The extraction of the $x$and y displacements of the upper and lower inspection points was used to estimate the value of the rotation of each substrate during the test by computing $\theta = \frac{dy}{dx}$, assuming a linear relationship between the x and y displacements(F. Sun and Blackman 2020).

The upper and lower inspection points at the crack tip were selected in the GOM software to measure the crack tip opening displacement (CTOD), by computing the difference between the two. To ensure that the measurement is performed perpendicularly to the adhesive layer, the same orientation was used in the software and, thus, the CTOD, δ_n, was determined .

Figure 3: Marked points in the DIC software to calculate the loading point rotations (left) and to determine the CTOD (right).

The CTOD and the J values obtained with the methods previously detailed were processed to obtain the cohesive law. The presence of experimental noise in the raw data required the adjustments of some parameters using a Savitsky-Golay filter. By adjusting the window length for the number of points considered and the order of the polynomial used in each filtering the results were smoothed. Finally, the cohesive law was estimated by:

$$\sigma = \frac{J_{t+\Delta t} - J_t}{\delta_{n_{t+\Delta t}} - \delta_{n_t}} \quad (4)$$

where t is the time in seconds and Δt is the increment between two measurements.

The experimental tests along with DIC analysis originated thousands of data and high values of noise associated with the results. To optimize and automate the analysis of this data, it was necessary to resort to a Python Script than can be used for data processing.

3. Results and discussion

The initial fracture tests of the present work were performed using steel substrates in order to characterize the fracture behavior of this joint by applying the direct method. However, soon after the first tests it was possible to conclude that, due to the low magnitude of the displacements essentially at the crack tip opening, it would be difficult to avoid the high noise values introduced in the experimental data. Since the cohesive law is obtained by differentiating the J values with respect to the CTOD, even the smallest variation of these last values would induce a large dispersion in the cohesive law values.

Considering this setback, the hypothesis of changing the joint substrate was considered, to obtain larger displacements during the tests. Firstly, an FEA was performed using a simulation model of the DCB tests, using steel and aluminum as materials for the substrates. The aim was to evaluate the CTOD for both cases, and, as can be observed in Figure 4, although there is a larger displacement for aluminum for most of the curve, when analyzing the beginning of them in more detail, it can be seen that the initial displacement of the crack tip is smaller for this material.

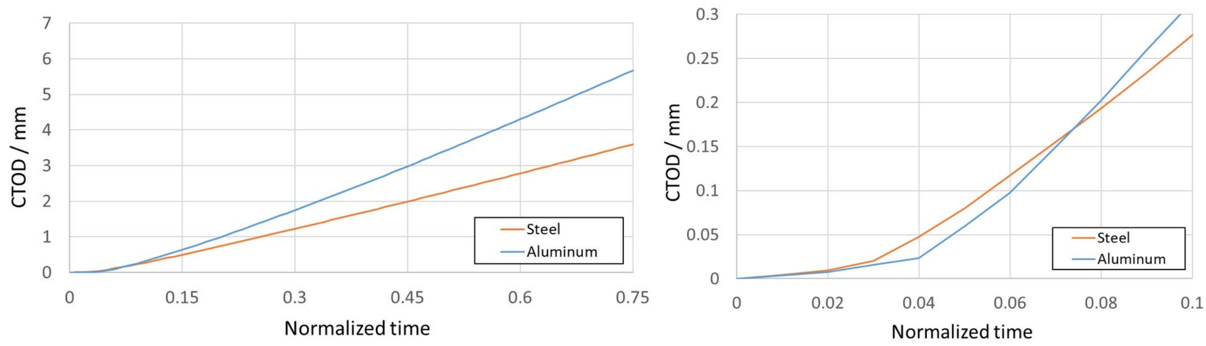

Figure 4: Comparison between the CTOD in steel and aluminum.

However, despite this preliminary result, it was considered possible that by having a larger generalized displacement for the aluminum substrates, their rotations at the loading point would be higher, and, in this way, the error introduced by the variations of the CTOD values would be mitigated during the cohesive law calculation. Thus, some tests were performed with both materials and, both their rotations and J values were subsequently analyzed. As can be observed in Figure 5 (left), the rotations values for aluminum are higher than the steel ones during the whole test, a result that reinforced the hypothesis that the change of the material could be beneficial to achieve the intended results. For this decision to be definitive, it would have to be undisputable that the fracture toughness values remained unchanged for both cases. Figure 5 (right) shows that the fact that the substrates are made of different materials, changes the recorded load values, as well as the rotations, but the result remains approximately the same. Steel substrates achieved a value of $0.21\ \pm 0.014\ N/mm$, while aluminium substrates achieved $0.22\ \pm 0.006\ N/mm$, which represents a variability of about 1.2%.

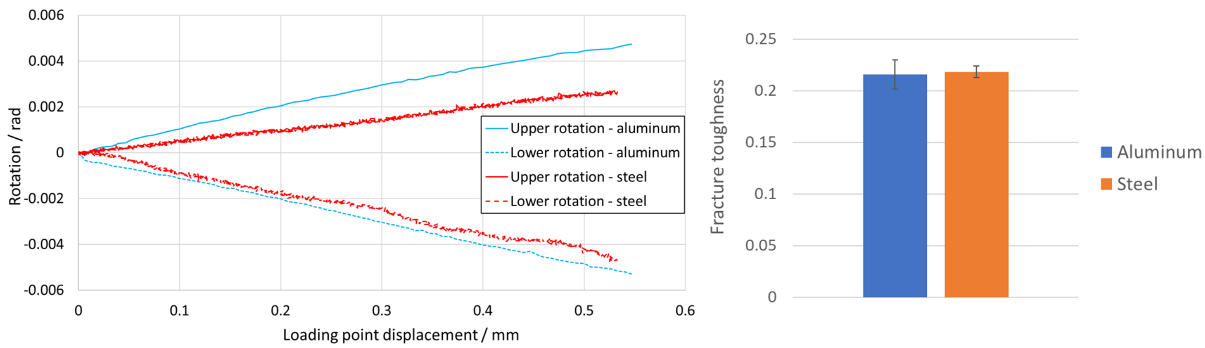

Figure 5: Comparison between the rotations measured in the experimental tests (left) for both materials and the obtained fracture toughness (right).

After establishing that the adhesive was to be characterized with DCB specimens with aluminum substrates, the necessary tests were carried out to obtain the cohesive law. **Figure 6** (left) depicts a representative load-displacement curve, recorded with the universal test machine. The adhesive exhibited cohesive failure, as well as a stable crack propagation. The average failure load observed was $1481 \pm 15.5\ N$.

For all the specimens, the crack propagation occurred in the middle-plan of the adhesive thickness. After the tests were performed, the J-integral approach in Mode I was used to calculate the fracture toughness of the adhesive. Subsequently, the results were compared to the CBBM values, as shown in **Figure 6** (right), where a good agreement between both methods can be observed. Both the J and G values showed similar behavior, having a steady increase until they reached a plateau of $0.21 \pm 0.014\ N/mm$ for the J-integral and $0.23 \pm 0.030\ N/mm$ for the CBBM method. This difference represents a variability of about 6.6%.

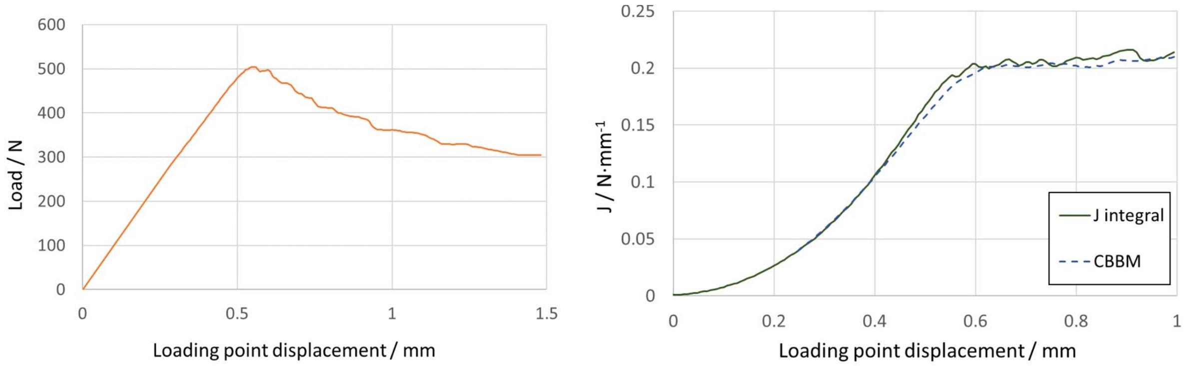

Figure 6: Load-displacement representative curve (left) and fracture toughness results for J-integral and CBBM (right).

The estimation of the cohesive law still generated data with considerable values of noise and, therefore, by implementing a Savitzky-Golay filter, the obtained curves were smoothened, as it can be observed in the representative curve in **Figure 7**. This process proved to be very sensitive to any change in the filter parameters and was performed carefully to prevent the loss of important information. The maximum value for the stress was $54.8 \pm 1.2\ MPa$, and it occurred at the end of the elastic behavior of the joint. Beginning at this point, where there was damage initiation, and its values started to increase, the stress values decreased, until they reached a zero value. In the representative curve, as well as for all the obtained curves, the stress values do not attain the zero value, considering that, from that CTOD value on, the area beneath the curve was negligible.

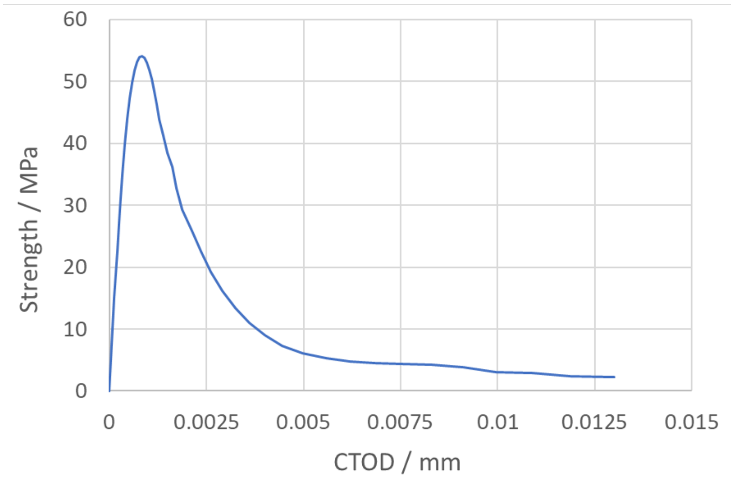

Figure 7: Cohesive law obtained by the direct method.

The average cohesive law obtained from the tests was used to validate the direct method, by using the average values for the damage of the adhesive and compute them in FEA, using CZM modelling with ABAQUS software. The numerical data was generated resorting to a DCB model that simulated the fracture test. The substrates were modeled with the elastic elements CPS4R and the adhesive was modeled with the cohesive elements COH2D4. **Figure 8** depicts the model used to perform the FEA, including geometry, boundary conditions and the respective mesh.

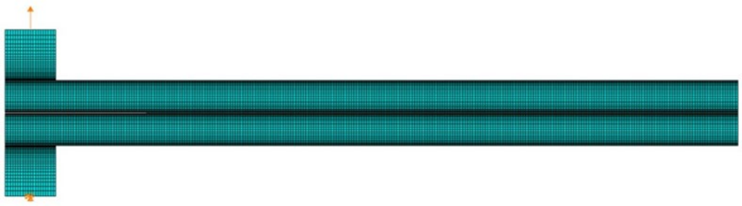

Figure 8: DCB model used in the numerical simulation.

The numerical validation resulted in the comparison of the CZM model values with the average load-displacement curve that represented the adhesive. The **Figure 9** represents both numerical and experimental representative curves. The value of the failure load for the numerical simulation was 557.9 N which is about 10.6% higher than the experimental results. The model could predict the stiffness of the adhesive but, by failing the maximum load, underestimated the damage values during crack propagation. Although the values of these parameter were not perfectly determined, the behavior of the fracture process seemed to be in a good agreement with the real joint, since the simulated damaged part of the curve is parallel to the experimental one.

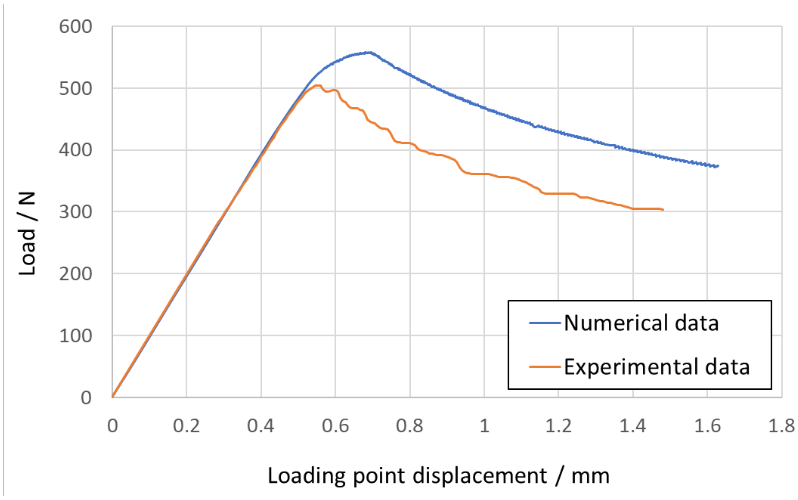

Figure 9: Comparison between the experimental load-displacement curve the curve obtained numerically.

4. Conclusions

In the present work the direct method was applied to characterize the fracture behavior in Mode I of an epoxy structural adhesive. Two substrate materials were compared in order to achieve the results with less noise, and the data was obtained resorting to DCB fracture tests and analyzed with the DIC technique, in conjunction with a Python script.

Results from the preliminary tests indicated that the aluminum substrates could introduce less noise in the data than the steel ones, due to its higher values of displacement. Although the CTOD was lower for aluminum in the beginning of the tests, the overall displacement created more rotation in the loading points, providing a less sensitive cohesive law, regarding the CTOD fluctuations. Additionally, the fracture toughness obtained for both materials was almost identical.

The fracture toughness values achieved with DCB tests using the J-integral approach were compared with the CBBM and a very good agreement was observed. A small difference between the plateau reached in the two methods was obtained and was approximately 6.6%.

The differentiation of the J values with respect to the CTOD resulted in a cohesive law that was smoothened by using a Savitzky-Golay filter, due to the inevitable noise produced by experimental data. A comparison between the experimental load-displacement curve and the one produced by numerical simulation, using the obtained TSL by the direct method, showed a good prediction of the stiffness of the adhesive. However, due to the underestimation of the failure load, the failure process of the curve created by CZM, did not present a perfect agreement. Nevertheless, the fracture behavior of the joint was similar to the experimental data and the overall features were acceptable.

References

Campilho, R. D. S. G., D. C. Moura, D. J. S. Gonçalves, J. F. M. G. da Silva, M. D. Banea, and L. F. M. da Silva (2013). "Fracture toughness determination of adhesive and co-cured joints in natural fibre composites." *Composites Part B: Engineering* 50: 120-126. DOI: 10.1016/j.compositesb.2013.01.025.

Carvalho, U. T. F., and R. D. S. G. Campilho (2016). "Application of the direct method for cohesive law estimation applied to the strength prediction of double-lap joints." *Theoretical and Applied Fracture Mechanics* 85: 140-148. DOI: 10.1016/j.tafmec.2016.08.018.

--- (2017). "Validation of pure tensile and shear cohesive laws obtained by the direct method with single-lap joints." *International Journal of Adhesion and Adhesives* 77: 41-50. DOI: 10.1016/j.ijadhadh.2017.04.002.

Ciardiello, R., L. Greco, M. Miranda, F. Di Sciullo, and L. Goglio (2020). "Experimental investigation on adhesively bonded U-shaped metallic joints using the Arcan test." *Journal of Advanced Joining Processes* 1: 100010. DOI: 10.1016/j.jajp.2020.100010.

Constante, C. J., R. D. S. G. Campilho, and D. C. Moura (2015). "Tensile fracture characterization of adhesive joints by standard and optical techniques." *Engineering Fracture Mechanics* 136: 292-304. DOI: 10.1016/j.engfracmech.2015.02.010.

Costa, M., G. Viana, R. Créac'hcadec, L. F. M. da Silva, and R. D. S. G. Campilho (2018). "A cohesive zone element for mode I modelling of adhesives degraded by humidity and fatigue." *International Journal of Fatigue* 112: 173-182. DOI: 10.1016/j.ijfatigue.2018.03.014.

de Moura, M. F. S. F., R. D. S. G. Campilho, and J. P. M. Gonçalves (2008). "Crack equivalent concept applied to the fracture characterization of bonded joints under pure mode I loading." *Composites Science and Technology* 68 (10): 2224-2230. DOI: 10.1016/j.compscitech.2008.04.003.

Lélias, G., E. Paroissien, F. Lachaud, and J. Morlier (2019). "Experimental characterization of cohesive zone models for thin adhesive layers loaded in mode I, mode II, and mixed-mode I/II by the use of a direct method." *International Journal of Solids and Structures* 158: 90-115. DOI: 10.1016/j.ijsolstr.2018.09.005.

Rice, James (1968). "A Path Integral and the Approximate Analysis of Strain Concentration by Notches and Cracks." *Journal of Applied Mechanics* 35: 379-386. DOI: 10.1115/1.3601206.

Sekiguchi, Yu, Asuka Hayashi, and Chiaki Sato (2020). "Analytical determination of adhesive layer deformation for adhesively bonded double cantilever beam test considering elastic–plastic deformation." *The Journal of Adhesion* 96 (7): 647-664. DOI: 10.1080/00218464.2018.1489799.

Sun, F., and B. R. K. Blackman (2020). "A DIC method to determine the Mode I energy release rate G, the J-integral and the traction-separation law simultaneously for adhesive joints." *Engineering Fracture Mechanics* 234: 107097. DOI: 10.1016/j.engfracmech.2020.107097.

Sun, Fengzhen, Ruoyu Zhang, and Bamber R. K. Blackman (2021). "Determination of the mode I crack tip opening rate and the rate dependent cohesive properties for structural adhesive joints using digital image correlation." *International Journal of Solids and Structures* 217-218: 60-73. DOI: 10.1016/j.ijsolstr.2021.01.034.

Tserpes, Konstantinos, Alberto Barroso-Caro, Paolo Andrea Carraro, Vinicius Carrillo Beber, Ioannis Floros, Wojciech Gamon, Marcin Kozłowski, Fabio Santandrea, Moslem Shahverdi, Davor Skejić, Chiara Bedon, and Vlatka Rajčić (2021). "A review on failure theories and simulation models for adhesive joints." *The Journal of Adhesion*: 1-61. DOI: 10.1080/00218464.2021.1941903.