

THESIS FOR THE DEGREE OF DOCTOR OF PHILOSOPHY

High Temperature Adhesives for Aerospace Applications

by

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ABSTRACT

Adhesives used in structural high temperature space and aerospace applications must operate in extreme environments. They need to exhibit high-temperature capabilities in order to maintain their mechanical properties and their structural integrity at the intended service temperature. One of the main problems caused by high temperature conditions is the fact that the adhesives have different mechanical properties with temperature, such as the stress-strain curve and the toughness. Information on the toughness of the adhesives at low or high temperatures is more difficult to find in the literature than the stress-strain curves of the adhesives and this is an area that needs to be investigated.

One of the applications of adhesive bonding in the aerospace industry is in the bonding of the tiles for temperature protection in the space shuttle. The adhesive used to bond the ceramic tiles is a RTV (room temperature vulcanizing silicone rubber). The strength of that adhesive is just enough to hold the tiles in place at the extreme temperature reached in the reentry of the space shuttle into the atmosphere due to the air friction. The RTV can give a bond that accommodates the different mechanical and physical behaviour of the tile and the metal, but has a low strength at high temperatures. An adhesive with a good strength at high temperatures would be too brittle at low temperatures when the shuttle is in orbit. One way to solve this problem is by bonding the tiles using two adhesives. A high temperature adhesive with a higher strength than the silicone adhesive would guarantee a better strength at high temperatures during the space shuttle reentry into the atmosphere and the silicone adhesive would be the load carrying adhesive when the space shuttle is in orbit.

The aim of this research is to investigate the mechanical behaviour of two different types of high-temperature adhesives, one for strength at high temperatures and one for strength at low temperatures, which will be subsequently used to design a mixed-adhesive joint suitable for use from low to high temperatures. An epoxy adhesive with a high strength at high temperatures but most likely very brittle at low temperatures would be combined with a silicone adhesive which is tough at low temperatures and not degrade at the high temperatures even being very ductile and creep.

However, the use of a combination of these adhesives is not a trivial task and requires careful design. A good prediction of the failure load of the mixed-adhesive joint proposed requires the adhesives mechanical properties. Furthermore, to predict the joint strength, the stress distribution and a suitable failure criterion are necessary. One of the simplest failure criteria is that based on a stress or strain limit state, i.e. based on a continuum mechanics approach for which the availability of the stress–strain curve of the high temperature adhesives is sufficient. However, for the more realistic and sophisticated methods such as progressive damage methods, damage laws of adhesives to be used under different temperatures are necessary. The parameters that define the damage law are the fracture toughness (G_c) and the maximum stress (σ_c^0) in each fracture mode.

Failure strength tests were carried out for determining the basic properties of the high temperature adhesives studied. Tensile bulk tests were performed to get the tensile properties of the adhesives as a function of temperature (the Young's modulus, the ultimate tensile stress and the failure tensile strain). The standard Thick Adherend Shear Test (TAST) was performed in order to measure the shear properties of the high temperature adhesives. In addition, single lap joints (SLJs) were fabricated and tested to assess the adhesives performance in a joint. The influence of temperature on the lap shear strength of the adhesives was investigated.

Numerical models of adhesively bonded joints using cohesive zone models were developed. The cohesive laws for use in the material models of the high temperature adhesives have been determined using dedicated test methods. The double cantilever beam specimen and the end notched flexure specimen were used with direct and inverse methods to determine cohesive laws in peel and shear, respectively. The cohesive laws were determined as a function of temperature in order to develop a suitable methodology for design-level predictions of the effect of temperature on the performance of structural adhesive joints for high temperature applications.

Keywords: High temperature adhesives; temperature tests; lap-shear strength; tensile strength; pure mode I and mode II fracture toughness, traction–separation laws, cohesive zone models.

RESUMO

Os adesivos utilizados em aplicações estruturais aeroespaciais a alta temperatura devem funcionar em ambientes extremos. Necessitam de características de resistência a altas temperaturas que lhes permitam manter as suas propriedades mecânicas e a sua integridade estrutural intactas à temperatura de serviço. Um dos maiores problemas resultantes das condições a temperaturas elevadas é a diferença registada nas propriedades mecânicas com o aumento da temperatura, tais como a curva de tensão-deformação e a rigidez. É mais difícil encontrar informação acerca da tenacidade dos adesivos a baixa e alta temperatura quando comparada com às curvas de tensão-deformação, sendo portanto uma área que necessita de maior investigação.

Uma das aplicações das ligações adesivas na indústria aeroespacial é a da união das placas cerâmicas de protecção térmica à estrutura de alumínio do vaivém espacial. O adesivo para colar as placas cerâmicas é um RTV (borracha de silicone vulcanizada à temperatura ambiente). A resistência desse adesivo é a suficiente para manter as placas cerâmicas integralmente ligadas durante as elevadíssimas temperaturas que se registam durante a reentrada do vaivém na atmosfera, devidas à fricção com o ar. O adesivo RTV promove uma adesão que permite acomodar os diferentes comportamentos mecânicos e físicos do metal e do cerâmico, mas tem uma reduzida rigidez a altas temperaturas. Um adesivo com boa rigidez a altas temperaturas seria demasiado frágil para as baixas temperaturas que ocorrem quando o vaivém está em órbita. Uma solução para este problema é utilizar dois adesivos distintos para colar as placas cerâmicas. Um adesivo para altas temperaturas com rigidez superior ao adesivo de silicone garantiria uma melhor rigidez a altas temperaturas durante a reentrada do vaivém na atmosfera e o adesivo de silicone suportaria as cargas durante a órbita do vaivém.

O objectivo desta tese é investigar o comportamento mecânico de dois tipos diferentes de adesivos para altas temperaturas, um com rigidez para altas temperaturas e outro com rigidez para baixas temperaturas, que serão subsequentemente utilizados para projectar uma junta mista, com dois adesivos, apropriada para uma ampla gama desde baixas até altas temperaturas.

Um adesivo epóxico com elevada rigidez para altas temperaturas, mas muito provavelmente frágil a baixas temperaturas será combinado com um adesivo de silicone que é rígido a baixas temperaturas e que apesar de ser dúctil e se deformar, não se degrada a altas temperaturas.

Contudo, a combinação destes adesivos não é uma tarefa trivial e requer um projecto cuidado. Uma boa previsão da força de rotura da junta mista proposta requer o conhecimento das propriedades mecânicas dos adesivos. Um dos critérios de rotura mais simples é baseado num estado limite de tensão ou deformação, i.e. baseado numa abordagem de mecânica contínua para a qual a utilização da curva de tensão-deformação para os adesivos de alta temperatura é suficiente. No entanto, para métodos mais realistas e sofisticados como o método de dano progressivo é necessário conhecer as leis de dano dos adesivos para diferentes temperaturas. Os parâmetros que definem as leis de dano são a taxa de libertação de energia de fractura (G_c) e a tensão máxima (σ_c^0) em cada modo de fractura.

Ensaio mecânicos foram realizados para determinar as propriedades básicas dos adesivos de alta temperatura estudados. Recorrendo a provetes maciços de adesivo, foram realizados ensaios de tracção para obter as diferentes propriedades em função da temperatura (módulo de Young, tensão de cedência e deformação após rotura). O ensaio TAST (*Thick Adherend Shear Test*) foi realizado para medir as propriedades de corte dos adesivos para altas temperaturas. Foram ainda fabricados juntas de simples sobreposição (SLJs) e ensaiados para aferir o comportamento dos adesivos numa junta adesiva. A influência da temperatura nestas juntas de simples sobreposição foi também investigada.

Foram desenvolvidos modelos numéricos de juntas adesivas utilizando modelos com elementos coesivos. As leis coesivas utilizadas nos modelos para simular os adesivos para altas temperaturas foram obtidas através de testes apropriados. O provete de dupla viga (DCB, *Double Cantilever Beam*) e o ensaio de flexão em três pontos foram utilizados para obtenção das leis coesivas em tracção e em corte respectivamente, recorrendo a métodos directos e inversos. As leis coesivas foram determinadas em função da temperatura de forma a desenvolver uma metodologia adequada para previsões, a nível de projecto, do efeito da temperatura na resistência de juntas adesivas estruturais para aplicações a altas temperaturas.

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LIST OF PUBLICATIONS

1. Banea M.D., da Silva L.F.M., Campilho R.D.S.G., Mode II Fracture Toughness of Adhesively Bonded Joints as a Function of Temperature: Experimental and Numerical Study, 2011, Journal of Adhesion, to be submitted.
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SUMMARY OF THESIS

1. Introduction

1.1 Background

Adhesives used in structural high temperature space and aerospace applications must operate in extreme environments. They need to exhibit high-temperature capabilities in order to maintain their mechanical properties and their structural integrity at the intended service temperature (resist thermal breakdown at elevated temperature). Adhesive systems that meet some of these requirements include: epoxies (having high strength and temperature resistance), silicones (excellent sealant for low stress applications, high degree of flexibility and very high temperature resistance), phenolics, polyimides, bismaleimides and ceramic adhesives.

One of the applications of adhesive bonding in the aerospace industry is in the bonding of the tiles for temperature protection in the space shuttle. The last disaster where some tiles detached from the internal aluminium fuselage of the shuttle shows that the technique of adhesive bonding still needs significant research [1]. The adhesive used to bond the ceramic tiles is a RTV (room temperature vulcanizing silicone rubber). The strength of that adhesive is just enough to hold the tiles in place at the extreme temperature reached in the reentry of the space shuttle into the atmosphere due to the air friction. The RTV promotes a bond that accommodates the different mechanical and physical behaviour of the tile and the metal, but has a low strength at high temperatures. An adhesive with a good strength at high temperatures would be too brittle at low temperatures when the shuttle is in orbit. One way to solve this problem is by bonding the tiles using two adhesives. A high temperature adhesive with a higher strength than the silicone adhesive would guarantee a better strength at high temperatures during the space shuttle reentry into the atmosphere and the silicone adhesive would be the load carrying adhesive when the space shuttle is in orbit.

The concept of a mixed-adhesive joint, which use more than one adhesive along the overlap (an adhesive bondline with variable modulus to relieve the high-stress concentrations at the end regions of the overlap) in order to have a more uniform stress distribution was studied in the past [2-7]. A flexible and ductile adhesive is placed at the extremities of the overlap and a

rigid and brittle adhesive is placed in the middle of the overlap (see Fig. 1). da Silva and Adams [5,6] investigated theoretically and experimentally mixed-adhesive titanium/composite joints to be used over a wide temperature range (-55 to 200°C) and showed that there is a real improvement in joint strength, especially if the difference of coefficients of thermal expansion is high. This concept could be extended to the bonding of ceramic tiles in the space shuttle. An epoxy adhesive with a higher strength than the silicone adhesive currently used, which will retain the strength and will transfer the entire load at high temperatures could be combined with the silicone adhesive which would be the load carrying adhesive at low temperatures.

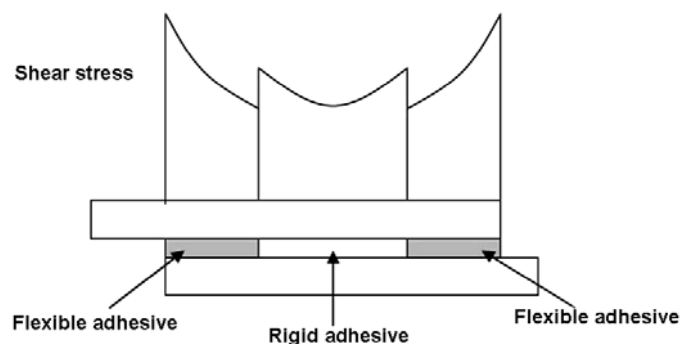


Fig. 1 Mixed - adhesive bonded joint.

The influence of temperature on the strength of adhesive joints is an important factor to consider in the design of adhesive joints. The most significant factors that determine the strength of an adhesive joint when used over a wide temperature range are: the cure shrinkage [8], the coefficients of thermal expansion (CTE), (especially when compared to the CTE of the substrates) [7], and different adhesive mechanical properties with temperature [6, 9-12]. However, due to the polymeric nature of adhesives, the most important factor to consider when designing a bonded joint is the variation of adhesive mechanical properties with temperature such as the stress-strain curve and the toughness.

Studies that present experimental results of adhesive joints with structural adhesives (especially epoxies) as a function of temperature generally show a decrease in strength with increasing and decreasing temperatures [13-19]. At high temperatures the cause is the lower adhesive strength while at low temperatures the high thermal stresses and the brittleness of the adhesive are the origin of such behaviour. However, little is known in terms of temperature when it comes to the fracture toughness of adhesively-bonded joints as the studies for

determination of the fracture toughness in tension or shear of thin adhesive layers in adhesively-bonded assemblies are often limited to room temperature testing.

In order to properly design a joint, the adhesive behaviour has to be characterized. Hence, to determine the stresses and strains in adhesive joints in a variety of configurations, it is necessary to know the adhesive mechanical properties. Furthermore, to predict the joint strength, the stress distribution and a suitable failure criterion are essential. One of the simplest failure criteria is that based on a stress or strain limit state, i.e. based on a continuum mechanics approach for which the availability of the stress–strain curve of the adhesive is sufficient. However, for the more realistic and sophisticated methods such as progressive damage methods, damage laws of adhesives to be used under different temperatures are necessary. A cohesive zone model (CZM), used to model the progressive damage and failure, models the fracture process extending the concept of continuum mechanics by including a zone of discontinuity modelled by cohesive zones, thus using both strength and energy parameters to characterize the debonding process. The parameters that define the damage law are the fracture toughness (G_c) and the maximum stress (σ_c^0) for each fracture mode.

This is still an innovative field under intense development, and it is also being implemented in commercial FE software packages such as ABAQUS® [20]. On the other hand, damage laws as a function of temperature are difficult to find in the literature and this was one of the challenges of this PhD research.

1.2 Objectives

The main objective of this research was to investigate the mechanical behaviour of two different types of adhesives, one for strength at high temperatures and one for strength at low temperatures, which will be subsequently used to design a mixed-adhesive joint suitable for use from low to high temperatures, for bonding the ceramic tiles to the space shuttle body.

The specific objectives are listed below:

- to determine the intrinsic mechanical properties of two different types of adhesives (a RTV silicone adhesive and an epoxy adhesive).
- to determine the appropriate cohesive laws for tension (pure mode I) loading as a function of temperature for the high temperature adhesives investigated.

- to determine the appropriate cohesive laws for shear (pure mode II) loading as a function of temperature for the high temperature adhesives.
- to perform end-use tests (SLJs) with the high temperature adhesives investigated in order to validate experimentally the damage laws obtained and to develop a suitable methodology of fracture characterization of adhesive joints for high temperature aerospace applications as a function of temperature.

1.3 Research methodology

In order to achieve the aim of this PhD research, the following approach was adopted:

- One of the main advantages of using adhesive bonding is the possibility to bond dissimilar materials, such as composite or ceramic to aluminium in many aerospace applications. Hence, a *literature review* of the investigations that have been made on adhesively bonded joints of composite structures was done in **Paper 1**.
- *Failure strength tests* were carried out for determining the basic adhesive properties in **Papers 2** and **3**. Tensile bulk tests were performed to get the tensile properties of the adhesives as a function of temperature, while the standard Thick Adherend Shear Test (TAST) was performed in order to measure the shear properties of the high temperature adhesives.
- *Fracture tests* for adhesive joints bonded with the high temperature RTV silicone adhesive were performed in **Paper 4** and pure mode I traction–separation laws were obtained as a function of temperature. The cohesive laws were obtained directly from the experiments by differentiation of simultaneously measured data (the J -integral and the end-opening displacement).
- *Fracture tests* for adhesive joints bonded with the epoxy high temperature adhesive were performed in order to get the fracture toughness in pure mode I (**Paper 5**) and pure mode II (**Paper 7**).
- *Numerical models* using cohesive zone models, in which the failure behaviour is expressed by a bilinear traction–separation law, were developed. From the numerical simulations, the parameters that describe the bilinear cohesive zone models were determined as a function of temperature in mode I (**Paper 6**) and mode II (**Paper 7**). The effect of the cohesive parameters on the output of numerical simulations of DCB and ENF tests was studied in **Paper 6** and **Paper 7**, respectively.

- In order *to validate the cohesive laws*, tensile tests on joints as a function of temperature were performed and numerical predictions based on the use of the cohesive damage model were obtained for the high temperature epoxy adhesive in **Paper 8**.

1.4 Outline of the thesis

This thesis consists of eight appended journal papers and a summary.

Paper 1 Banea M.D., da Silva L.F.M., Adhesively Bonded Joints in Composite Materials – An Overview, Proc. IMechE, Part L: Journal of Materials: Design and Applications, 223(1), 2009, p. 1-18.

Abstract of Paper 1: *Adhesively Bonded Joints in Composite Materials – An Overview.* A review of the investigations that have been made on adhesively bonded joints of fiber-reinforced plastic (FRP) composite structures (single skin and sandwich construction) is presented. The effects of surface preparation, joint configuration, adhesive properties and environmental factors on the joint behaviour are described briefly for FRP composite structures adhesively bonded. The analytical and numerical methods of stress analysis required before failure prediction are discussed. The numerical approaches cover both linear and non-linear models. Several methods that have been used to predict failure in bonded joints are described. There is no general agreement about the method that should be used to predict failure as the failure strength and mode are different according to various bonding methods and parameters, but progressive damage models are quite promising since important aspects of the joint behaviour can be modelled using this approach. However, a lack of reliable failure criteria still exists, limiting in this way a more widespread application of adhesively bonded joints in principal load bearing structural applications. An accurate strength prediction of the adhesively bonded joints is essential to decrease the amount of expensive testing at the design stage.

Paper 2 Banea M.D., da Silva L.F.M., Static and Fatigue Behaviour of Room Temperature Vulcanizing Silicone Adhesives for High Temperature Aerospace Applications, Materialwissenschaft und Werkstofftechnik 2010, 41(5), p. 325-335.

Abstract of Paper 2: *Static and Fatigue Behaviour of Room Temperature Vulcanizing Silicone Adhesives for High Temperature Aerospace Applications.* This paper focuses on the mechanical properties of room temperature vulcanising (RTV) silicone adhesives for aerospace applications. In view of the very limited amount of published data on this subject, we analyzed the performances of two RTV silicone rubber adhesives through adhesive joint tests. The standard Thick Adherend Shear Test (TAST) was performed in order to measure the shear properties of the adhesives. Single lap joints (SLJs) were fabricated and tested to assess the adhesive performance in a joint. The influence of temperature on the lap-shear strength of the adhesives was investigated. It is shown that the lap-shear strength of both adhesives decreased with increasing of temperature. The effect of bondline thickness and overlap length on the lap-shear strength of the adhesives was also studied. The reduction of failure load with an increase of the bondline thickness is a very common situation when dealing with structural adhesives. However, for the tested RTV silicone adhesives the failure loads increased as the bondline was made thicker. Also, the failure loads of RTV silicone adhesive joints increased almost proportionally with increasing the overlap length. Fatigue tests were also performed and show a low dispersion of the results.

Paper 3 Banea M.D., de Sousa F.S.M., da Silva L.F.M., Campilho R.D.S.G., de Bastos Pereira A.M., Effects of Temperature and Loading Rate on the Mechanical Properties of a High Temperature Epoxy Adhesive, *Journal of Adhesion Science and Technology*, 2011, 25, p. 2461-2474.

Abstract of Paper 3: *Effects of Temperature and Loading Rate on the Mechanical Properties of a High Temperature Epoxy Adhesive.* In this study, the combined effect of the temperature and test speed on the tensile properties of a high temperature epoxy adhesive was investigated. Tensile tests were performed at three different test speeds and various temperatures (room temperature (RT) and high temperatures (100°C, 125°C and 150°C)). The glass transition temperature (T_g) of the epoxy adhesive investigated is approximately 155°C. The ultimate tensile stress linearly decreased with temperature (T) while logarithmically increased with the loading rate, which is in the accordance with the Airing's molecular activation model.

Paper 4 Banea M.D., da Silva L.F.M., Campilho R.D.S.G, Temperature Dependence of the Fracture Toughness of Adhesively Bonded Joints, Journal of Adhesion Science and Technology, 2010, 24, p. 2011–2026.

Abstract of Paper 4: *Temperature Dependence of the Fracture Toughness of Adhesively Bonded Joints.* As is known, adhesive strength generally shows temperature dependence. Similarly, the fracture toughness is expected to show temperature dependence. In this study, the pure mode I fracture toughness for adhesive joints bonded with a high temperature RTV silicone adhesive was measured over a wide range of temperatures. Double cantilever beam (DCB) tests were performed on specimens at room temperature (RT), 100 and 200°C. Mode I traction–separation laws were obtained as a function of temperature, directly from the experiments, by differentiation of simultaneously measured data (the J -integral and the end-opening displacement). Results showed that the fracture toughness, the peak cohesive stress and the respective end-opening displacement all decreased with the temperature rise.

Paper 5 Banea M.D., da Silva L.F.M., Campilho R.D.S.G., Effect of Temperature on Tensile Strength and Mode I Fracture Toughness of a High Temperature Epoxy Adhesive, Journal of Adhesion Science and Technology, 2011,
DOI: 10.1163/156856111X593649.

Abstract of Paper 5: *Effect of Temperature on Tensile Strength and Mode I Fracture Toughness of a High Temperature Epoxy Adhesive.* In this work, an experimental study is performed to evaluate the effect of the temperature on the adhesive strength and fracture toughness of a high temperature epoxy adhesive. Bulk specimens of cured adhesive were produced and tested in tension at room temperature (RT), 100°C, 150°C and 200°C, in order to obtain a strength profile of the adhesive over this temperature range. Results showed that as the temperature increases the adhesive tensile strength reduces but the ductility increases. Pure mode I adhesive fracture toughness (G_{Ic}) tests were performed on double cantilever beam (DCB) specimens at room (RT) and high temperatures (100°C, 150°C and 200°C) and the fracture toughness G_{Ic} as a function of temperature was obtained. It is shown that at temperatures below T_g , the fracture toughness, G_{Ic} , is relatively insensitive to temperature, while above T_g (at 200°C) a drastic decrease in fracture toughness was observed.

Paper 6 Banea M.D., da Silva L.F.M., Campilho R.D.S.G., Mode I Fracture Toughness of Adhesively Bonded Joints as a Function of Temperature: Experimental and Numerical Study, International Journal of Adhesion and Adhesives, 2011, 31(5), p. 273-279.

Abstract of Paper 6: *Mode I Fracture Toughness of Adhesively Bonded Joints as a Function of Temperature: Experimental and Numerical Study.* In this work, the Double Cantilever Beam (DCB) test is analysed in order to evaluate the effect of the temperature on the adhesive mode I fracture toughness of a high temperature epoxy adhesive. Cohesive zone models, in which the failure behaviour is expressed by a bilinear traction–separation law, have been used to define the adhesive behaviour and to predict the adhesive P – δ curves as a function of temperature. The simulation response for various temperatures matched the experimental results very well. The sensitivity of the various cohesive zone parameters in predicting the overall mechanical response as a function of temperature was examined as well for a deeper understanding of this predictive method. Also, issues of mesh sensitivity were explored to ensure that the results obtained were mesh independent.

Paper 7 Banea M.D., da Silva L.F.M., Campilho R.D.S.G., Mode II Fracture Toughness of Adhesively Bonded Joints as a Function of Temperature: Experimental and Numerical Study, to be submitted to Journal of Adhesion.

Abstract of Paper 7: *Mode II Fracture Toughness of Adhesively Bonded Joints as a Function of Temperature: Experimental and Numerical Study.* In this work, an experimental and numerical study is performed to evaluate the effect of temperature on the mode II fracture toughness of a high temperature epoxy adhesive. Mode-II fracture testing on End Notch Flexure (ENF) test specimens was performed at room temperature (RT) and high temperatures (100°C, 150°C and 200°C) and the mode II fracture toughness G_{IIc} as a function of temperature was obtained. It is shown that at temperatures well below the glass transition temperature, the fracture toughness, G_{IIc} , increases with temperature and decreases as the temperature approaches T_g , while above T_g a drastic decrease in fracture toughness was observed. Furthermore, cohesive zone models, in which the failure behaviour is expressed by a bilinear traction–separation law, were used to define the adhesive behaviour in mode II and to predict the adhesive load-displacement (P – δ) curves as a function of temperature. The simulation response matched the experimental results very well at temperatures below T_g . The

sensitivity of the various cohesive zone parameters in predicting the overall mechanical response of the joint was examined as well for a deeper understanding of this predictive method.

Paper 8 Banea M.D., da Silva L.F.M., Campilho R.D.S.G., Effect of Temperature on the Shear Strength of Aluminium Single-lap Bonded Joints for High Temperature Applications, to be submitted to Journal of Adhesion Science and Technology.

Abstract of Paper 8: *Effect of Temperature on the Shear Strength of Aluminium Single-lap Bonded Joints for High Temperature Applications.* An experimental and numerical investigation into the shear strength behaviour of adhesive single-lap joints (SLJs) was carried out in order to understand the effect of temperature on the joint strength. The adherend material used for the experimental tests was an aluminium alloy in the form of thin sheets, and the adhesive used was a high strength high temperature epoxy. Tensile tests as a function of temperature were performed and numerical predictions based on the use of a bilinear cohesive damage model were obtained. It is shown that at temperatures below T_g , the lap-shear strength of SLJs increased, while at temperatures above T_g , a drastic drop in the lap-shear strength was observed. Comparisons between the experimental and numerical maximum loads representing the strength of the joints show a reasonably good agreement.

2. Adhesives tested

In this thesis, two different types of adhesives, one for strength at high temperatures and one for strength at low temperatures were investigated.

Initially (in **Paper 2**), two RTV adhesives were studied (for strength at low temperatures):

- AS1805, a high temperature thixotropic adhesive sealant, room temperature vulcanising (RTV) silicone rubber supplied by ACC Silicones LDT (Bridgwater, UK) and
- RTV106, a high temperature acetoxo adhesive sealant supplied by GE Bayer Silicones (USA).

After screening tests, as the adhesive RTV106 showed a better behaviour at high temperatures than adhesive AS1805, the adhesive RTV106 was chosen for the design of the mixed-adhesive joint. Therefore, fracture tests (Mode I) were performed only with RTV106 adhesive (**Paper 4**).

A rigid high temperature adhesive for strength at high-temperatures was chosen:

- XN1244, a one-component high temperature paste epoxy adhesive, supplied by Nagase ChemteX (Japan).

Failure strength tests were performed in **Paper 3**. Fracture tests were performed in **Papers 5, 6** (Mode I) and **Paper 7** (Mode II). Single Lap Joints were performed in **Paper 8**.

Structural adhesives, such as XN1244, have high glass transition temperatures (155°C obtained by DMTA method), high elastic modulus but low extensions to failure. Flexible adhesives, such as RTV106 silicone adhesive, have low glass transition temperatures (-60°C, data provided by supplier), low elastic modulus but high extensions to failure. However, combinations of these adhesives in a mixed-joint are possible and this would be particularly beneficial for bonded adherends with dissimilar coefficients of thermal expansion or with different stiffnesses [6], as in the case of bonding the ceramic tiles to the shuttle body. The flexible and ductile adhesive (RTV106) is placed at the ends of the overlap and the rigid and brittle adhesive (XN1244) is placed in the center of the overlap (see Fig. 2). At high temperatures, the XN1244 in the middle of the joint will retain the strength and will transfer the entire load while the RTV106 will be the load bearing component at low temperatures.

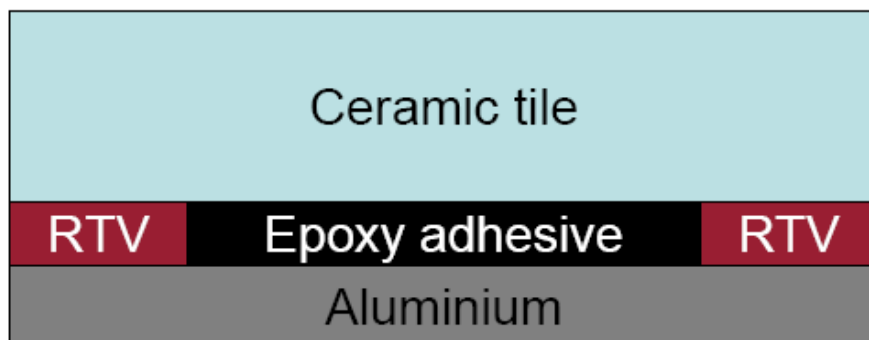


Fig. 2 Mixed - adhesive bonded joint proposed.

3. Test methods

Before an adhesive can be specified for an application, screening tests should be conducted in order to compare and evaluate the various adhesion parameters. This is especially true for structural adhesives where failures during actual use can have devastating consequences. Properties of adhesives can vary greatly and an appropriate selection is essential for a proper joint design. Thus, to determine the stresses and strains in adhesive joints in a variety of configurations, it is necessary to characterize the adhesive behaviour in order to know its mechanical properties, particularly the stress-strain curve and the modulus.

The approaches used for determining the properties of adhesives are the measure of the properties of bulk adhesive specimens and the use of specially designed joint geometries with a thin bond line (often referred to as “*in-situ*” testing).

A great variety of test geometries and specimens are used to obtain adhesive properties. The measured parameters are the load and strain needed to create failure. The test geometry should provide a pure state of stress, uniformly distributed across the contact surface and through the bondline, free of stress concentrations, and the surface treatment should be sufficient to ensure cohesive failure in the adhesive layer.

Currently there are many ASTM and ISO standards, which have been written to analyze and experimentally verify adhesive properties. These standards provide a basis for testing. Commonly test methods that have been developed and used to obtain properties of the adhesives include: tensile tests, shear tests, compression, peel, durability tests and dynamic tests.

3.1 Failure strength tests

In this thesis, the following tests were performed for the determination of the basic adhesive mechanical properties:

- Tensile bulk tests were performed to get the tensile properties of the adhesives.
- The thick-adherend shear test method (TAST) was performed in order to measure the shear properties of the adhesives.

3.1.1 Tensile tests

In the following section the manufacture of “dogbone” adhesive specimens, the test method and the results of the tensile tests on “dogbone” RTV silicone adhesives at RT are briefly presented.

Tensile tests of RTV silicone adhesives

Manufacture of the specimens and test method: The fully cured RTV adhesives are very soft and machining to the right dimensions was not feasible. Therefore, a moulding technique to avoid any problems associated with the cutting of the specimens and to produce the best possible specimens for tensile testing was used. The metallic mould designed for this purpose is shown in Fig. 3a. It consists of three individual parts: (1) the base part, which is used as the support, (2) the middle part, which determines the shape and the thickness of the specimen and (3) the top part, which closes the mould. To ensure that the adhesive does not bond to the mould, three layers of release agent were applied to all the parts, prior to adhesive application. The three parts were fitted together with screws. The RTV dogbone specimens were cured at RT (Fig. 3b). For a fast and complete curing process the specimens were removed from the mould after 10 days and left at 25°C and 50% relative humidity (RH) for a period of three more weeks. In this way, the diffusion of water from all sides into the bulk of the samples was allowed to promote complete curing of the specimen.

The geometry of the dogbone tensile specimens (2 mm thick and 10 mm wide) used correspond to BS 2782 standard and is shown in Fig 4.

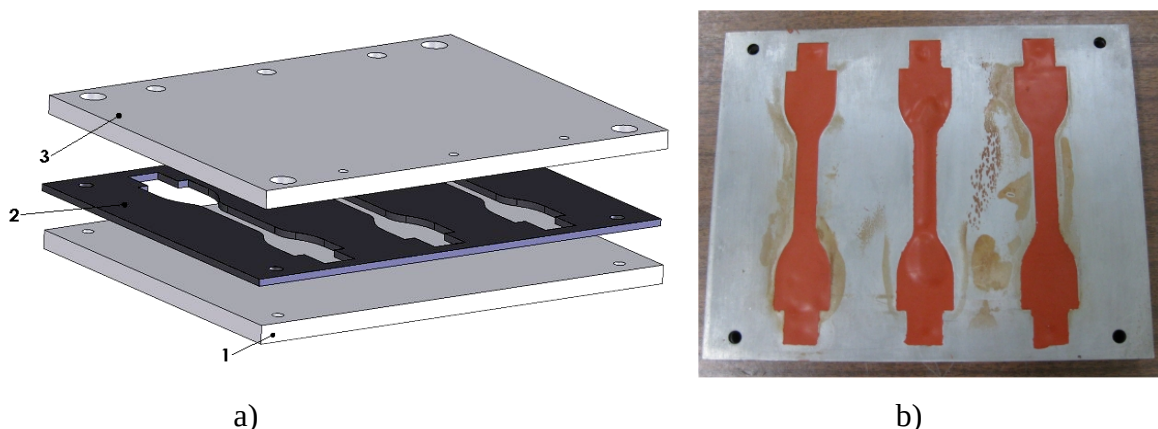


Fig. 3. a) Mould for producing bulk specimens. b) Bulk specimens in the mould.

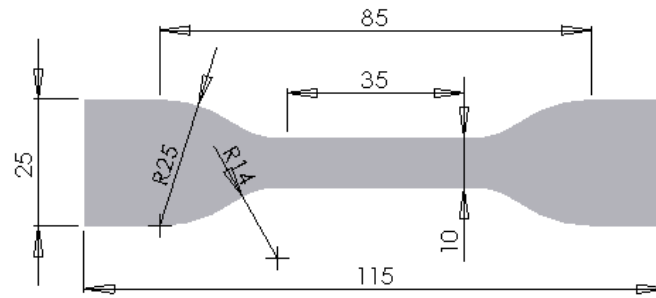


Fig. 4. Tensile test specimen geometry for the bulk specimens (dimensions in mm).

The “dogbone” adhesive specimens were tested at RT in tension using a TIRA testing machine model 2705, under a constant crosshead rate of 10 mm/min. A load cell of 5 kN was used. The TIRA testing machine recorded both the load and the crosshead displacement. When soft elastomeric materials are to be tested, contacting strain measurement techniques, such as strain gauging and ‘clip-on’ mechanical extensometry are not recommended in general. The reason is that their weight and/or method of attachment can influence the results and the point of failure. Additionally, most mechanical extensometers have limited travel and require removing from the specimen before fracture occurs. However, non-contacting strain measurement techniques offer the advantage of measuring the actual strain on the gauge length of the specimen, when using flexible materials, without any interaction over a very large strain range. Thus, a digital camera monitoring the separation of the two lines inscribed on the test specimen (see Fig.5), which define the gauge length, was used. The digital camera was set to take pictures of the gauge length every 10 sec recording the change in separation of the two lines throughout the test. The digital images were then analysed using an image processing and analysis software and the strain was extracted for each specimen. Three specimens were tested to failure for each adhesive.

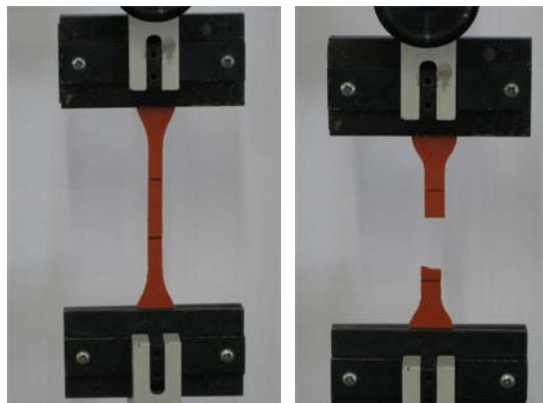


Fig. 5. A RTV 106 “dogbone” specimen clamped in the TIRA machine during the test (a) and after failure (b).

Results: The RTV “dogbone” specimens were tested at RT. For each specimen, a tensile stress-strain curve was produced based on load, displacement values and specimen dimensions. From the stress-strain curve, the elastic modulus was calculated (Table 1). The values for Young’s modulus were calculated from the tangent to the tensile stress-strain curve at the origin (a polynomial approximation of the curve was made). Representative tensile stress-strain curves of the RTV silicone adhesives can be seen in Fig. 6.

Table 1. Tensile modulus and strength data.

Adhesive	Young’s Modulus (MPa)	Tensile Strength (MPa)	Tensile Strain (%)
AS1805	2.63 ± 0.03	1.4 ± 0.01	120 ± 24
RTV106	1.60 ± 0.05	2.3 ± 0.02	270 ± 15

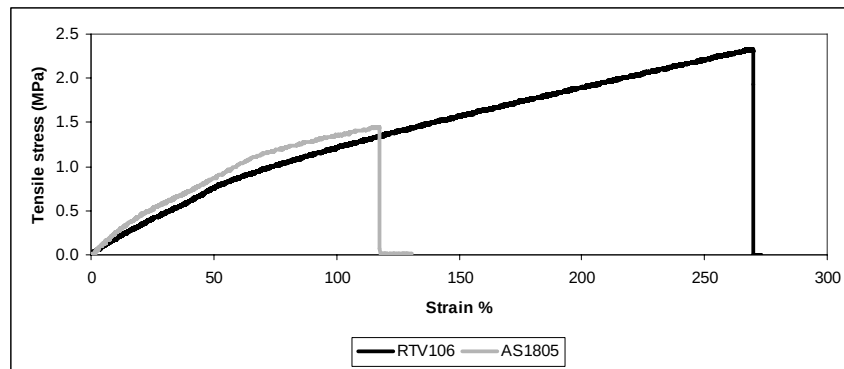


Fig. 6. Typical tensile stress-strain curves of AS1805 and RTV106 adhesives.

Tensile tests of the epoxy adhesive

The tensile properties (mode I loading) of the XN1244 adhesive were determined using “dogbone” specimens, produced from bulk adhesive plates cured in a steel mould, using a silicone rubber frame, according to the French standard NF T 76-142 [21]. Curing of the bulk plates was carried out in a hot press (1h at 140°C). The XN1244 specimens were tested in tension using a universal testing machine Shimadzu® model AUTOGRAPH, under a constant crosshead rate of 1 mm/min. Strains were measured by a video extensometer Messphysik ME46, along a length of 50 mm between hand-painted marks. For high temperatures, the environmental chamber of the machine was used to reach the desired temperature (see **Paper 3**).

3.1.2 Thick Adherend Shear Test (TAST)

Thick Adherend Shear Tests (TAST) for mode II loading followed the guidelines of standard ISO 11003-2:1999 [22], using steel substrates of dimensions 110 x 25 x 12 mm³. The joint surfaces were grit blasted and degreased with acetone prior to the application of the adhesive. The bondline thickness was nominally 0.7 mm and the length of the overlap test section was 5 mm. Two spacers (1.5 mm thick) were inserted in the gaps between the adherends after the application of the adhesive and prior to curing in order to provide the necessary spacing between the two adherends. These spacers were removed after the adhesive was cured. The joints were cured following the manufacturer's suggested curing conditions. TAST tests were performed at room temperature on a MTS servo-hydraulic machine, model 312.31, at a constant crosshead rate of 0.1 mm/min. The displacement was measured with two methods: a 25 mm length MTS extensometer and a non-contact method (video microscopy). As the extensometer is mounted in the metallic substrate, the extensometer measures not only the displacement of the adhesive, but also the displacement of the adherend. Therefore, it is necessary to apply a correction to the measured displacements. At the same time a video microscopy was used to record the displacements, which gives only the adhesive displacement. For the XN1244 epoxy adhesive the strains were calculated using the spatial correlation method developed by Choual [23]. However, da Silva *et al.* [24] showed that the steel deformation can be neglected in the case of flexible adhesives, so that the RTV silicone adhesives displacement can be measured only by the MTS extensometer method.

3.2. Fracture tests

For the design of adhesively bonded joints for high temperature applications, the availability of reliable damage models to predict their fracture behaviour in these extreme conditions is fundamental. Therefore, in order to apply a fracture mechanics or damage mechanics approach, it is essential to have the fracture toughness of the material.

The fracture toughness varies with the type of loading, i.e., modes I, II, III, as it can be seen in Fig. 7. However, in the adhesive layer of a joint, the direction of the crack and the resulting failure path is usually constrained and thus cracks may truly propagate under mixed-mode and mode II loading conditions. Modes I and II and combinations of I and II have received the most attention in the literature and are considered in this thesis. In addition, the adhesives used in structural high temperature aerospace applications must operate in extreme environments and have to maintain their mechanical properties at elevated temperatures.

Hence, their high-temperature mechanical performances, especially fracture toughness, should be taken into consideration.

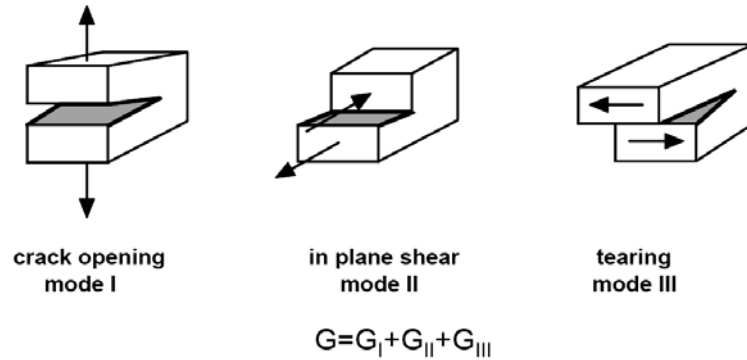


Fig. 7. Fracture modes.

Throughout this thesis, the double cantilever beam (DCB) specimen (see Fig 8) is used for pure peel (mode I) deformation and the end notched flexure (ENF) specimen (see Fig. 9), for pure shear (mode II).

3.2.1. Mode I Double Cantilever Beam (DCB) Test

In order to get the mode I fracture toughness of adhesives, the DCB test is most commonly employed. This test was initially described in the ASTM D3433 standard [25] and has been developed more recently in the international standard, ISO 2009 [26]. A schematic representation of the DCB test is shown in Fig. 8.

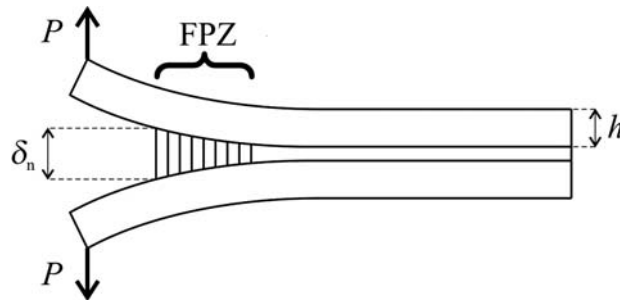


Fig. 8. Schematic representation of the DCB test.

3.2.2. Mode II End Notched Flexure (ENF) Test

For the determination of the toughness in mode II there are various test methods available: the end notched flexure (ENF) test, the end loaded split (ELS) test and the four-point notched flexure (4ENF) test. The ELS test presents large displacements and is sensitive to the clamping device [27]. The 4ENF is more sophisticated but has problems of friction due to the

loading mode in the pre-crack region [28]. Therefore, the most suitable testing method for mode II appears to be the ENF test [29,30].

The ENF test is essentially a three-point flexure test on a pre-cracked specimen causing a shear mode loading in the adhesive. However, currently no joint geometry has been standardised for mode II testing.

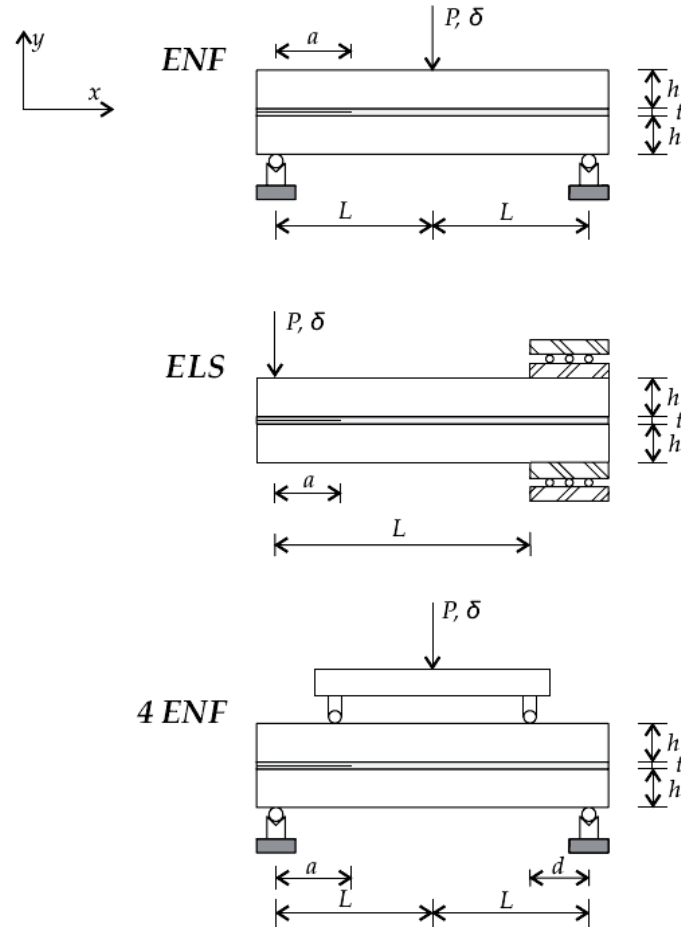


Fig. 9. Schematic representation of the end notched flexure (ENF) test, end loaded split (ELS) test, and four-point notched flexure (4ENF) test methods.

3.3. Single Lap Joints (SLJ)

It is well known, that the state of stress in the adhesive in a SLJ is neither pure shear nor uniform. There are stress intensifications and significant peel stresses exist towards the adhesive edges due to the eccentric loading. Although bending of the adherends and rotation at the overlap introduce non-uniform stress states in the adhesive preventing the accurate determination of material properties this test is often used for screening tests to compare

several adhesive systems and the effects of the environment on the adhesive properties in the selection process of the adhesive. The SLJ test is standardized in ASTM D1002-99 [31] and ISO 4587:1995 [32].

After the characterization of the high temperature adhesives, the behavior of joints made using these adhesives were analyzed. A schematic representation of the SLJ specimen used in this thesis is shown in Fig. 10. Alignment tabs, cut from the same material, were bonded at each end to improve alignment and reduce the eccentricity of the load path that causes out-of-plane bending when the specimen is tested.

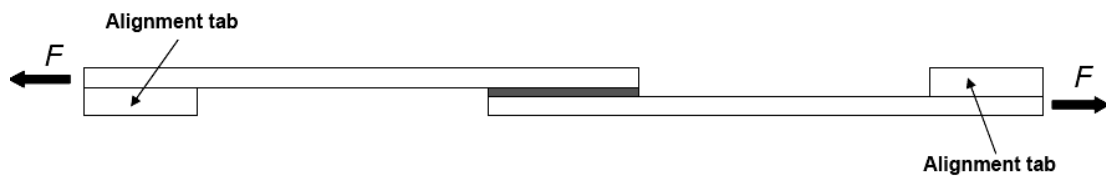


Fig. 10. Schematic representation of the single lap joint (SLJ) test.

4. Numerical modelling

The Finite Element Method (FEM) has been used for strength prediction, traditionally by using stress/strain or fracture mechanics criteria. These techniques showed acceptable results, but have some limitations: stress/strain methods are mesh dependent, while fracture criteria such as the Virtual Crack Closure Technique (VCCT) are restricted to Linear Elastic Fracture Mechanics (LEFM) and rely on the existence of an initial crack. One method that surpasses these limitations is the Cohesive Zone Models (CZM's).

CZM has received considerable attention in the last years and has been employed for a wide variety of problems and materials including metals, ceramics, polymers and composites. A CZM models the fracture process extending the concept of continuum mechanics by including a zone of discontinuity modelled by cohesive zones, thus using both strength and energy parameters to characterize the debonding process (see Fig. 11 from [33]).

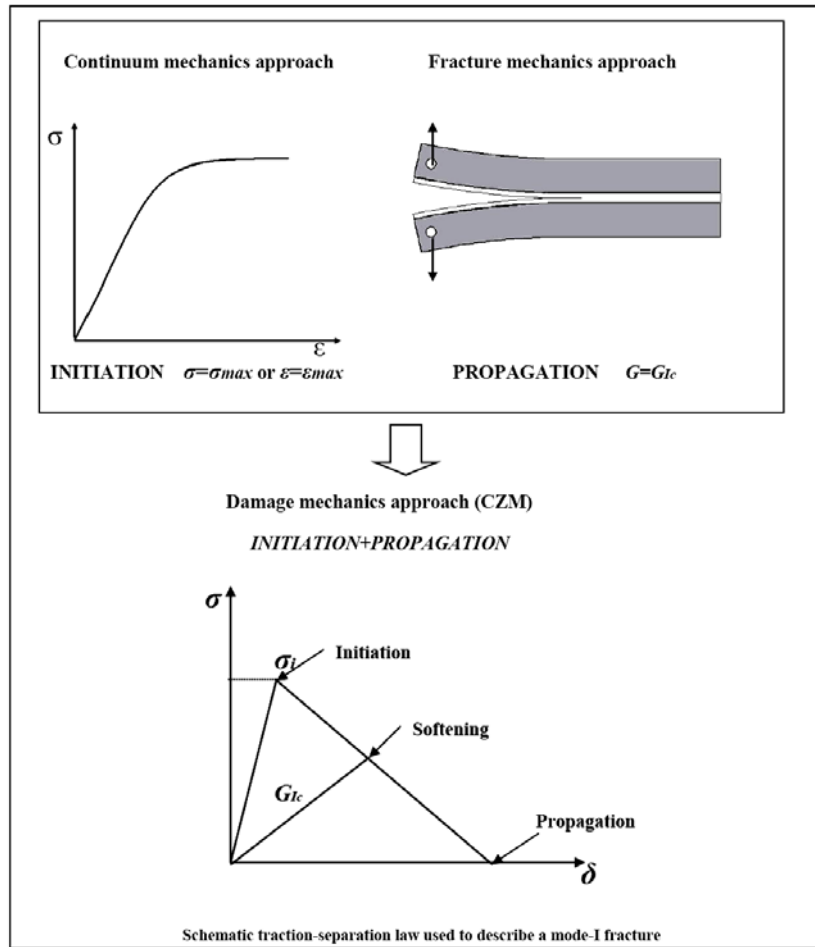


Fig 11. Schematic of a CZM for failure prediction of adhesively bonded joints [33].

The key concept of CZM is that the fracture process zone (FPZ) can be described by a traction–separation law relating stresses and displacements between the crack faces, thus simulating the gradual degradation of the material properties. In this thesis, the shape of the traction–separation law was defined as a bilinear model (Fig. 12).

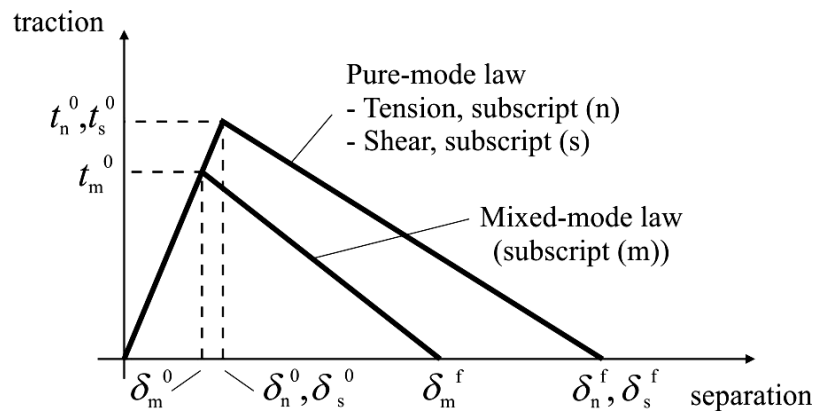


Fig. 12. Traction-separation law with linear softening available in ABAQUS®.

The subscripts n and s relate to pure normal (tension) and shear behaviours, respectively. t_n and t_s are the corresponding current stresses, and δ_n and δ_s the current values of δ . A CZM requires the knowledge of G_n and G_s along the fracture paths and respective values of G_n^c and G_s^c . Additionally, the cohesive strengths must be defined (t_n^0 for tension and t_s^0 for shear), relating to the onset of damage, i.e., cancelling of the elastic behaviour and initiation of stress softening. δ_n^0 and δ_s^0 are the peak strength displacements, and δ_n^f and δ_s^f the failure displacements. The values of δ_n^f and δ_s^f are defined by G_n^c and G_s^c , respectively, as these represent the area under the CZM laws. For the mixed mode CZM law t_m^0 is the mixed mode cohesive strength, δ_m^0 the corresponding displacement, and δ_m^f the mixed mode failure displacement.

Under pure mode loading, the respective t - δ response attains its peak at the cohesive strength (t_n^0 or t_s^0), corresponding to damage initiation by the induced reduction of stiffness of the cohesive element. Softening follows and, when the values of t are completely cancelled, the crack propagates up to the adjacent set of paired nodes in the failure path, permitting the gradual debonding between crack faces. Under mixed loading (i.e., when two or three modes of loading are simultaneously present), stress and/or energetic criteria are often used to combine the pure mode laws, thus simulating the typical mixed mode behaviour inherent to bonded assemblies.

Determination of the cohesive parameters

For the estimation of the cohesive law parameters, a few data reduction techniques are available: the property determination technique, the inverse method and the direct method.

The property determination method, consisting on the isolated definition of all the cohesive law parameters by suitable tests, is particularly critical if bulk tests are used, owing to reported deviations between the bulk and thin adhesive bond cohesive properties [34]. This is caused by the strain constraining effect of the adherends in bonded assemblies, and also by the typical mixed mode crack propagation in adhesive bonds [34, 35]. Actually, in bulk materials cracks tend to grow perpendicularly to the direction of maximum principal stress. In thin bonds, cracks are forced to follow the bond length path since, as the adhesive is typically weaker and more compliant than the components to be joined, failure is often cohesive within the adhesive [36].

The inverse method consists on iterative comparisons between experimentally measured data and the respective numerical predictions, considering a precise description of the experimental geometry and approximate cohesive laws with a parameterized shape, established based on the typical behaviour of the material that the law simulates. Using this technique, the value of fracture toughness, which corresponds to the plateau value of the respective R -curve of the fracture characterization test, is used as an input parameter in the numerical model. These models also include typical values for the cohesive strength. A few numerical iterations must be performed until a good accuracy between the experimental and numerical data is obtained. This method was used to determine the cohesive parameters of the epoxy high temperature adhesive studied as a function of temperature in **Papers 6 and 7**.

The direct method is also available, allowing the extraction of the cohesive law of an adhesive layer directly from the measured data from the experiments without requiring such a laborious numerical analysis. This method is based on the simultaneous measurement of the J -integral and the end-opening (normal or shear) of the cohesive zone. This has been successfully employed in the extraction of traction–separation laws for the RTV106 silicone adhesive bonds in **Paper 4**.

Validation of the CZM

In order to validate the cohesive laws obtained, SLJs as a function of temperature were performed and numerical predictions based on the use of the CZM were obtained for the high temperature epoxy adhesive in **Paper 8**. Hence, the tension and shear cohesive parameters (G_n^c , G_s^c , t_n^0 , t_s^0) determined in **Papers 5-7** were combined with a mixed mode propagation criteria to provide quantitative predictions of the deformation and failure load of SLJs under a mixed mode loading. The FE predictions for different temperatures showed a reasonable agreement with the experimental results.

5. Conclusion

In this thesis, the mechanical behaviour of two different types of adhesives, one adhesive suitable for high temperatures and another suitable for low temperatures was investigated. The combination of these two adhesives would guarantee a joint to be suitable from low to high temperatures, for bonding of temperature protection tiles on the space shuttle.

The main conclusions of this research are summarized below:

The performances of two different RTV silicone adhesives were investigated in **Paper 2**. TAST was performed in order to measure the shear properties of the adhesives. SLJs were fabricated and tested to assess the adhesive performance in a joint. The influence of temperature on the lap-shear strength of the adhesives was investigated. The lap-shear strength of both adhesives decreased with increasing of temperature. The effect of bondline thickness and overlap length on the lap-shear strength of the adhesives was also studied. The reduction of failure load with an increase of the bondline thickness is a very common situation when dealing with structural adhesives. However, for the tested RTV silicone adhesives the failure loads increased as the bondline was made thicker. Also, the failure loads of RTV silicone adhesive joints increased almost proportionally with increasing the overlap length.

In **Paper 3**, tensile data obtained from tests at RT showed that the XN1244 adhesive has a high strength and high stiffness. Large increases in ductility (strain to failure) and reductions in ultimate strength were measured at elevated temperatures (100°C, 125°C and 150°C). The ultimate tensile stress linearly decreased with temperature while logarithmically increased with the test speed. The Young's modulus nonlinearly decreased with the temperature, while shows a similar behaviour as the ultimate tensile stress as a function of test speed. The ultimate tensile strain nonlinearly increased with temperature, while has a more complex behaviour in relation to the test speed. As expected, the temperature effect on the properties of the adhesive is more significant than that of the strain rate.

In **Paper 4**, pure mode I traction–separation laws were obtained for RTV106 silicone/steel DCB specimens. The cohesive laws were obtained directly from the experiments by differentiation of simultaneously measured data (the J -integral and the end-opening displacement). The fracture toughness and the traction–separation laws exhibited temperature dependence. The fracture toughness, the peak cohesive stress and the respective end-opening displacement decreased with the temperature rise, while the critical opening displacement increased at 100°C and decreased at 200°C from the RT.

In **Papers 5 and 6**, the mode I cohesive parameters (mode I adhesive fracture toughness (G_{Ic}) and maximum tensile stress) of XN1244 adhesive at different temperatures (RT, 100°C,

150°C and 200°C) were determined. Tensile tests and pure mode I DCB adhesive fracture toughness tests were performed at room and high temperatures. The results of the tensile tests showed that the ultimate tensile stress decreased monotonically with the temperature. At 200°C, as the temperature increased above the T_g of the adhesive, the adhesive strength and strain drastically reduces. The value of G_{Ic} was found to be relatively insensitive to temperature up to T_g , while above T_g (at 200°C) a drastic decrease in G_{Ic} was found. CZMs have been used to characterize the adhesive behaviour and to predict the adhesive $P-\delta$ curves as a function of temperature. The simulation response for various temperatures matches the experimental results very well. From the numerical simulations, the parameters that describe the bilinear CZM were determined.

In **Paper 7**, the mode II cohesive parameters (mode II adhesive fracture toughness (G_{IIc}) and maximum shear stress) of XN1244 adhesive at different temperatures (RT, 100°C, 150°C and 200°C) were determined. Pure mode II ENF adhesive fracture toughness (G_{IIc}) tests were performed at room and high temperatures (100°C, 150°C and 200°C). The mode II fracture toughness was found to increase at 100°C and decrease at 150°C, as the temperature approaches T_g . Moreover, a drastic drop in fracture toughness was observed at 200°C, when the T_g of the adhesive was overpassed. CZMs have been used to characterize the adhesive behaviour and to predict the adhesive $P-\delta$ curves as a function of temperature. The simulation response matched the experimental results very well at temperatures below T_g , while above this value of temperature it was not possible to mimic the experiments as a clear drop of load could not be related to crack propagation due to the excessive degradation of the adhesive. From the numerical simulations, the parameters that describe the bilinear CZM were determined.

Finally, the main objective of **Paper 8** was to evaluate the capabilities of using the current implementations of CZM available in ABAQUS® to simulate the behaviour and strength of adhesively-bonded joints as a function of temperature and validate experimentally the damage laws for pure mode I and pure mode II, determined previously. With this purpose, SLJs were fabricated and tested at RT and high temperatures. CZMs have been used to characterize the adhesive behaviour as a function of temperature. The simulation response for various temperatures matches reasonably well the experimental results.

A database of mechanical properties of the high temperature adhesives studied was established which will be used in finite element (FE) analysis to simulate the deformations at different testing conditions. Also, the experimental determination of the cohesive parameters at different temperatures allows the definition of cohesive laws that will be subsequently used in FE simulations to model the fracture of bonded structures (e.g. ceramic/aluminium) as a function of temperature. The major contribution of this research is the development of methods to measure temperature dependence of cohesive laws and the determination of the cohesive laws for different temperatures of the high temperature adhesives studied.

The methods presented in this thesis have potential for improving the prediction of joint behaviour over a range of temperatures. However, the field is still developing and significant research is still needed.

6. Future work

Low temperatures tests

In this thesis, the mechanical behaviour of the high temperature adhesives selected for the joint proposed was evaluated at high temperatures. The logical step forward should be to investigate the behaviour of these adhesives under conditions that better approach the conditions occurring in the space shuttle in service. Therefore, the behaviour at low temperatures should be investigated.

Effect of adhesive thickness

The thickness used in **Papers 5-8** for XN1244 adhesive was 0.2 mm. However, it would be very useful if the effect of adhesive thickness could be assessed and, if necessary, incorporated into the CZM. Fracture tests may be completed with more thicknesses between 0.1 mm and 1 mm. This will help understand the complete shape of the thickness dependence P - δ curves as a function of temperature.

Rate effects

It is well known that the tests performed under different states of stress should be carried out under similar test temperatures and rates of strain in the adhesive layer. In this thesis, all the tests were carried out in environmental chambers with the temperatures controlled and under similar test speeds. However, for generalization of the cohesive laws to structural predictions additional investigations on the effect of strain rate in the cohesive parameters is required.

In **Paper 3**, the combined effect of the temperature and test speed on the bulk tensile properties of XN1244 adhesive was investigated. However, adhesives in the form of bonds behave differently than as a bulk material, because of the strain constraining effects of the adherends and the typical mixed mode crack propagation. Hence, the possible rate effects in the mode I and II fracture toughness may be investigated, and, if necessary, incorporated into the CZM.

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Paper 1

Literature review

Adhesively bonded joints in composite materials: An Overview

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Abstract

A review of the investigations that have been made on adhesively bonded joints of fiber-reinforced plastic (FRP) composite structures (single skin and sandwich construction) is presented. The effects of surface preparation, joint configuration, adhesive properties and environmental factors on the joint behaviour are described briefly for FRP composite structures adhesively bonded. The analytical and numerical methods of stress analysis required before failure prediction are discussed. The numerical approaches cover both linear and non-linear models. Several methods that have been used to predict failure in bonded joints are described. There is no general agreement about the method that should be used to predict failure as the failure strength and mode are different according to various bonding methods and parameters, but progressive damage models are quite promising since important aspects of the joint behaviour can be modelled using this approach. However, a lack of reliable failure criteria still exists, limiting in this way a more widespread application of adhesively bonded joints in principal load bearing structural applications. An accurate strength prediction of the adhesively bonded joints is essential to decrease the amount of expensive testing at the design stage.

Keywords: Adhesively bonded joints, fiber-reinforced plastic composite materials, sandwich panels, analytical methods, finite element modelling.

1. Introduction

Adhesive bonding is a material joining process in which an adhesive, placed between the adherend surfaces, solidifies to produce an adhesive bond. Adhesively bonded joints are an increasing alternative to mechanical joints in engineering applications and provide many advantages over conventional mechanical fasteners. Among these advantages are lower structural weight, lower fabrication cost, and improved damage tolerance. The application of adhesively bonded joints in structural components made of fiber reinforced composites has increased significantly in recent years. The traditional fasteners usually result in the cutting of fibres and hence the introduction of stress concentrations, both of which reduce structural integrity. By contrast, bonded joints are more continuous and have potential advantages of strength-to-weight ratio, design flexibility and ease of fabrication. In fact, adhesive bonding has found applications in various areas from high technology industries such as aeronautics, aerospace, electronics, and automotive to traditional industries such as construction, sports and packaging. These applications are in the form of single skin as well as sandwich configurations. The structures could potentially be made up using different fibre types, fibre architectures and weaves, and resins.

Bonded joints are frequently expected to sustain static or cyclic loads for considerable periods of time without any adverse effect on the load-bearing capacity of the structure. A lack of suitable material models and failure criteria has resulted in a tendency to “overdesign” composite structures. Safety considerations often require that adhesively bonded structures, particularly those employed in primary load-bearing applications, include mechanical fasteners (e.g. bolts) as an additional safety precaution. These practices result in heavier and more costly components. The development of reliable design and predictive methodologies can be expected to result in more efficient use of composites and adhesives. In order to design structural joints in engineering structures, it is necessary to be able to analyse them. This means to determine stresses and strains under a given loading, and to predict the probable points of failure. There are two basic mathematical approaches for the analyses of adhesively bonded joints: closed-form analysis (analytical methods) and numerical methods (i.e. finite element analyses).

On the other hand, sandwich composite constructions are increasingly used in various applications in many industries due to the combination of high strength and low weight, leading to a highly efficient structure. Other advantages offered by sandwich construction are elimination of welding, high insulating qualities and design versatility. In its simplest form,

this type of construction usually consists (Figure 1) of two thin outer face sheets of stiff, strong material (in this case a composite) separated by a thick, lightweight layer of core material (e.g. foam, honeycomb or corrugated). Virtually all types of composites from sheet molding compound to prepregs can be used as skins with a wide range of polymeric, metallic and ceramic materials used as core materials.

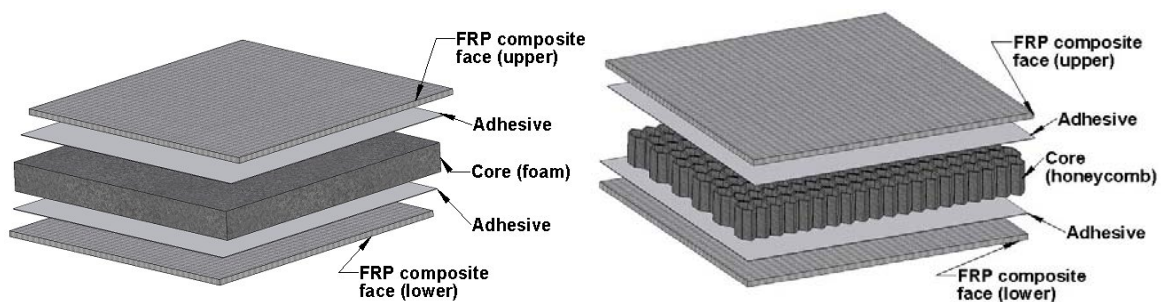


Figure 1. Sandwich principle.

Most of the early work on adhesively joining of composites was done in the seventies and early eighties for the aerospace industry. There have been many analytical, finite elements, and experimental studies performed over the years. Matthews et al. [1] give a very comprehensive review on the strength of adhesive joints in fibre-reinforced plastics. Since then, a considerable number of research studies have been conducted, new analytical approaches have been developed and finite element methods are more widely used. This present review is focused on published works concerning adhesively joining of FRP composites structures. The literature dealing with joining of composite structures with adhesives concentrates on investigating the bond strength. The topics of particular interest are: surface preparation, joint configuration, adhesive properties, environmental conditions, analytical and finite element analyses of joints and test methods. Despite composite (single skin and sandwich) structures having gained substantial importance in many applications where the weight saving has become more and more important, a lack of reliable material models still exist limiting in this way a more widespread application of adhesively bonded joints of composite structures in principal load bearing structural applications.

2. Effect of surface preparation

The surfaces play an important role in the bonding process and is, perhaps, the most important process step governing the quality of an adhesive bond joint [2]. Appropriate pre-treatment

can sometimes confer additional properties to the surfaces. Surface treatments prior to the application of adhesives are recommended in order to achieve maximum mechanical strength. Bond strength can be significantly improved by surface treating the adherends prior to bonding. The formation of a suitable surface chemistry is the most important step in the surface preparation process because the integrity of this surface directly influences the durability of the adhesive bond as pointed out by Davis and Bond [3]. They have investigated the factors affecting the durability problem of adhesively bonded joints using the “clean surface concept” approach. The most common misconception in surface preparation is that the only requirement for a good bond is a clean surface. A clean surface is a necessary condition for adhesion but it is not a sufficient condition for bond durability. Most structural adhesives work as a result of the formation of chemical bonds (mainly covalent, but some ionic and static attractive bonds may also be present) between the adherend surface atoms and the compounds constituting the adhesive [4]. These chemical links are the load transfer mechanism between the adherends. Most adhesive bond failures can be attributed to poor processes during fabrication, with lack of quality surface preparation being the most significant deficiency [3].

Typical composite surface treatments include traditional abrasion/solvent cleaning techniques for thermoset composites, while thermoplastic composites require surface chemistry and surface topographical changes to ensure strong and durable bond strength. For thermoplastic composites the primary aim of the surface treatment is to increase the surface energy of the adherend as much as possible. Surface treatments decrease water contact angle, increase surface tension and as a result increase bond strength [5].

A variety of surface treatments have been used with various degrees of success to increase surface tension, increase surface roughness, change surface chemistry and thereby increase bond strength and durability of polymer composite adhesive joints: abrasion/solvent cleaning, grit blasting, peel-ply, tear-ply, acid etching, corona discharge treatment, plasma treatment and laser treatment. However, the recommended surface preparation for thermoplastic composites is a light aluminum oxide grit blast in dry nitrogen [3]. The abrasion should just remove the surface of the resin without exposing fibers. Because the epoxy surface bonds well to other epoxies, no chemical modification is required.

By increasing surface tension, increasing surface roughness and changing surface chemistry, a more intimate bond can be formed, which allows for increase in strength and durability.

3. Effect of joint configuration

Joints represent one of the greatest challenges in the design of structures in general and in composite structures in particular since they entail discontinuities in the geometry of the structure and/or material properties, and introduce high local stress concentrations.

Unlike surface preparation, joint configuration, is usually a product of design. A wide variety of joints are available to the designer as discussed by Adams and Wake [6]. Commonly joint configurations which have been analysed in the literature are single-lap joints, double-lap joints, scarf joints, and stepped-lap joints (Figure 2). Other configurations have also been studied, such as strap joints, butt joints, butt strap joints, corner joints, stepped scarf joints, T-shaped joints, L-shaped joints, double-doubler joints, tubular lap joints, etc. Chamis and Murthy [7] describe a step-by-step procedure for the preliminary design of composite adhesive joints (including single, double and step lap joints, and scarf joints) for use in hot/wet service environments under static and cyclic loads.

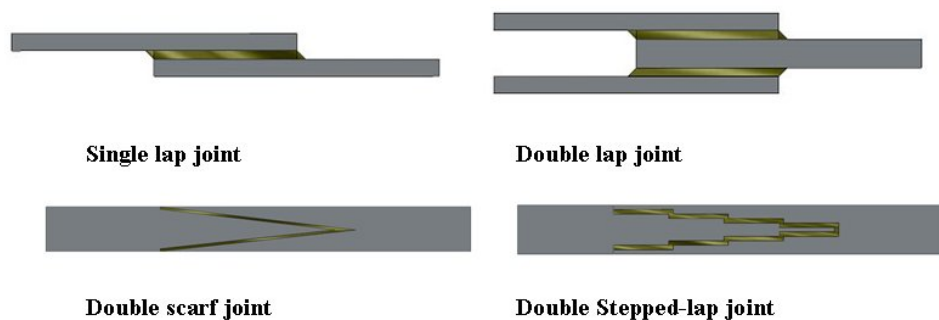


Figure 2. Adhesive bonded joints configurations.

The strength of a given type of joint depends, for a given type of load, on the stress distribution within the joint. This stress distribution depends on the joint geometry and the mechanical properties of adhesive and adherend. In the case of FRP composite substrates, the high through-thickness stresses at the overlap ends are of particular concern, due to the relatively low through thickness strength of most composite materials. This often means that joints made with high strength adhesives are more likely to fail prematurely in the composite before failure in the adhesive occurs (Figure 3). The joint should be designed to minimize stress concentrations. Some stresses, such as peel and cleavage, should be minimized and others maximized, i.e. shear, compressive stresses.

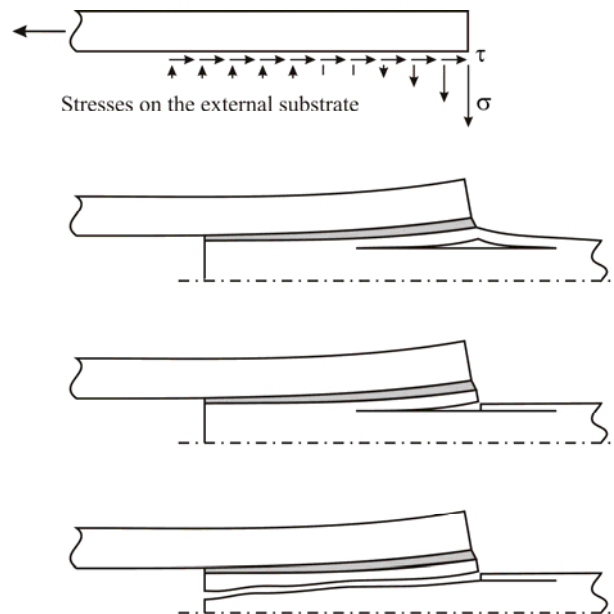


Figure 3. Failure in adhesive double lap joints due to transverse (through the thickness) stresses of the composite substrates.

In composite/metallic adhesive joints, the layered nature of composite adherends and relative weakness in the through-the-thickness direction makes the failure mechanism more complex [8]. Due to these uncertainties in joint strength many designers use higher safety margins in their structure resulting in a non optimum use of materials. For example, slight variation in joint design can vary the peel stresses. The double lap, the scarf and the stepped joints are designed to decrease the peel stresses when comparing to single lap shear joints. Adherend shaping is also used to decrease the peel stresses in the composite joints (see Figure 4).

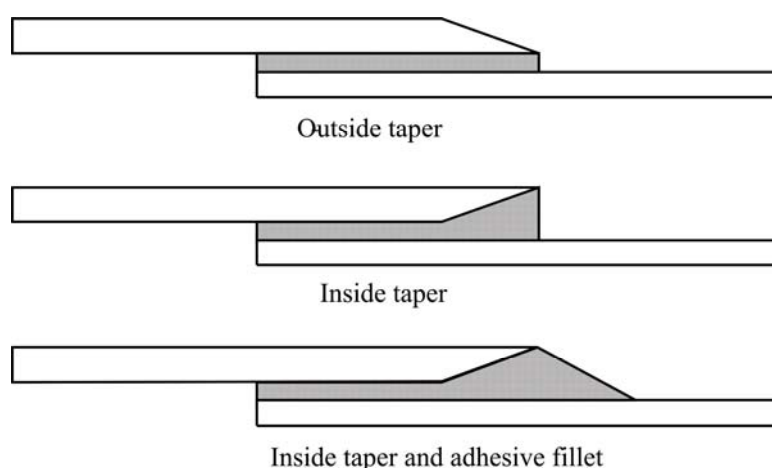


Figure 4. Adherend shaping to decrease the peel stresses in the composite joints.

The single-lap joint is the most common joint used mainly due to its simplicity and efficiency. However, one of the problems associated to this joint is the fact that the stress distribution (shear and peel) is concentrated at the ends of the overlap. Various techniques have been adopted by researchers to improve the efficiency of single-lap joints. These include altering the adherend geometry [9-11], adhesive geometry [12,13] and spew geometry [14-17].

Studies related to altering adherend geometry have been focused towards adherend tapering and stepping. Taper angle and taper edge thickness were the variable parameters considered in most of the studies and design guidelines exist for specific application.

Lang and Mallick [13] introduced a new bonding technique by removing portions of the adhesive layer (recessing) from the overlap. Their study indicates that the average strength of the bond increases with recessing. This positively contributes towards weight and cost reduction. However, the small effective lap length due to recessing may adversely affect the fatigue failure.

Spew shape and size, (the spew is the result of the adhesive squeezed out of the lap region at the moment of the joint manufacture), were the other parameters studied to reduce stress concentration [14-17]. The peak stress is dependent on the size and shape of the spew. It is shown that shaping the spew can provide smoother transition in joint geometry significantly reducing the stress concentration [14].

Zeng and Sun [18] proposed a novel “wavy” lap joint configuration (Figure 5). In this joint, the through-thickness stresses at the edges of the overlap are compressive, which provided a significant improvement in joint strength, especially in fatigue performance. However, geometrical changes have constraints in terms of manufacture. For example, from the fabrication point of view, adherend shape other than tapering and stepping would be complex. The fabrication constraint applies equally to adhesive geometry alterations.



Figure 5. “Wavy” lap joint configuration (redrawn from [18]).

One technique that is being intensively investigated at this moment is the use of more than one adhesive along the overlap (an adhesive bondline with variable modulus to relieve the

high-stress concentrations at the end regions of the overlap) in order to have a more uniform stress distribution [19-23]. A flexible and ductile adhesive is placed at the ends of the overlap and a rigid and brittle adhesive is placed in the center of the overlap (Figure 6). Although this approach has been discussed theoretically [19,20], there have been relatively few published experimental demonstrations of a practical method that yields significant improvements in the joint performance. da Silva and Adams [22,23] investigated theoretically and experimentally dual adhesive metal/composite joints and showed that there is a real improvement in joint strength, especially if the difference of coefficients of thermal expansion is high. To date, few studies are available where the material property of the adhesive are gradually altered to achieve better joint performance.

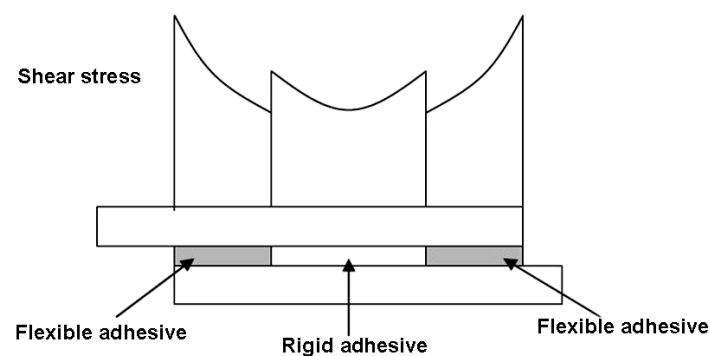


Figure 6. Mixed - adhesive bonded joint.

Another technique is varying the adherend material properties in the overlap region. Ganesh et al. [24] have shown that composite materials with continuously varying material properties can be fabricated by modifying the conventional braiding technology of fiber placement. With this technology, locally varying the adherend modulus by design in the overlap region is possible. Ganesh and Choo [25] showed that spatial grading of adherend elastic modulus of an adhesively bonded single-lap joint reduced the peak elastic shear stresses and caused more uniformly distributed stresses in the adhesive layer. Boss et al. [26] also compared modulus grading of the adherends with geometrical grading of the adherends, and showed that the modulus grading provided reduced stresses and that combining modulus and geometrical grading could also yield a better performing adhesive joint design.

In practice, adhesive joints are subjected to thermal as well as structural loads. The mismatches in the thermal and mechanical properties of the adhesive and composite adherends cause deformations and stresses induced by thermal fields in all members of the adhesive joint, especially in the adhesive layer. For example, adhesives with high curing

temperatures may be unsuitable for some low temperature applications because of large thermal stresses which develop as the joint cools down from the curing temperature.

Thermal loads are especially important when bonding dissimilar materials (with different coefficients of thermal expansion), as large differences in thermal expansion characteristics between adherends can cause severe problems [27]. For example, for aluminium/composite adhesively bonded joints, the difference in thermal expansion between the adherends is relatively large, giving considerably higher thermal stresses. In addition, carbon/epoxy has a particularly low thermal expansion, so that when bonded to metals these materials tend to produce higher thermal stresses than when are bonded to other materials.

Rastogi et al. [27] studied three-dimensional (3D) thermal stress distributions in aluminium-to-composite, symmetric, double lap joints subjected to uniform temperature loads. They found that the joint corners are critical regions for debonding initiation.

Owens and Lee-Sullivan [28] studied stiffness loss due to crack growth in composite-to-aluminium joints. They tested single lap joints at room temperature and at -40°C at quasi-static conditions and found that the joint stiffness is more affected by the response of the adherends to the test temperature than by the modulus of the thin adhesive layer.

Studies on composite scarf joints or repairs were employed by many researchers [29-33]. However, a scarf joint in a composite structure is more complex because, unlike lap or stepped-lap joints, the stiffness of the bonded surface varies along the bondline. The stress distribution (peel and shear) can vary significantly along the bondline in response to changes in ply orientation [34]. Significant stress concentrations exist in scarf joints between composite adherends of identical lay-up [31,32]. The stress concentration factor depends strongly on the stacking sequence and thickness of the laminates. Since fibres themselves do not cross the bondline, the large difference in stiffness between the adhesive and the composite plies, especially 0° plies, induces significant stress variations along the scarf.

The development of adhesively bonded sandwich joints has been driven by the requirement for reduced structural weight and the ever-increasing demand for more efficient structures. Several typical joint configurations for sandwich panels (foam core) are illustrated in Figure 7.

Joining of sandwich structures is a complex task. For example, in a sandwich T-joint, joining two sandwich panels at right angle to each other with continuous fiber reinforcement at the corners is considerably difficult [35-39]. Continuous fiber-reinforcement facilitates efficient load transfer between the two composite parts and increases the joint strength substantially.

The composite T-joint is being used extensively in the marine and aerospace industries. A typical design of this type of joint consists of panels joined by filler and overlaminates (Figure 8). Various improved T-joints have been designed and investigated [37-46], some with focus on improved strength, and others with focus on reduced weight. Also, T-joint connection can have foam fillets in the form of a pad or two triangular inserts combined with overlaminates [40]. Sandwich panels joined by use of filler and two triangular PVC foam fillets (core triangles) were investigated theoretically and experimentally by Toftegaard and Lystrup [46].

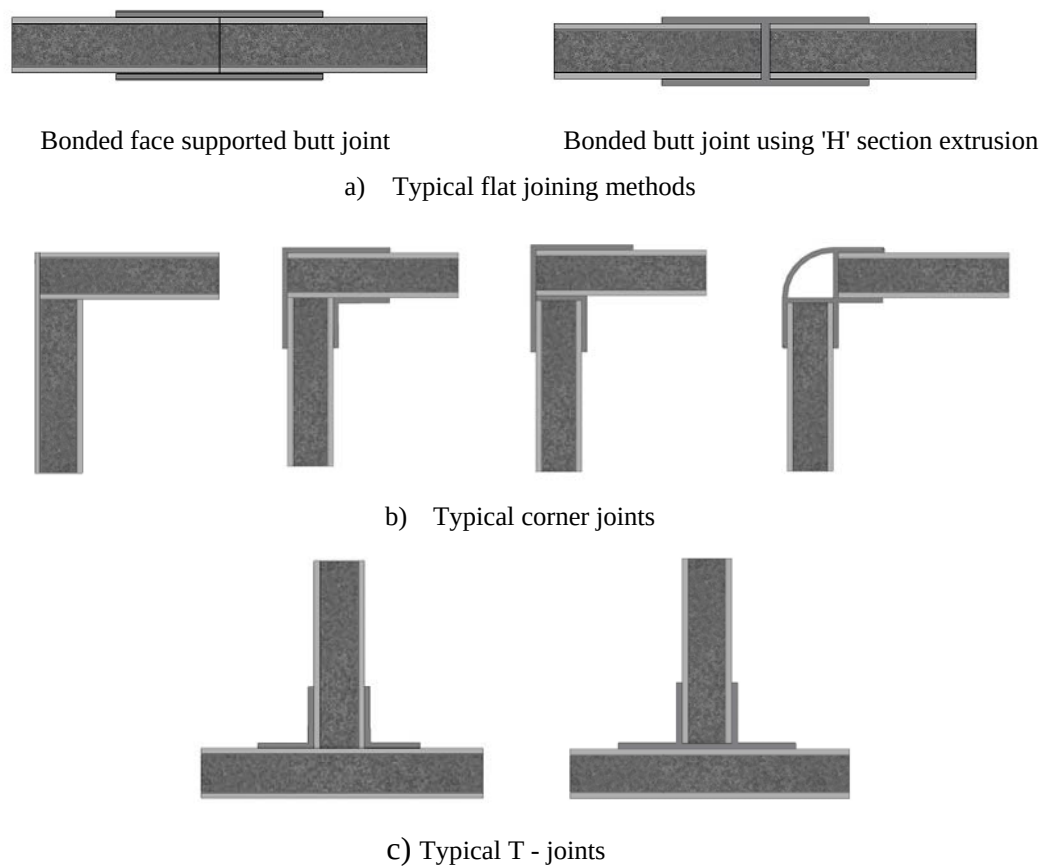


Figure 7. Typical joints configurations for sandwich panels.

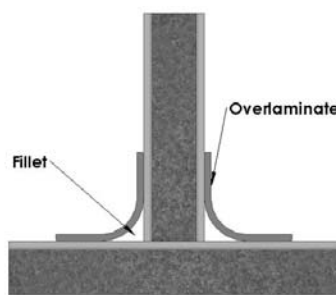


Figure 8. Typical T-joint configuration with circular fillet.

4. Adhesive properties

Adhesives used in structural applications include: epoxies (having high strength and temperature resistance), cyanoacrylates (fast bonding capability to plastic and rubber but poor resistance to moisture and temperature), anaerobics (suitable for bonding cylindrical shapes), acrylics (versatile adhesives with capabilities of fast curing and tolerate dirtier and less prepared surfaces), polyurethanes (good flexibility at low temperatures and resistant to fatigue), silicones (excellent sealant for low stress applications, high degree of flexibility and very high temperature resistance) and high-temperature adhesives (phenolics, polyimides and bismaleimides). Table 1 presents several typical properties for different types of adhesives.

The increased usage of high-temperature resin-matrix systems for composite materials has necessitated the development of compatible and equally heat stable adhesive systems. Epoxy adhesives, which are very frequently used for the composite matrixes, are commonly used to bond composites based on epoxy matrix because of the compatibility between resin and adhesive. Typical mechanical properties values for different types of adhesives are presented in Table 2.

Before an adhesive can be specified for an application, screening tests should be conducted in order to compare and evaluate the various adhesion parameters. This is especially true for structural adhesives where failures during actual use can have devastating consequences. Properties of adhesives can vary greatly and an appropriate selection is essential for a proper joint design. Thus, to determine the stresses and strains in adhesive joints in a variety of configurations, it is necessary to characterize the adhesive behaviour in order to know its mechanical properties, particularly the stress-strain curve and the modulus [47]. The approaches used for determining the properties of adhesives are the measure of the properties of bulk adhesive specimens and the use of specially designed joint geometries with a thin bond line (often referred to as “*in-situ*” testing).

As it is well known, to achieve a good bond, first it is necessary to start with a good adhesive. The adhesive selection process is difficult as there is no universal adhesive that will fulfil every application and the selection of the proper adhesive is often complicated by the wide variety of available options. However, adhesive selection includes many factors such as: type and nature of substrates to be bonded, cure and adhesive application method and the expected environments and stresses that the joint will face in service. Also the cost of the adhesive may sometimes be an important factor of adhesive selection in a particular production situation.

Table 1. Typical properties of adhesives.			
	Comments	Service temperature (°C)	Cure
Epoxy	High strength and temperature resistance, relatively low cure temperatures, easy to use, low cost.	-40 – 100 (180*)	One-part epoxies cure with temperature. Two-part epoxies cure at room temperature (cure can be accelerated with temperature).
Cyanoacrylates	fast bonding capability to plastic and rubber but poor resistance to moisture and temperature	-30 - 80	Fast cure (second or minutes) upon exposure to moisture at room temperature.
Anaerobics	designed for fastening and sealing applications in which a tight seal must be formed without light, heat or oxygen, suitable for bonding cylindrical shapes	-55 - 150	Cure in the absence of air or oxygen at room temperature.
Acrylics	versatile adhesives with capabilities of fast curing and tolerate dirtier and less prepared surfaces	-40 - 120	Cure through a free radical mechanism.
Polyurethanes	good flexibility at low temperatures and resistant to fatigue, impact resistance, and durability	-200 - 80	Room temperature
Silicones	excellent sealant for low stress applications, high degree of flexibility and very high temperature resistance, capability to seal or bond materials of various natures, long cure times and low strength.	-60 - 300 (350**)	Room temperature
Phenolics	good strength retention for short periods of time, limited resistance to thermal shocks	-40 – 175 (260**)	Cure with temperature and high pressure.
Polyimides	thermal stability, dependent on a number of factors, difficult processability	-40 – 250 (300**)	Cure with temperature and high pressure.
Bismaleimide	very rigid, low peel properties	-50–200 (230**)	Cure with temperature and high pressure.

*with different filler materials,

**intermittent

Table 2. Typical adhesives mechanical properties values.

Adhesive	Type	Shear Modulus G MPa	Shear Strength MPa	Shear Strain %
AV138 [129]	Epoxy	1559	30	7.8
DP805 [129]	Acrylic	159	8.4	180
SikaFlex 265*	Polyurethane	0.7	4.5	450
RTV 106*	Silicone	-	1.3	400
AS1805	Silicone	0.68	1.47	330
Redux 326 [22]	Bismaleimide	1180	36.5	3.63

* Manufacturer data

A great variety of test geometries and specimens are used to obtain adhesive properties. The measured parameters are the load and strain needed to create failure. The test geometry should provide a pure state of stress, uniformly distributed across the contact surface and through the bondline, free of stress concentrations, and the surface treatment should be sufficient to ensure cohesive failure in the adhesive layer.

Currently there are many ASTM and ISO standards, which have been written to analyze and experimentally verify adhesive properties. These standards provide a basis for testing. Commonly test methods that have been developed and used to obtain properties of the adhesives include: tensile tests, shear tests, compression, peel, durability tests and dynamic tests. The typical test for strength characterization is lap shear test; for the fracture toughness it is the double cantilever beam test; and for the assessment of the resistance to solvents it is the wedge test.

There are several test methods in order to get the shear properties of the adhesives, such as: notched beam shear method (Iosipescu), notched plate shear method (Arcan), torsion of bulk material, butt torsion (napkin ring or solid specimen) and thick-adherend shear test method (TAST). The TAST (ISO 11003-2:1993) is usually preferred for determining design parameters as the thick, rigid adherends reduce (but not eliminate) the peel stresses. The state of stress is predominantly shear but there are peel stresses at the end of the overlap. However, the most widely used adhesive-bond test specimen is the single lap tension test. The failure mode of the single lap joint is rarely controlled by the shear strength of the adhesive but is largely the result of joint deflections and rotations and induced peel stresses. Because of the rotation at the overlap, data from single overlap tension test specimen cannot be used to obtain adhesive shear design data but are often used for screening tests to compare several

adhesive systems and the effects of the environment on the adhesive properties in the selection process of the adhesive.

5. Environmental factors

Adhesively bonded joints may be exposed to various environmental conditions during their service life. As has been shown, the performance of adhesive systems can be considerably deteriorated when exposed to harsh environments. As stated by Vodicka [48] the environmental factors which can affect the properties of an adhesively bonded joint can affect in turn the ultimate mechanical performance of the joint. These factors must be considered a critical factor in determining the long-term durability of adhesively bonded joints and need to be carefully identified and related to the type of service the material will see.

The main environmental factors in climatic exposure are temperature and humidity. The prolonged exposure or even short term exposure to elevated temperatures will often produce irreversible chemical and physical changes within adhesives. As the temperature increases, the bond strength decreases. Also, the moisture absorbed in a polymeric material can lead to a wide range of effects, both reversible and irreversible, including plasticization, swelling, and degradation. At temperatures below the glass transition temperature T_g , polymer property reduction is reversible upon dehydration, whereas above T_g , matrix properties are permanently degraded.

The presence of moisture in adhesive joints may not only weaken the physical and chemical properties of the adhesive itself but also the interface between the adhesive and the substrate. In the case of composite joints exposed to humid environments the mechanisms of degradation are quite different compared to adhesively bonded metal joints. Unlike metals, the work of adhesion for composite to epoxy joints remains positive in the presence of water [49], thus decreases the likelihood of interfacial failure on ageing. Also, the composite adherend will absorb water, which can affect the kinetics of water absorption into the adhesive. Temperature and moisture can also influence the mechanical properties of the composite matrix material and the interface between fibres and matrix may be weakened in the presence of moisture [50].

Kinloch [50] suggested a wide range of degradation mechanisms including plasticization, hydration, microcracking of the polymer and fiber-matrix weakening in composite joints. The degradation can be assessed by constitutive and fracture tests. Constitutive tests on adhesives

show a significant lowering of adhesive strength and stiffness, often accompanied by an increase in ductility with increasing moisture content [51].

Parker studied the effect of environmental effects on carbon fibre reinforced plastic (CFRP) materials bonded with epoxy adhesives [52-54]. The studies which investigated the effect of pre-bond moisture on the strength of bonded joints by exposing dried laminates to a humid environment before bonding have shown that moisture contained within the composite, prior to bonding, can exert a serious deleterious effect on joint performance. However, drying of the composite prior to bonding has been shown to alleviate these deleterious effects and those drying procedures should form part of any pre-bond surface treatment regime. Storing a laminate for long periods of time was also seen to result in a decrease in the strength of the bonded joints, even when stored in a freezer in a sealed bag [52]. Whether the laminate had been dried or not before bagging had no effect. This was attributed to absorption of moisture during the storage period and indicates that laminates should be dried before bonding if they have been stored for any length of time.

Many studies for durability predictions for composite bonded joints were performed. One approach which has been extensively used to predict the durability of adhesively bonded joints exposed to humid environments is the cohesive zone model (CZM) modelling, methodology discussed in more detail in section 8.3 [55-59].

Recently, Hua et al. [60] proposed a strain-based failure model to deal with progressive cohesive failure in ductile aluminum and composite single-lap adhesive bonded joints studied for a range of environmental degradations. This model can predict not only the failure loads of the joints but also the damage initiation and propagation in the degraded adhesives. The elastic-plastic response of the adhesive and the substrates, both obtained from the bulk tensile tests, were incorporated in the coupled mechanical-diffusion analyses. The main limitation of this strain-based model is the mesh dependence of the material failure parameters. They proposed a displacement based continuum damage model [61] in order to overcome this mesh dependence, validated by undertaking progressive damage modelling on single-lap joints. They found good correlation between predicted and experimental results, demonstrating that the continuum damage model was an efficient and reliable method to model environmental degradation in ductile adhesive bonded joints, where failure is predominantly within the adhesive layer.

Earl et al. [62] studied experimentally, using a thermoelastic stress analysis, marine sandwich construction composite T-joints which were exposed to a hygrothermal (heat/moisture) ageing environment and were simultaneously subjected to one of three loading conditions: free

standing, static compression and cyclic compression. Data obtained from the aged joints was compared with data from an unaged specimen and significant differences were found that could only be attributed to the ageing process. They concluded that thermoelastic stress analysis can be used as a tool for establishing damage severity levels resulting from mechanical loading during hygrothermal exposure.

Various studies were employed on the effects of various environments on some adhesive properties, but it is still necessary to address the performance of specific adherend-adhesive combinations and to combine environmental, fatigue, and fracture studies of bonded systems. For example, it is known that moisture absorption results in varying degrees of plasticization, strength loss, and increased ductility of some epoxy adhesives. However, the effect of moisture on the fatigue and fracture properties of bonded joints employing these adhesives is still not fully understood. In addition, since adhesive joints are systems comprised of adherends, adhesives, and inter-phase regions, the performance of each of these components may strongly affect the performance of the joint. Thus, general knowledge of the behavior of adhesives exposed to various environments must be supplemented by knowledge of the behavior of specific bonded systems. Also, the influence of environmental aspects has specific relevance for multi-material structures (sandwich structures), where components with different reactions to the same environmental conditions can significantly alter the behaviour of the structure as a whole.

6. Failure modes

Failure modes are determined by the quality of the bond at each interface, specimen geometry, and loading. The modes of failure must be characterized in order to gain a full understanding of the properties of the adhesive and the joint being investigated. In FRP composite adhesive joints, according to the standard ASTM D5573 [63], there are seven typical characterized modes of failure. These failure modes are: adhesive failure, cohesive failure, thin-layer cohesive failure, fiber-tear failure, light-fiber-tear failure, stock-break failure or mixed failure. These modes are illustrated in Figure 9.

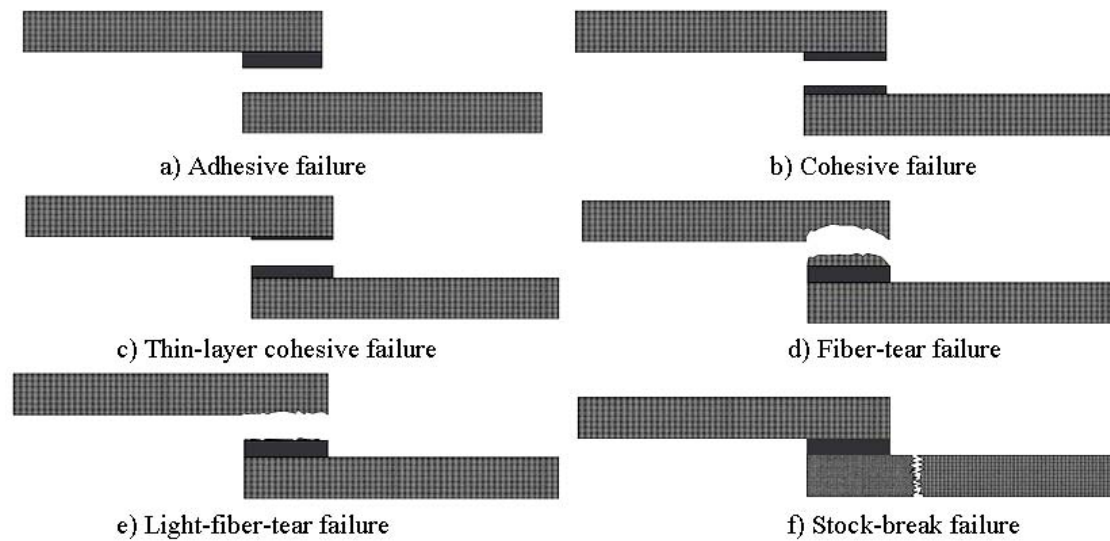


Figure 9. Possible failure modes in bonded joints between FRP composite adherends.

Many researchers experimentally investigated the influences of various parameters on the failure behaviours on composite bonded joints [64-75]. In these studies, the typical bonding parameters are surface conditions (e.g., contamination, abrasion, and plasma treatment), fillet, bondline thickness, surface ply angle, stacking sequence, environmental conditions, etc. Also, numerous studies for failure predictions for composite bonded joints were performed [70-77]. However, the failure prediction of the composite bonded joints is still difficult because the failure strength and mode are different according to various bonding methods and parameters. Primary failure modes for composite sandwich structures are buckling, local delamination, and fatigue/fracture. Buckling is one of the most important failure modes for composite structures, which have low modulus of elasticity. For all kinds of loading on skins and joints between composite materials, local delamination is one of the most severe failure modes since it can result in catastrophic failure for the global system structure. Interlaminar shear strength and through-thickness normal strength must then be carefully designed to prevent composite structures from local delamination. The initiation of the various failure modes depends on the material properties of the constituents (facings, adhesive and core), geometric dimensions and type of loading.

There are various studies referred to failure mode of sandwich structures bonded joints. Sheno et al. [37] used finite element analysis and experimentation to investigate the failure mechanisms in composite T-joints. Significant results of this study indicate that the behavior and failure modes of T-joints are dependent upon geometry and materials.

Interfacial debonding and shear failure of the core in the flange, for pullout loading of a T-joint with filler fillet and overlaminates, were the two major failure modes found by Theotokoglou and Moan [38]. They reported that the ultimate failure loads for these failure modes did not vary significantly.

Toftegaard and Lystrup [46] investigated theoretically and experimentally the failure mode in sandwich panels joined. They observed two different types of failures. One is the shear failure of the base panel, and the other is the failure through the T-joint itself. The shear failure is a classical shear failure of sandwich panels consisting of shear fracture of the core and delamination between the core and skin laminates. The two types of failure resemble the failure types found for pullout loading of a T-joint with filler fillet and relatively thick overlaminates [43].

Turaga and Sun [78] investigated various T-joints of composite sandwich panels and identified the corresponding modes of failure at the joint. The modes of failure included debonding between the two sandwich components to be joined, debonding between the attachment and the sandwich panel, and cracking in the core of the sandwich. They studied a new type of T-joint incorporating an aluminum U-channel in the web sandwich which provided much improvement over the conventional circular fillet T-joint. They also studied two types of bolted joints to investigate the effectiveness of using bolts for suppressing debonding at the joint. It was shown that using bolts in a circular fillet joint could cause early failure in the core and would not help much to increase the ultimate joint strength. The experimental failure modes of the tested joints were explained with the help of finite element analysis.

7. Analysis of adhesively bonded joints: Analytical methods

A simple shear lag model for adhesively bonded lap joints with the assumption that the adherends are in tension and the adhesive is in shear only and both stresses are constant across the thickness was presented for the first time, in 1938, by Volkersen [79]. However, the Volkersen solution does not reflect the effects of the adherend bending and shear deformations, which are potentially significant for composite adherends with a low shear and transverse modulus and strength. Goland and Reissner [80] extended this study by taking into consideration the effects of the adherend bending leading to peel stress in the adhesive layer, in addition to the shear stress. Goland and Reissner's adhesive shear and peel stress

distributions for aluminium alloy adherends and an epoxy adhesive can be seen in Figure 10 [81].

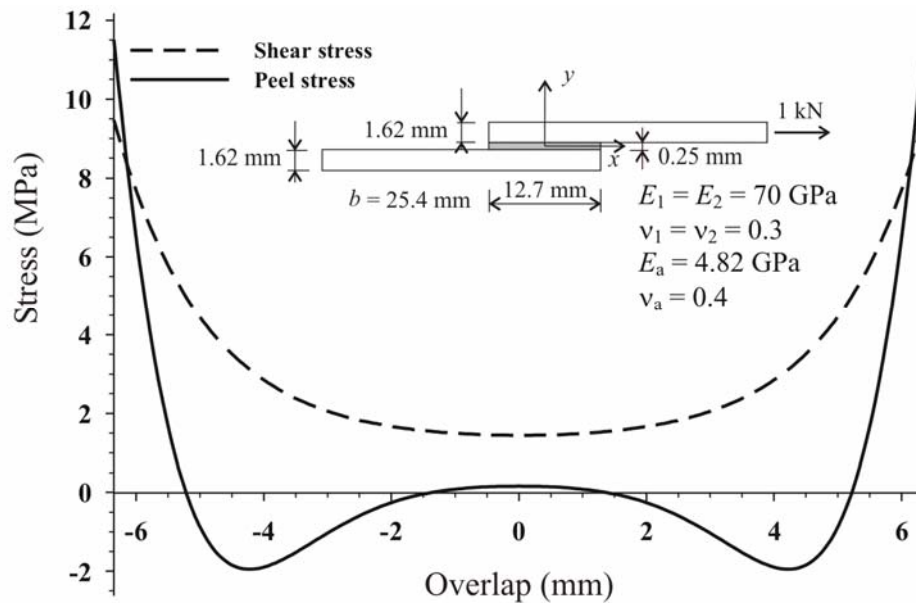


Figure 10. Goland and Reissner's adhesive shear and peel stress distributions for aluminium alloy adherends and an epoxy adhesive [81].

Hart-Smith [82] proposed a simple analytical model by considering that the adhesive layer has a perfect elastoplastic behavior. He could show that the maximum load which an adhesively bonded joint can transfer depends on the shear deformation energy of the adhesive layer, regardless of the stress-strain curve. This approach allows a better prediction of the mechanical behavior of ductile adhesive layers.

Hart-Smith analysed in detail single-lap, double-lap, scarf, and stepped-lap joints in which the adherends were isotropic or anisotropic elastic, and the adhesive was modelled as elastic, elastic-plastic, or bielastic. Thermal effects were also included in the analysis and it was shown that reduction in joint strength, due to thermal mismatch increases as adherends thickness and/or stiffness increases. A review of most of his work can be found in reference [83], and the main results are summarised there.

However, the effects of transverse shear deformation, which is important when adherends have relatively low transverse shear modulus, as in the case of laminated composite adherends were not included in the early classical analytical theories.

The low transverse stiffness that is often present as a result of the high and ultra-high modulus fibers combined with much lower-modulus polymer resins is an important characteristic of

adhesively bonded joints with composite adherends. Renton and Vinson [84] as well as Srinivas [19] accounted for these low transverse stiffness effects by including first-order shear deformation in their analysis. Renton and Vinson [84] developed an analytical solution of a single-lap joint geometry that included shear deformation for the composite adherends, and determined the linear elastic response for the adherends and adhesive. Srinivas [19] developed a similar method for single-lap and double-lap joints that included shear deformation as a part of the analytical solution while attempting to approximate the nonlinear geometric effects. Dattaguru et al. [85] as well as Pickett and Hollaway [86] provided studies that used nonlinear analysis methods to evaluate composite bonded joint configurations. They studied the performance of single-lap, double-lap, and tubular joints through analytical and finite-element models considering nonlinear material behavior in the form of elastic-perfectly plastic adhesive modelling. They used the elastic-plastic analysis method of Hart-Smith to perform their analytical joint evaluations. Additionally, the results from the elastic-plastic joint analyses predicted an increase in joint strength over the linear elastic results while the results from the geometric nonlinear analyses suggested that adhesive properties were a significant factor in the joint responses.

Renton and Vinson [87] have developed a higher-order analytical model that includes the adherend transverse shear and normal strains to analyze adhesive-bonded joints and predicted that the actual peak adhesive shear stress was shifted inboard from the edge of the joint and reduced when compared to the value found by traditional analysis methods. They did not include in their analysis coupling of the shear and bending responses of the joint, which has a significant effect on the stress distribution.

Delale et al. [88] extended the classical analysis of a bonded joint to include coupling between bending and extension, which resulted in modified constitutive equations for the adhesive that include the longitudinal stresses in the adhesive, derived from the average extensional strains in the adherends. Their results were shown to be accurate when compared to detailed finite element results for the adhesive stresses at the edge of a bonded joint. They concluded that it is not important to impose a stress-free boundary condition at the edge of the adhesive, since it is well known that the peak stresses occur at the singularity at the interface of the adherend and adhesive at the extreme edge of the joint.

Using an alternate approach, Allman [89] has expressed the stresses in the joint as a set of stress functions, and, by minimizing the strain energy in the joint, developed solutions that satisfied the adhesive stress-free boundary condition and allowed the satisfaction of the full equilibrium equations for the adherends.

Adams and Mallick [90] used Allman's approach to develop a one-dimensional finite element solution including the nonlinear adhesive behaviour. The adherends could be dissimilar with different material properties and/or different thicknesses. This model allows the variation of the adhesive stresses through the thickness. The results were found to be in close agreement with a finite element model, but it generally under-predicts the peak stresses.

Yang and Pang [91,92] also derived analytical models using classical laminated plate theory with first-order shear deformation to analyze symmetric and asymmetric single-lap joints subjected to tensile and bending loading. Their approach correlated the asymmetry of the adherend laminates as well as the effects due to the transverse shear deformation. The results were found to be in very close agreement with a finite element model.

Wu et al. [93] proposed a set of several differential equations for the analysis of joint-edge loads in dissimilar adherends with different thicknesses and lengths. They were developed for the stress analysis of adhesive joints with fibre reinforced plastics adherends.

Tong [94] used a simplified one-dimensional model as well as a finite element model to predict the strength of adhesively-bonded double-lap composite joints. He showed that the failure loads predicted by the nonlinear model were in good agreement with the measured failure loads, while those given by the linear model were only about half of the measured failure loads.

Frostig et al. [95] developed a closed-form high-order theory, which was an extension of their previous work, [96], on the analysis of sandwich panels with a transversely flexible or stiff core, analyses which has the advantage of modelling the shear stress free condition at the ends of the overlap. The adhesive shear stress was considered constant through the adhesive thickness and the transverse normal stress (peel stress) in the adhesive was allowed to vary through its thickness. The joints consisted of two metallic or composite laminated adherends that were interconnected through equilibrium and compatibility requirements by a two-dimensional linear elastic adhesive layer. However, the shear deformation in the adherends was neglected, which is important when adherends with relatively low transverse shear and normal modulus are present, as the case of laminated composite adherends. Recently, das Neves et al. [97] extended the model of Frostig to include two adhesives along the overlap (mixed adhesive joint). The use of a flexible and ductile adhesive at the ends of the overlap (low temperature adhesive in Figure 11) decreases the peel stress [98] which is of paramount importance when working with composites.

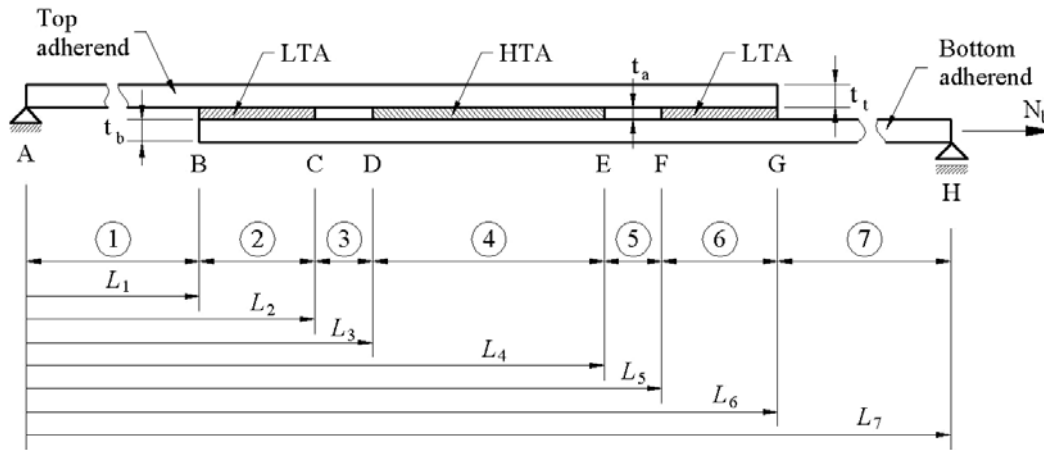


Figure 11. Geometry and loading of a typical mixed adhesive single lap joint. LTA stands for low temperature adhesive and HTA for high temperature adhesive [97].

Mortensen and Thomsen [99] developed a unified approach for adhesive composite bonded joints. They modelled the adherends as beams or wide plates in cylindrical bending and the composite adherends as generally orthotropic laminates using classical laminate theory and the adhesive as a linear elastic material. The analysis accounts for coupling effects induced by adherends made as asymmetric and unbalanced laminates and allow specification of any combination of boundary conditions and external loading. The results obtained using this approach were compared with finite element results and results obtained using a high-order theory approach (both including spew-fillets), and were in very good agreement.

Yang et al. [100] developed an analytical model using laminated anisotropic plate theory for determining adhesive stress distributions within adhesively bonded single-lap composite joints. The composite adherends were assumed linear elastic while the adhesive was assumed elastic-perfectly plastic following von Mises yield criterion. The adhesive was assumed to be very thin and the adhesive stresses were assumed constant through the bondline thickness. The entire coupled system of equations was determined through the kinematics relations and force equilibrium of the adhesive and the adherends. The overall system of governing equations was solved analytically with appropriate boundary conditions. The developed stress model was verified with finite element analysis by comparing the adhesive stress distributions.

Zou et al. [101] used classical laminate plate theory to model single lap and single strap metal and CFRP balanced and symmetric joints subjected to tensile and/or bending moment, as well as transverse shear loading. The model was verified by comparing with finite element models.

More recently, Zhang et al. [102] developed a method for multiaxial stress analysis of composite joints based on Mortensen and Thomsen [99] unified approach, to accommodate transverse in-plane strain and hygrothermal loads and to compute the in-plane and interlaminar stresses in the adherends. Compared with other analytical methods for bonded joint analysis, this method is capable of handling more general situations, including various joint geometries, linear and nonlinear adhesives, asymmetric and unbalanced laminates, and various loading and boundary conditions.

All the analyses presented considered the adherends to have linear elastic behaviour, some considered plastic behaviour only for the adhesive layer and some included variations in the distribution of adhesive stresses through the thickness of the adhesive [89,90,95]. However, material nonlinearity due to plastic behaviour is difficult to incorporate because the analysis becomes very complex in the mathematical formulation, the nonlinear material behaviour being implemented only in finite element models.

8. Analysis of adhesively bonded joints: Finite elements method

The finite element method has the great advantage that the stresses in a body of almost any geometrical shape under load can be determined. Linear and nonlinear finite element analyses have been carried out on different types of adhesive joints, and the adhesive effective stresses and strains have been evaluated. As the adhesive layer is thin compared with the thickness of the adherends, a fine mesh in these areas is required so the number of degrees of freedom in a joint is rather high. A full finite element analysis should include the effects of bending, adherend shear, end effects and nonlinear behaviour of the adhesive and adherends.

Numerous studies on the analysis of bonded joints with composite adherends have been published, many of them being concerned with the definition of appropriate failure criteria for improved joint strength predictions.

Adhesive bonded joints contain inherent defects from their manufacture. From these defects the crack initiation starts and leads later to failure of the assembly. The fracture strength of adhesive joints depends on a number of factors and their combinations-e.g. adhesive type, cure cycle, adherend type, bondline thickness, etc. However, a lack of reliable failure criteria still exist limiting in this way a more widespread application of adhesively bonded joints in structural applications. An accurate strength prediction of adhesively bonded joints is essential to decrease the amount of expensive testing at the design stage.

Currently used approaches for predicting the strength of adhesively bonded joints are: the continuum mechanics approach (stress based), fracture mechanics and damage mechanics approach.

8.1. Continuum mechanics approach

The continuum mechanics approach has been used by many researchers to predict the joint strength. The adhesive and adherends are modelled using continuum elements, assuming that the adhesive is perfectly bonded to the adherends. The assumption of a perfect bond means that the finite element analysis takes no account of the adhesion properties of the interface.

The criterion that is used varies between maximum values of stress [82, 103-105], strain [104] or plastic energy density [106]. However it is difficult to use maximum stress or strain criteria due to the bi-material singularities inherent in a bonded joint (see Figure 12). As was shown by Bogey [107] a bi-material wedge give rise to a singular strain distribution. As there will be always a singularity at the ends of idealised bonded joints, the maximum strain for such a model will coincide with the value at the singularity and thus will vary greatly with mesh refinement.

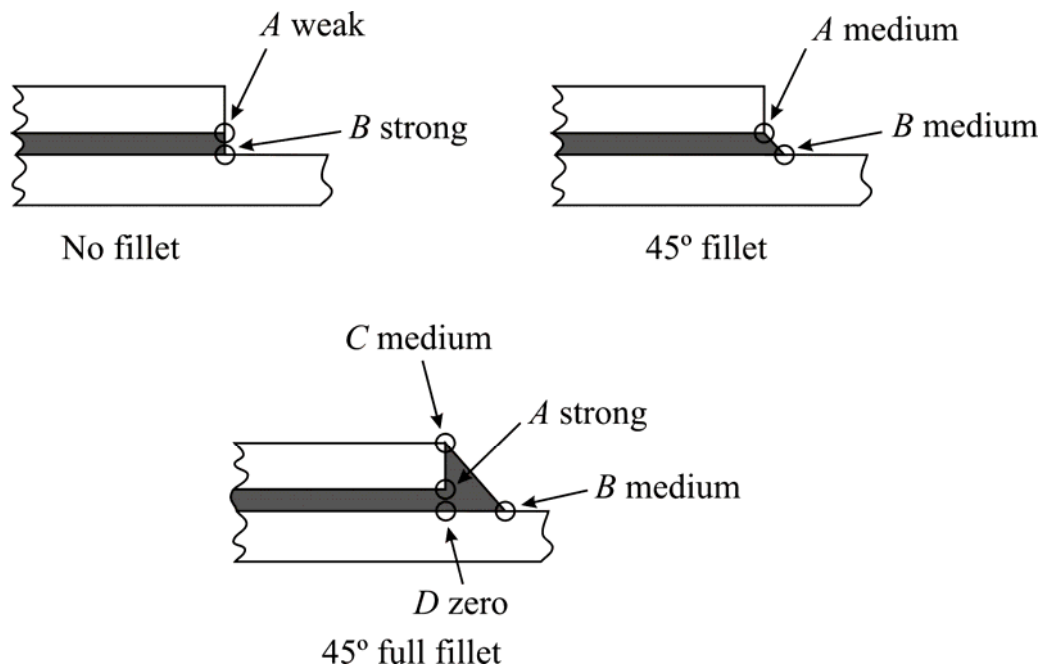


Figure 12. Strength of singularities in lap joints.

Adams and Harris [106] showed that by introducing rounding, the singularity was removed although the level of peak stress (or plastic strain energy density for the non-linear analyses) became dependent upon the degree of rounding. The problem now is shifted toward deciding

how much rounding to use in order not to affect the joint strength. Knowledge of the exact end shape becomes then essential for accurate strength predictions. Consequently, the continuum mechanics approach often uses the same operators as mentioned above, but this time applied at a certain distance from the singularity. This method is known as stress or strain at a distance [108].

8.2. Fracture mechanics approach

In the fracture mechanics approach, an energy parameter (toughness) is used as the failure criterion. In principle, it is possible to determine values of toughness as a function of the relative amounts of normal and shear deformation acting at the crack tip (in the adhesive, along the interface, or through the adherend), and to use the concepts of mixed-mode fracture mechanics (failure occurs if local mixed mode energy release rate exceeds a critical value) to predict the appropriate crack path to calculate the strength of the joint under different loading conditions [109].

This is the case of linear elastic fracture mechanics, which relies on the existence of a crack and linear elasticity. However, well-fabricated joints may not have macroscopic defects large enough to be considered cracks. Also, with laminated structures, it is easy to induce large-scale plasticity in the adherends. These factors fundamentally limit the use of linear elastic fracture mechanics approach in practical applications and alternative approaches need to be sought.

Another approach is the stress singularity approach (the fracture mechanics approach with no initial crack). The use of a generalized stress-intensity factor, analogous to the stress-intensity factor in classical fracture mechanics, to predict fracture initiation for bonded joints was investigated by some researchers [110-112]. Groth [111] suggested a fracture initiation criterion at the interface corners for bonded structure. It was assumed that initiation of fracture occurs when the generalized stress-intensity factor reaches its critical value. Gleich et al. [112] carried out a similar study and calculated the singularity strength and intensity for a range of adhesive thicknesses. They found that the intensity increased with increasing adhesive thickness and observed that this would account for decreasing joint strength with increasing adhesive thickness, which is verified experimentally.

8.3. Damage modelling

The progressive damage modelling approach enables the complete response of structures up to the final point of failure to be modelled in a single analysis without the need for additional

post-processing of finite element analysis results. This is an emerging field and the techniques for modelling damage can be divided into either local or continuum approaches. In the continuum approach the damage is modelled over a finite region. The local approach, where the damage is confined to zero volume lines and surfaces in two and three dimensions respectively is often referred to as cohesive zone approach.

In a cohesive zone model (CZM) approach, used to model the progressive damage and failure, a pre-defined crack path has to be defined [113-115]. A CZM simulates the macroscopic damage along this path by specification of a traction-separation response between initially coincident nodes on either side of the pre-defined crack path. In most of the CZMs, the traction-separation relations for the interfaces are such that with increasing interfacial separation, the traction across the interface reaches a maximum (crack initiation), then decreases (softening) and finally crack propagates permitting a totally de-bond of the interfaces. The whole failure response and crack propagation can thus be simulated.

A CZM models the fracture process extending the concept of continuum mechanics by including a zone of discontinuity modelled by cohesive zones, thus using both strength and energy parameters to characterize the debonding process (see Figure 13).

The main difference in CZMs is in the shape of the traction-separation curves and the parameters that describe this shape. In general, the parameters that describe the cohesive law are: the area under the traction-separation curve (the toughness), and a characteristic strength (typically, the cohesive strength) or a characteristic displacement that represents the failure strain of the cohesive zone. General traction-separation laws for damage-softened composites and adhesives can be obtained from Ungsuwarungsri and Knauss [114].

Studies demonstrated that it is possible to determine the appropriate cohesive-zone parameters of an adhesive layer experimentally, and to incorporate them into numerical analyses that have excellent predictive capabilities [116-121]. For example, Sorensen and Jacobsen [116] determined the cohesive laws by a J -integral approach using a double cantilever beam (DCB) specimen. The end-opening displacement was measured with extensometers at the neutral axis on the beam at the initial crack tip. By differentiating the J -Integral (determined from the load) with respect to the end-opening displacement a meaningful cohesive law was obtained.

Li et al. [117,118] used a cohesive-zone approach to model the mode-I fracture of adhesive composite joints. They have shown that a two parameter CZM parameter can be obtained for a polymer matrix composite joint from the experimental results.

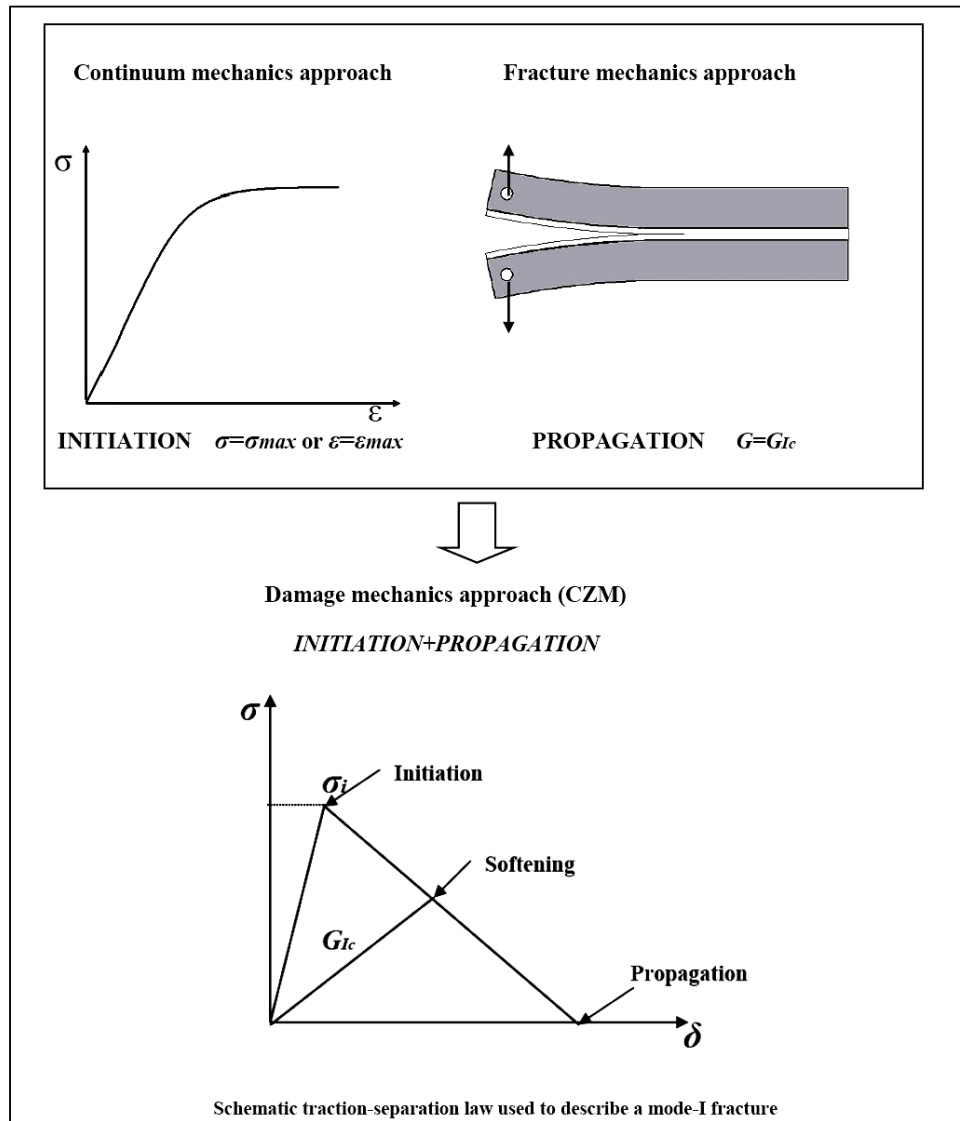


Figure 13. Schematic of a CZM for failure prediction of adhesively bonded joints.

The toughness parameters were obtained by correlation of the part of the load–displacement curve that was relatively independent of the cohesive strength parameter and the cohesive strength parameter was varied until the whole simulated load–displacement characteristics were consistent with the experimental results. A simulation of a DCB test and the experimental results were in excellent agreement. However, for specimens with a very short-characteristic length (butt joint) a third parameter had to be included in traction-separation law, corresponding to fibre bridging and pull out.

In subsequent works Li et al. [119,120] used the CZM approach to model the mixed-mode fracture of adhesive composite joints using different mixed-mode bonded geometries. The mode-II cohesive-zone parameters were obtained using sandwich end-notch flexure specimens [119] and single-lap shear and asymmetrical double-cantilever beam specimens

[120]. They showed that numerical simulations provided quantitative predictions, including predictions for both the strengths of the joints and for the failure mechanisms.

Liljedahl et al. [121] used the CZM in conjunction with both elastic and elastoplastic continuum behaviour to predict the response of a mixed mode flexure and three different lap shear joints. Excellent correlation was found between the experimental and predicted joint strengths.

Blackman et al. [122] applied CZMs to a range of bonded composite configurations and investigated the physical significance of the maximum (tripping) stress ($\sigma_{\max}$) used in the CZM. They concluded that it cannot be stated whether or not the traction stress in the separation law has a physical meaning.

Parametric studies, in order to determine the effect of incorporating plasticity with a CZM, were employed by many researchers [115,123-126]. They showed that the total amount of fracture energy is increased by including plastic dissipation. It was shown that slow hardening of the plastically deforming layer increases the steady state fracture toughness more than a sudden, step hardening [115,126]. It was also showed that the total toughness depend on the thickness of the adhesive layer. The ratio of the substrate elastic modulus to that of the adhesive layer was shown to have a fairly significant effect such that joints with stiffer substrates exhibit higher toughness [124,127].

An important concept of the CZM approach is that both strength and energy parameters are used to characterize the debonding process along the crack path, allowing the approach to be of much more general utility than conventional fracture mechanics. However, the cohesive models present a limitation, as it is necessary to know a priori the critical zones where damage is prone to occur and place the cohesive elements in accordance. Continuum damage models constitute a valuable alternative when crack path is not known a priori. Also, continuum damage models acquire special relevancy when adhesive thickness has to be considered [128]. de Moura et al. [128] used two different methods to simulate damage propagation to fracture characterization of bonded joints in pure modes I and II. The cohesive damage model was based on a trapezoidal law to account for the ductile behaviour of the adhesive and was applied to DCB and ENF tests. The cohesive parameters were determined using an inverse method applied to fracture characterization tests. The simulation of single-lap bonded joints indicates that only one parameter should be accurately determined, which simplifies the procedure. A continuum mixed-mode damage model was also developed to evaluate the adhesive thickness mechanical influence on fracture energy. It was verified that plateau values of the *R*-curves, which correspond to the fracture energy, presented similar values for

different adhesive thicknesses which means that there is no mechanical reason for the referred variation. The model was also able to simulate the different shape of the Fracture Process Zone as a function of adhesive thickness and its influence on the *R*-curve profile.

In summary, there are a large number of approaches and studies in the literature for predicting the strength of adhesively bonded joints and the number of possibilities in choosing a failure criterion is overwhelming. It is difficult to assess which one is the best and there is no general agreement about the method that should be used to predict failure as the failure strength and mode are different according to various bonding methods and parameters. However, progressive damage models are quite promising since important aspects of the joint behaviour can be modelled using this approach.

For adhesively bonded sandwich structures, much of the literature deals with design and analysis of T-joint sandwich structures. Linear finite element analyses of composite sandwich T-joints were developed by Sheno and Violette [40], Dodkins [42] and Phillips and Sheno [44]. Non-linear analyses are provided in [38,39,41,43] and may include damage in the form of delamination [42] or cracks [44].

Theotokoglou [38,39,43] developed a non-linear finite element model to predict joint performance of composite sandwich T-joints under pull-off load. The behavior of the sandwich T-joint in tension was shown to be characterized by geometric non-linearities due to out-of-plane loads and by material non-linearities due to local yielding.

Dulieu-Barton et al. [45] determined the stresses in sandwich construction T-joints experimentally and numerically under static loading. They showed that the features of interest in the T-joint are the use of a boundary angle laminate which reinforces the connection between the two component parts of the T-joint, the use of a fillet at the connection and the effect of any gap at the joint. The results of the finite element study of the T-joints were compared with the experimental data.

9. Conclusions

There is an increasing interest in the use of lightweight fiber reinforced composite structures for a variety of applications in different industries. Adhesive bonding is a viable technique for joining composite materials although the low interlaminar shear and tensile strength limit the joint efficiency.

Suitable surface treatments and adhesives for a given application have to be chosen. The choice of which adhesive is best is usually dictated by the type of composite to be bonded, the application, the service environment, and cost. Also, the behavior of specific bonded systems exposed to various environments should be taken into account for durability design. The choice of failure criterion will primarily be dictated by the type of joint considered and is dependent on the joint design philosophy.

An accurate composite bonded joint analysis method must be able to predict failure in the adhesive, at the adhesive-adherend interface, within the surface plies of the laminate itself and must also account for nonlinear material behavior.

There is no general agreement about the method that should be used to predict failure as the failure strength and mode are different according to various bonding methods and parameters, but progressive damage models are quite promising since important aspects of the joint behaviour can be modelled using this approach. However, a lack of reliable failure criteria still exists, limiting in this way a more widespread application of adhesively bonded joints in principal load bearing structural applications. An accurate strength prediction of the adhesively bonded joints is essential to decrease the amount of expensive testing at the design stage.

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Failure strength tests of high temperature adhesives for aerospace applications (AS1805 and RTV106)

Static and Fatigue Behaviour of Room Temperature Vulcanising Silicone Adhesives for High Temperature Aerospace Applications

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Abstract

This paper focuses on the mechanical properties of room temperature vulcanising (RTV) silicone adhesives for aerospace applications. In view of the very limited amount of published data on this subject, we analyzed the performances of two RTV silicone rubber adhesives through adhesive joint tests. The standard Thick Adherend Shear Test (TAST) was performed in order to measure the shear properties of the adhesives. Single lap joints (SLJs) were fabricated and tested to assess the adhesive performance in a joint. The influence of temperature on the lap-shear strength of the adhesives was investigated. It is shown that the lap-shear strength of both adhesives decreased with increasing of temperature. The effect of bondline thickness and overlap length on the lap-shear strength of the adhesives was also studied. The reduction of failure load with an increase of the bondline thickness is a very common situation when dealing with structural adhesives. However, for the tested RTV silicone adhesives the failure loads increased as the bondline was made thicker. Also, the failure loads of RTV silicone adhesive joints increased almost proportionally with increasing the overlap length. Fatigue tests were also performed and show a low dispersion of the results.

Keywords: RTV silicone adhesives; Temperature tests; Lap-shear strength; Fatigue

1. Introduction

Room temperature vulcanising (RTV) silicone adhesives are being increasingly used in a variety of space applications (i.e. satellites construction and space flight) as they are able to withstand the temperature extremes that are experienced in the space environment and are able to maintain a good degree of flexibility at very low temperatures. They are used when considerable expansion and contraction is expected in the joint and flexibility is required, as in the case of materials that have dissimilar coefficients of thermal expansion (CTE), since they can reliably compensate for the materials' different expansion properties. Actually, if considerable and rapid temperature changes produce thermomechanical stress within the adhesive, the flexible RTV silicone adhesives can dissipate this stress and keep it away from the interfaces.

RTV silicone rubber adhesives cure from the moisture in the air and form low strength structural joints. They retain excellent properties over a temperature range from nearly 300°C down to cryogenic temperatures, providing low-temperature flexibility and very high-temperature resistance. Also, they are used where organic materials (based on carbon) cannot withstand harsh environmental conditions. Silicone joints are designed to take advantage of the good peel strength of the silicone elastomer rather than its tensile or lap-shear properties [1]. However, the room temperature (RT) mechanical properties of RTV silicone adhesives are quite low compared to typical polymers. Moreover, silicones have not found broad use as adhesives, relatively to the total consumption volume, because of their relatively high cost.

The stress-strain characteristics of two moisture curing RTV elastomeric adhesives were investigated in tensile mode and in the lap-shear configuration by Geiss and Vogt [2]. They found that ageing in a humid atmosphere significantly influenced the mechanical properties of the adhesives. Banea and da Silva [3] studied the performances of a high temperature adhesive sealant (RTV silicone) through adhesive joint tests, concluding that the lap-shear RTV joint retains its flexibility and different fractions of strength at a temperature range from RT to 300°C. The pure mode I fracture toughness for adhesive joints bonded with a high temperature RTV silicone adhesive was also studied over a wide range of temperatures showing that the fracture toughness and the traction-separation laws exhibit temperature dependence [4]. However, very limited data is available relatively to the mechanical properties of RTV silicone adhesives.

It is well known that, in order to properly design a joint, the adhesive behaviour has to be characterized. Thus, to determine the stresses and strains in adhesive joints in a variety of configurations, it is necessary to know the adhesive mechanical properties. The thick adherend shear test (TAST) was performed in order to measure the shear properties of the adhesives according to standard ISO 11003-2:1993 [5]. In addition, SLJs were fabricated and tested to assess the adhesives performance in a joint. The influence of temperature on the lap-shear strength of the adhesives was also investigated.

The influence of temperature on the lap-shear strength of RTV silicone adhesives is an important factor to consider as they are designed for use in adhesive joints that undergo significant dimensional changes during their service life. For example, one of the applications of RTV silicone adhesives is to bond the ceramic tiles to the internal aluminium fuselage of the space shuttle. The most significant factors that determine the strength of an adhesive joint when used over a wide temperature range are the CTE (especially when compared to the CTE of the substrates) [6], and different adhesive mechanical properties with temperature [7-9]. Studies that report on experimental results of adhesive joints with structural adhesives (especially epoxies) as a function of temperature generally show a decrease in strength with increasing and decreasing temperatures [7,10,11]. At high temperatures the cause is the low adhesive strength while at low temperatures it is the high thermal stresses.

The effect of the bondline thickness on single lap joints is well documented in the literature. Most of the results are for typical structural adhesives and show that the lap joint strength decreases as the bondline increases [12-14]. There are many theories that attempt to explain this fact. Some researchers [14] explained that an increase in the bondline thickness increase the probability of having internal imperfections in the joint (voids and microcracks), which will lead to premature failure of the joints. Gleich *et al.* [15] showed with a finite element analysis on single lap joints that increases in the interface stresses (peel and shear) as the bondline gets thicker causes the failure load of a bonded joint to decrease. They found that for the low bondline thickness range an optimum distribution of stresses along the joint interface exists for maximum joint strength. Crocombe [16] shows that single lap joints with thicker bondlines have a lower strength by considering the plasticity of the adhesive. Grant *et al.* [17] found a reduction in joint strength with increasing the bondline thickness when testing SLJs for the automotive industry with an epoxy adhesive. The strength reduction was attributed to the higher bending moments for the lap joints with thick bondlines due to the increase in the

loading offset. However, it is known that small variations in the bondline thickness can result in significant changes in the bond strength and, for comparative studies, careful consideration should therefore be given to ensure that the stress and strain distributions (i.e. maximum peel and shear stresses at the ends of the joint) for different systems are at least similar [18]. For example, Bryant [19] tested SLJs with elastomeric flexible adhesives and concluded that it was the applied strain rate that was responsible for the performance of joints with different bondline thicknesses. Crocombe [16] suggested that this is not the case for modern structural adhesives, which are not so strain rate dependent. More recently, Giannis [20] tested two flexible sealants choosing the applied crosshead rate for each joint and concluded that the reduction in joint strength with increasing the bondline thickness cannot be explained by Bryant's suggestion.

Other researchers suggested that the adhesive/substrate interface properties may be modified as the bondline thickness increases. This may be due to internal stresses developing at this interface [21], to migration of species from the substrates into the adhesive (oxides) or to changes of stoichiometry within the adhesive near the substrate [22].

The overlap length is another parameter that can affect the joint strength. Studies showed that, in contrast to joints with brittle adhesives, the strength of joints with ductile adhesives increases almost proportionally with increasing overlap length [23,24].

The fatigue behaviour of adhesively-bonded joints is also important and is influenced by many factors, such as the adhesive type, the adherends, joint geometry, environmental conditions, loading and the quality of the joint fabrication process. For these reasons, fatigue tests can provide only comparative data and not design data. Although considerable research has been conducted to investigate the fatigue performance of adhesively-bonded joints [25,26], the fatigue behaviour has to be investigated for every particular adhesive/adherend system and applications. As is known, generally, the crack propagation resistance and fatigue resistance is greater with tough, flexible adhesives rather than with brittle adhesives. This is mainly due to more uniform stress distributions and high internal energy damping.

The objective of the present study was to investigate the mechanical behavior of two RTV silicone adhesives to further develop adhesive joints with these adhesives. The influence of temperature, adhesive thicknesses and the overlap length on the lap shear strength was studied. The fatigue behaviour of the tested RTV adhesives was also investigated.

2 Experimental details

2.1 Adhesives selected

Two adhesives were chosen for this study: AS1805, a high temperature thixotropic adhesive sealant, RTV silicone rubber supplied by ACC Silicones LDT (Bridgwater, UK) and RTV106, a 1-part high temperature acetoxy adhesive sealant supplied by GE Bayer Silicones (USA). The working temperature of AS1805 RTV adhesive is from -50°C to 300°C while for RTV106 it is from -60°C to 260°C.

A key parameter in the testing of adhesive joints is the glass transition temperature (T_g) of the adhesive. When the adhesively bonded joints are tested below this temperature, the adhesive will behave like a low-strain rigid material while above this temperature it will have a more rubber-like behaviour. For example, the elastomeric adhesives (the case of the RTV silicones studied here) have a T_g below 0°C and are in a rubbery state at room temperature, so that they have different behaviour in adhesively bonded joints when tested as a function of temperature. The glass transition temperature (T_g) of the AS1805 and RTV106 adhesive is -60°C (data provided by suppliers).

2.2 Specimens manufacture

2.2.1 TAST

For the TAST, steel substrates of dimensions 110 x 25 x 12 mm³ (see Fig. 1) were used. The joint surfaces were grit blasted and degreased with acetone prior to the application of the adhesive. The bondline thickness was nominally 0.7 mm and the length of the overlap test section was 5 mm. Two spacers (1.5 mm thick) were inserted in the gaps between the adherends after the application of the adhesive and prior to curing in order to provide the necessary spacing between the two adherends. These spacers were removed after the adhesive was cured. A mould with spacers for correct alignment of the specimens was used and is shown in Fig. 2. The adhesives were cured at RT for a week.

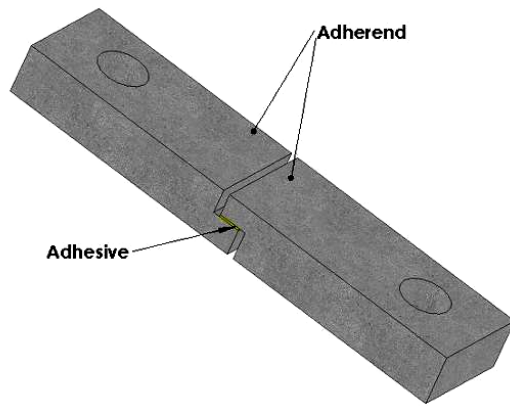


Fig. 1 Standard TAST specimen.

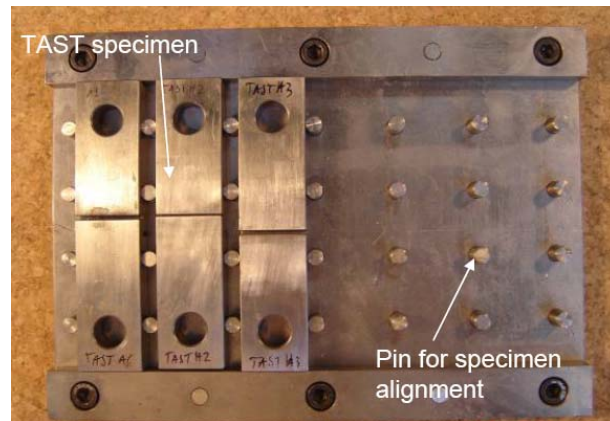


Fig. 2 Mould for TAST specimens fabrication.

Prior to testing, each specimen must be dimensioned for use in calculations and to assure conformity to the standards dimension set out in ISO 11003-2:1993 [5]. Measurements for each specimen were taken and recorded for the width, length, and thickness of the bondline.

2.2.2 Single lap joints

Aluminium alloy 6082-T651 (Al Si1MgMn) substrates with a thickness of 3 mm and 25 mm width were used for SLJs made with both adhesives. Overlap lengths of 12.5, 25 and 50 mm were tested. The values of bondline thickness selected for this study were 0.5 and 1 mm. In order to achieve that, packing shims of different thicknesses were used to provide the necessary spacing between the two adherend halves. The geometry of the lap shear joint specimens is shown in Fig. 3.

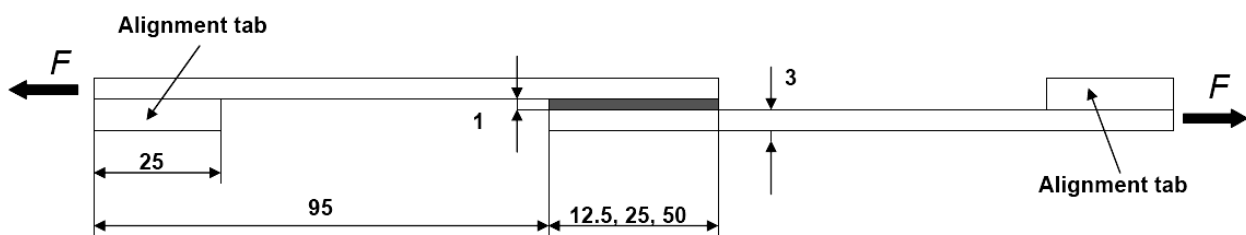


Fig. 3 SLJs specimen geometry (dimensions in mm).

The joint surfaces were grit blasted and degreased with acetone prior to the application of the adhesive. A mould with spacers for correct alignment of the substrates was used (see Fig. 4). The substrates were bonded and then the joints were left under pressure for 24 hours at RT in a hydraulic press. They were then removed from the mould and left for another 7 days to fully cure the adhesive, following the manufacturer's suggested curing conditions. At the end of the curing process, any excess of adhesive was carefully removed.



Fig. 4 Mould for SLJ specimens fabrication.

2.3 Test method

2.3.1 TAST

The TAST was performed at room temperature on a MTS servo-hydraulic machine MTS, (Eden Prairie, MN, USA), model 312.31, at a constant crosshead rate of 1 mm/min. The displacement was measured with a 25 mm length MTS extensometer (Eden Prairie, MN, USA). As the extensometer is mounted in the metallic substrate, the extensometer measures not only the displacement of the adhesive, but also the displacement of the adherend. However, da Silva *et al.* [27] showed that the steel deformation can be neglected in the case of flexible adhesives, so that the adhesives displacement can be measured by the MTS extensometer method. Five joints were tested for each adhesive.

2.3.2 Single lap joint tests

At RT, testing was conducted at a constant displacement rate of 1 mm/min using the MTS 312.31 servo-hydraulic machine. Loads and displacements up to failure were recorded.

At high temperatures, the SLJs were tested using a universal testing machine Instron model 8801 (Instron Co., USA), under a constant crosshead rate of 1 mm/min. For the high temperature tests the environmental chamber of the machine was used to reach the desired temperatures. Three joints were tested up to failure at each temperature. For each joint tested, load-displacement curves were produced.

2.3.3 Fatigue tests

The fatigue behaviour of the adhesives AS1805 RTV and RTV106 was investigated by testing SLJ specimens in force control with a sinusoidal waveform, load ratio (minimum to

maximum load) of $R = 0.1$ and frequency of 5 Hz. The specimen geometry (ISO 9664:1993 [28]) was a SLJ (Fig. 3) with a bondline thickness of 1 mm and an overlap of 25 mm.

The fatigue experiments were conducted at different load values from 30% up to 90% of the average quasi-static failure load of the SLJ specimens. The quasi-static failure load was calculated as the average of the maximum load sustained by three specimens tested at a displacement rate of 1 mm/s.

The servo-hydraulic MTS 810 testing machine was used. The tests were performed in ambient laboratory conditions. During testing, thermocouples were placed at various points on the surfaces of the specimens in order to investigate any thermal (heat up) effects. However, no change in temperature was observed. The fatigue experiments were conducted up to failure or to a maximum of 1 million load cycles.

3 Results and discussion

3.1 TAST

Typical shear stress-strain curves for the AS1805 RTV and RTV106 adhesives tested at RT are shown in Fig. 5. From the shear stress-strain curves, the shear modulus and shear strength were calculated. In general, elastomeric materials exhibit a nonlinear stress-strain behaviour and the definition of the modulus is very difficult. However, at small shear strains they obey Hooke's law and the modulus can be found. The values for shear modulus were calculated from the tangent to the shear stress-strain curve at the origin (a polynomial approximation of the curve was made).

The shear modulus, shear strength and strain data for the adhesives AS1805 RTV and RTV106 are presented in Table 1.

Typical failure modes of the TAST specimens are presented in Fig. 6. The failure was mainly cohesive. In some cases the failure was close to the interface but after close inspection, it was evident that a thin layer of adhesive remained on the substrate.

Table 1. Shear modulus and strength data of AS1805 and RTV106 adhesives obtained from the TAST.

Adhesive	Shear modulus G (MPa)	Shear strength (MPa)	Shear strain (%)
AS1805	0.68 ± 0.03	1.47 ± 0.02	332 ± 17
RTV106	0.55 ± 0.05	1.97 ± 0.03	408 ± 21

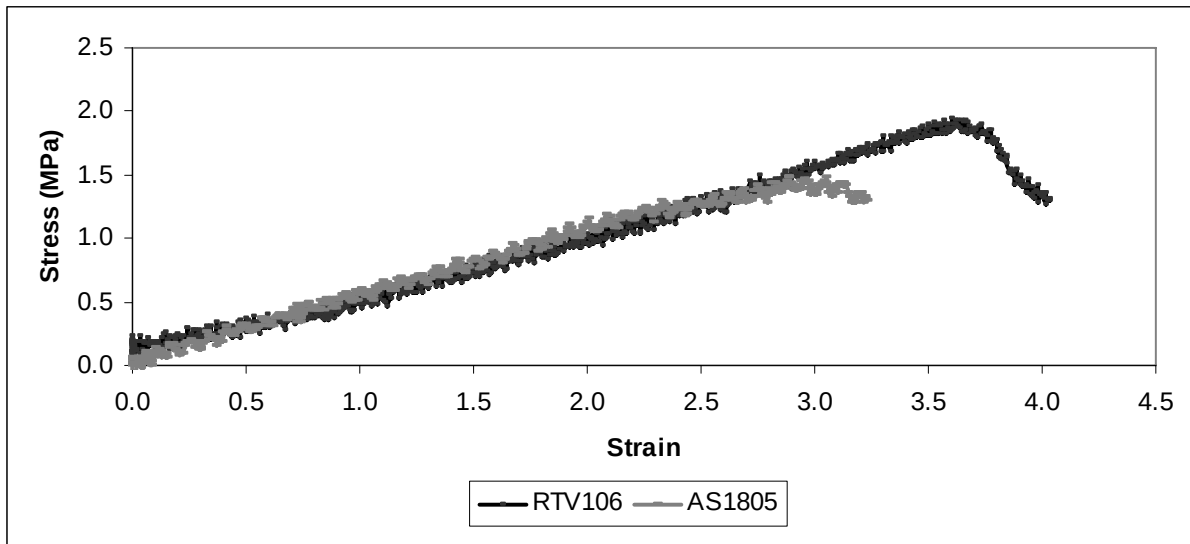


Fig. 5 Typical TAST shear stress-strain curves of AS1805 RTV and RTV106 adhesives tested at room temperature.



Fig. 6 Failure mode in TAST specimens: a) AS1805 RTV adhesive; b) RTV106 adhesive.

3.2 SLJ tests

3.2.1 Effect of temperature

AS1805 RTV adhesive

Representative load-displacement curves of AS1805 RTV adhesive SLJs (1 mm adhesive thickness) as a function of temperature are presented in Fig. 7. The failure displacement decreased with increasing temperature. The AS1805 SLJs stiffness does not substantially vary with temperature until 200°C.

The average lap-shear strength (τ_{av}) is given by:

$$\tau_{av} = P/bL \quad (1)$$

where P is the maximum load, b is the joint width and L is the joint overlap length.

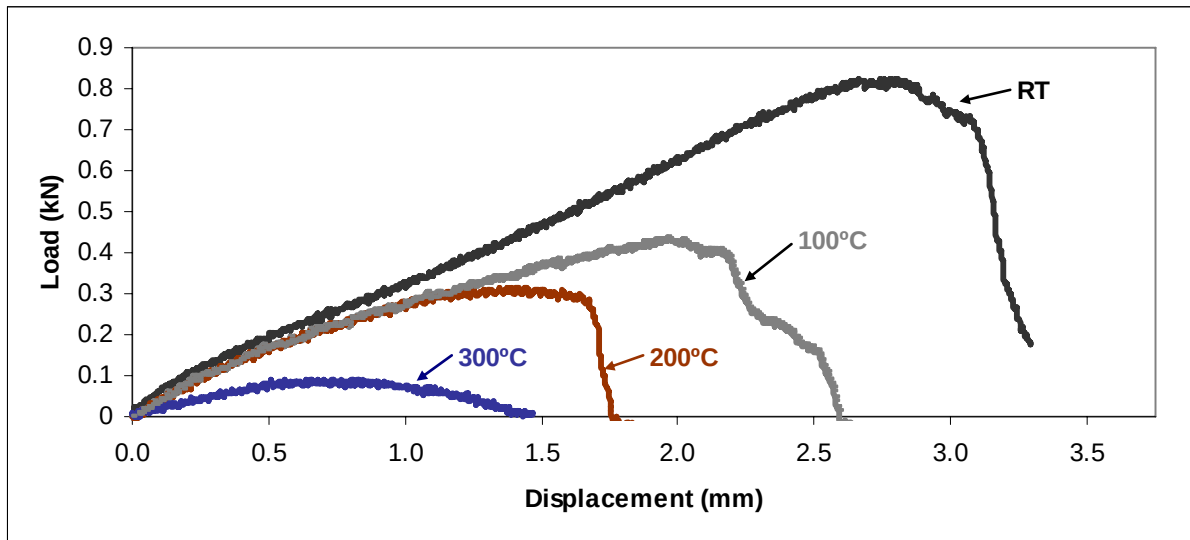


Fig. 7 Representative load-displacement curves of the AS1805 RTV SLJs as a function of temperature.

A summary of maximum loads and average lap shear strengths for AS1805 RTV adhesive SLJs tested at RT, 100, 200°C and 300°C is presented in Table 2.

Table 2. Maximum load and average lap shear strength for the AS1805 RTV SLJs as a function of temperature.

Temperature °C	Maximum load (N)	Average lap shear strength (MPa)
RT	782.8 ± 51.81	1.25 ± 0.08
100	413.7 ± 22.22	0.66 ± 0.03
200	264.4 ± 73.11	0.42 ± 0.11
300	103.3 ± 16.54	0.16 ± 0.02

The silicone systems can withstand exposure to temperatures of 200°C for a long period without degradation [29]. Also, the retention of flexibility and a fraction of strength at a temperature range from cryogenic to $> 260^{\circ}\text{C}$ is one of the advantages of silicone adhesives. Figure 7 and Table 2 show that the lap shear strength of the AS1805 RTV adhesive is affected by variation of temperature. Actually, the failure loads of the SLJs tested at 100°C fell to about half of the RT values. Also, the failure loads of joints tested at 200°C decreased by approximately 66%. After visual examination, the adhesive seems to not physically degrade at 300°C (maximum working temperature of the adhesive) but a dramatic fall in lap shear strength was observed.

RTV106 adhesive

Representative load-displacement curves of RTV106 adhesive SLJs (1 mm adhesive thickness) as a function of temperature are presented in Fig. 8. The failure displacement decreased with increasing temperature similarly to the AS1805 SLJs. It can also be seen that the RTV106 SLJs stiffness does not vary with the temperature.

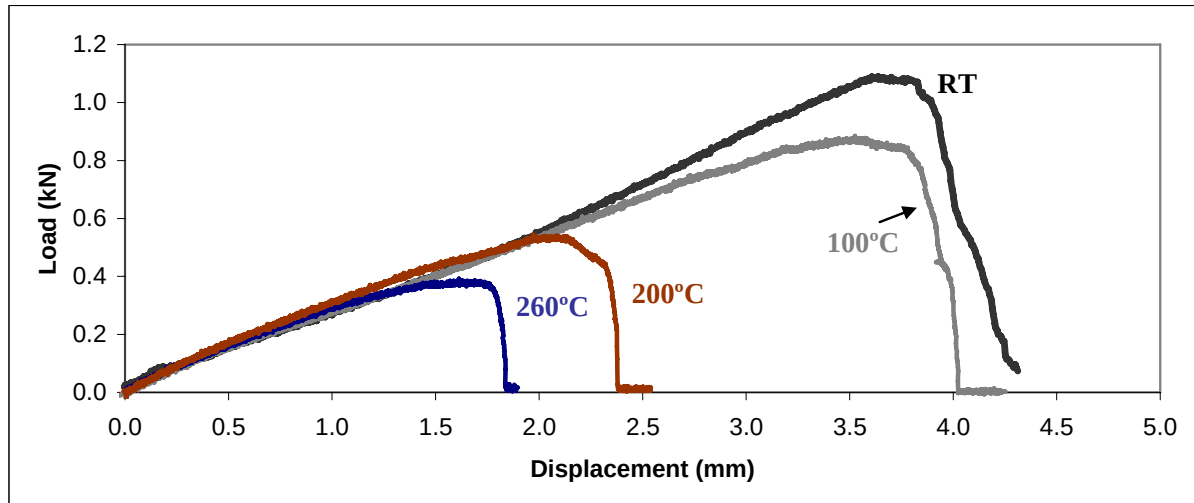


Fig. 8 Representative load-displacement curves of the RTV106 SLJs as a function of temperature.

A summary of maximum loads and average lap shear strengths for RTV106 adhesive SLJs tested at RT, 100, 200°C and 260°C is presented in Table 3.

The lap shear strength of the RTV106 adhesive decreases as the temperature is increased. The failure load of the adhesive joints tested at 100°C was reduced to about 17.4% of the RT failure load. Also, the failure load of joints tested at 200°C decreased by approximately 38%. At 260°C (maximum working temperature of the adhesive), a drop of approximately 66% in the lap shear strength was observed.

Table 3. Maximum load and average lap shear strength for the RTV106 SLJs as a function of temperature.

Temperature °C	Maximum load (N)	Average lap shear strength (MPa)
RT	1017.66 ± 88.79	1.64 ± 0.13
100	840 ± 41.72	1.35 ± 0.06
200	622.66 ± 68.19	1.00 ± 0.10
260	339.66 ± 48.23	0.54 ± 0.17

Similar to the adhesive AS1805, the lap shear strength of the RTV106 adhesive is affected by variation of temperature. However, the behaviour of RTV106 SLJs at high temperatures is considerably better than that of the AS1805 adhesive.

Failure modes

After the tests, the failure modes of the specimens were evaluated visually. A mixed cohesive/adhesive mode failure was found for the adhesive AS1805 RTV (Fig. 9). Ridges can be observed in the fracture surface, which are oriented perpendicularly to the loading direction. Pascal *et al.* [30] investigated the simple shear behaviour of a rubber-like adhesive. They observed noticeable ridges that appeared on the fracture surface of the specimens, perpendicularly to the loading direction. Using finite element analysis, they also found that the principal stresses acting in the rubber-like adhesive were practically oriented along the shearing direction. The appearance of the failure bond surfaces substantially varied with the temperature (Fig. 9). At 200°C, no sign of the ridges was detected on the failure surface. The failure surfaces at 200° and 300°C (Fig. 9b, c) suggest that the adhesive becomes less ductile at high temperature.

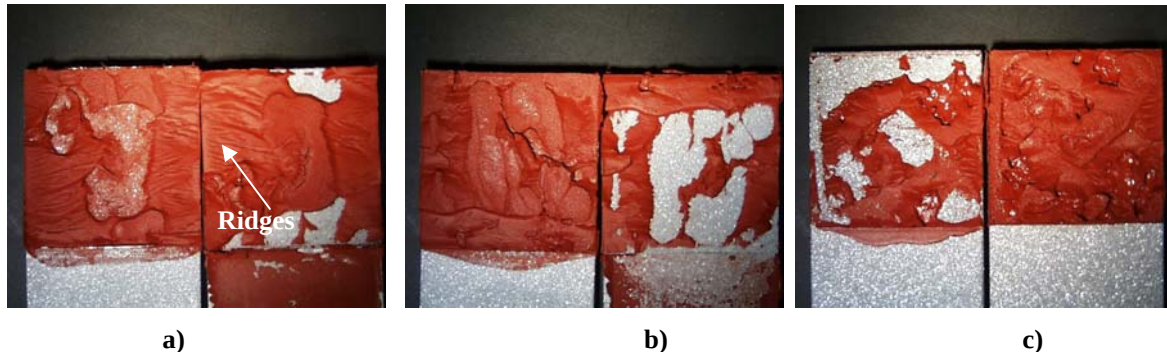


Fig. 9 Failure mode of the AS1805 RTV SLJs tested at a) 100°C, b) 200°C and c) 300°C.

The failure was cohesive for the RTV106 adhesive (Fig. 10). However, at RT the failure surface has some regions where the failure takes place close to the interface. Also, similarly to the adhesive AS1805, at high temperatures, a reduced amount of ridges can be observed in the fracture surfaces. The appearance of the failure surfaces for this adhesive also varied with temperature, as can be seen in Fig. 10.

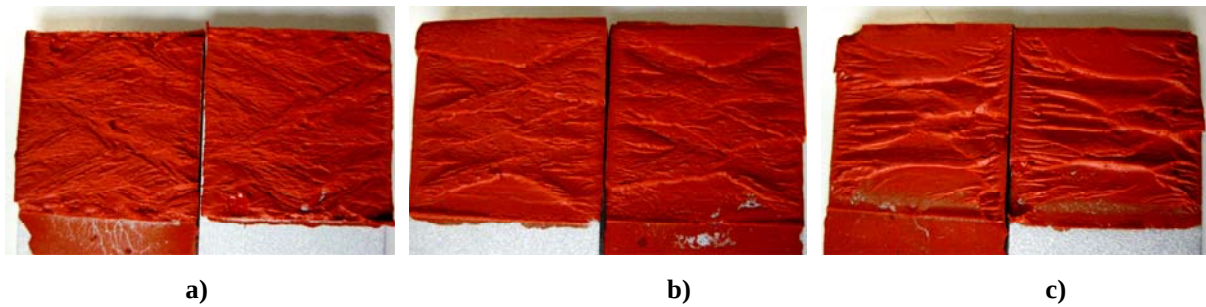


Fig. 10 Failure mode of the RTV106 SLJs tested at a) 100°C, b) 200°C and c) 260°C.

Microscopic examination

A scanning electron microscopy (SEM) analysis was performed on the fracture surfaces of RTV106 SLJs. Samples were coated with an Au layer prior to examination. The fracture surfaces of SLJs specimens tested at RT were examined to check whether or not a small layer of adhesive remains on the substrate surface where the failure takes place close to the interface. Figure 11 presents the SEM micrographs of a SLJ specimen. The SEM micrographs show that the failure took place inside of the adhesive layer, with a region where the failure was close to the interface, but it can be seen that a significant amount of adhesive remained on the substrate.

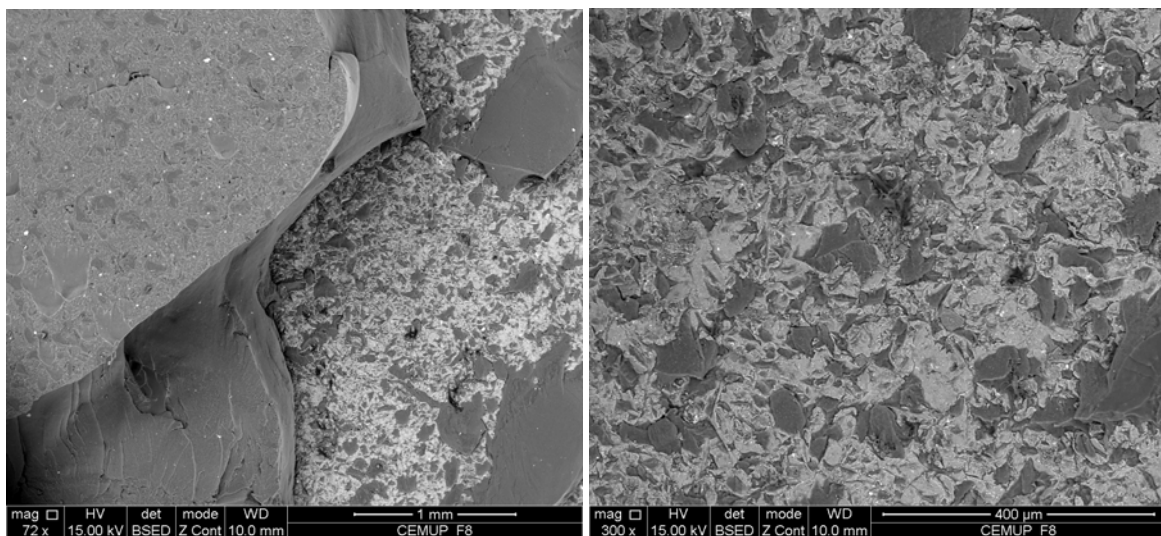


Fig. 11 SEM micrographs of the fracture surface of a SLJ RTV106 specimen tested at RT.

3.2.2 Effect of overlap length and bondline thickness

The effect of the overlap length on the joint performance was examined by testing SLJs with 12.5, 25 and 50 mm of overlap length. The behaviour of the RTV adhesives in SLJs with various bondline thicknesses was also of interest. It is known that for structural adhesives, smaller than expected bondline thicknesses may produce insufficient wetting, while thicker

than expected bonds may exhibit significant defects. Either case can lead to reduced joint performance. For RTV silicone adhesives, the optimum bondline thickness to be used in a joint, recommended by the manufacturer, is from 1 to 2 mm. However, it was decided to experiment values of bondline thickness of 0.5 mm and 1 mm. Figure 12 plots the average failure loads of SLJs with both RTV silicone adhesives as a function of the bondline thickness and overlap length. The failure loads of SLJs increased almost linearly with the overlap length. Figure 12 also shows that the average failure loads of the SLJs with the AS1805 RTV adhesive increases as the bondline gets thicker from 0.5 to 1 mm, although for joints with 12.5 and 50 mm overlaps this trend is not obvious due to the high experimental scatter. The same behaviour was observed for the RTV106 SLJs. The failure surface of the SLJs with 0.5 mm thickness presents a high area of adhesive failure (see Fig. 13a for AS1805 and Fig. 13c for RTV106, respectively). Thus, it can be concluded that for the SLJs with 0.5 mm thickness, the adhesion strength was smaller than the cohesive strength of the adhesive, which resulted in an adhesive failure. The failure mode, and subsequently the joint strength, changed when SLJs with 1 mm thickness were used. For this case, the failure took place cohesively with a very few spots of adhesive failure regions (Fig. 13b for AS1805 and Fig. 13d for RTV106, respectively). The explanation is probably not due to a joint mechanics argument as other flexible adhesive studied by the authors [31] with similar bulk properties gave an increased joint strength for thinner bondlines. The reason for this phenomenon might be due to different cure and interfacial chemical reactions as the RTV silicone adhesives thickness varies. Also, the adhesive structure may change as the thickness increases. This may be caused, for example, by differences in the curing conditions. Heterogeneous thermal behaviour, such as exotherm dissipation, will depend on the proximity of conductive substrates.

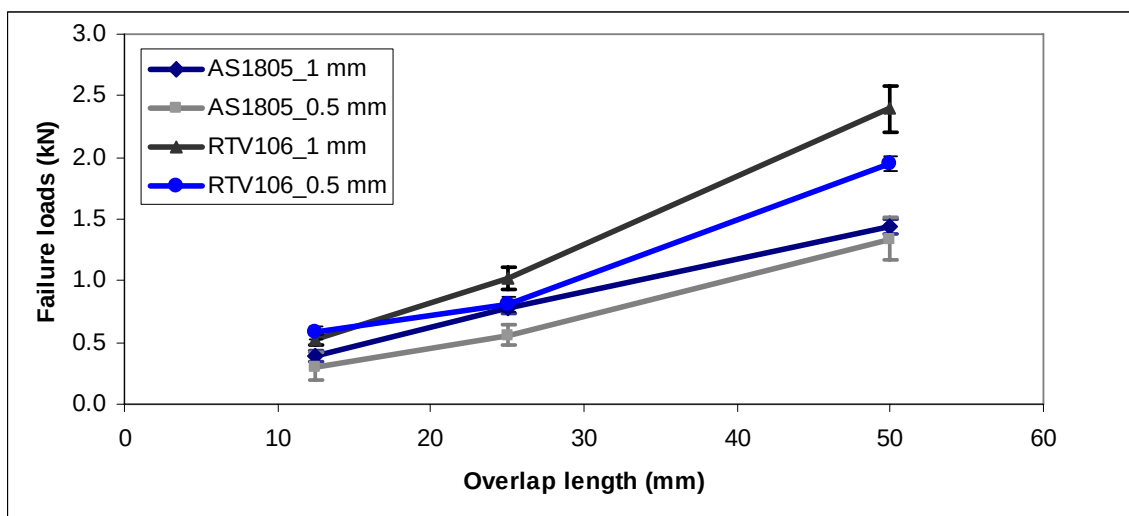


Fig. 12 Failure loads of the RTV SLJs with various bondline thicknesses and overlap length.

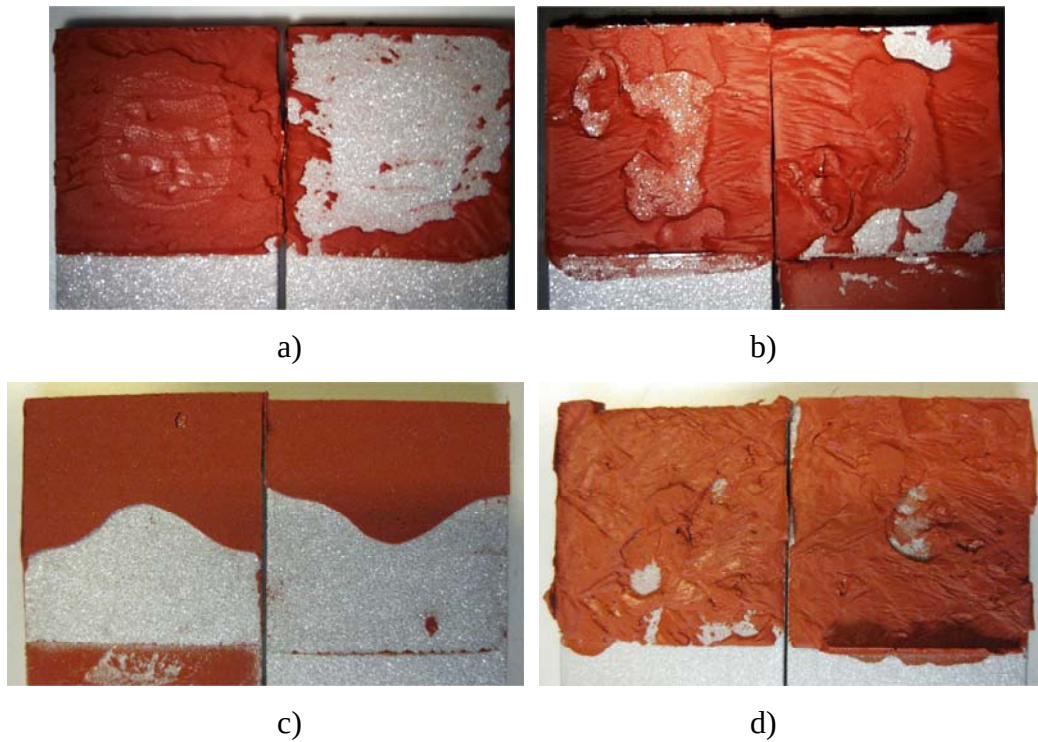


Fig. 13 Failure mode of the RTV SLJs (25 mm overlap) as a function of bondline thickness a) AS1805 0.5 mm; b) AS1805 1 mm, c) RTV106 0.5 mm; d) RTV106 1 mm.

Failure load prediction

As referred above, Pascal *et al.* [30] showed that flexible adhesives fail by tension, in the shear load direction, due to the very high adhesive deformation. However, this approach needs a numerical tool. In the present work, a simpler approach was used. The failure load of single lap joints can be predicted using the simple design methodology proposed by Adams *et al.* [32], based on the shear stress of the adhesive. The load corresponding to the total plastic deformation of the adhesive (global yielding) is given by:

$$P = \tau_y \times b \times L \quad (2)$$

where P is the failure load of the adhesive, τ_y is the shear yield strength of the adhesive, b is the joint width and L is the overlap length. Figure 14 shows the experimental and predicted failure loads of the single lap joints (with values of shear stress obtained from TAST). It can be seen that the simple criterion adopted for the SLJs gives failure loads that compare quite well with the experimental results.

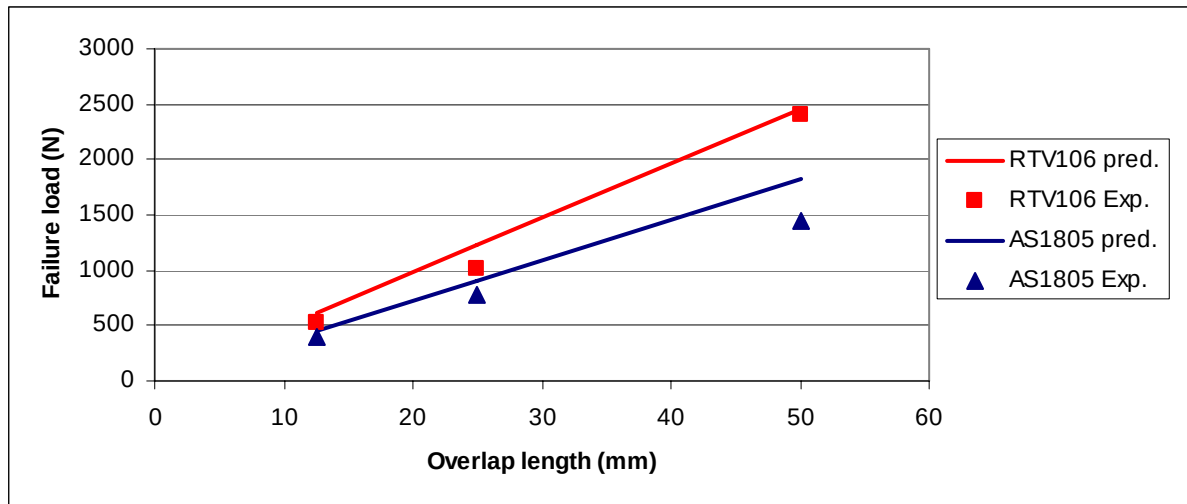


Fig. 14 Experimental and predicted failure loads of SLJs.

For the SLJs bonded with the RTV flexible adhesives tested in this work, the average lap shear stress for the SLJs at failure in quasi-static loading is very similar to the measured shear strength with the TAST (for RTV106 from TAST, $\tau_r = 1.97$ MPa and from SLJs (12.5 mm overlap length) $\tau_{av} = 1.64$ MPa, whereas for AS1805 from TAST, $\tau_r = 1.47$ MPa and from SLJs $\tau_{av} = 1.25$ MPa). This indicates that the shear stresses in the joints are essentially uniformly distributed whenever TAST or SLJ specimens are used. In other words, the SLJ can be used to determine the shear strength of flexible adhesives, contrarily to stiff and rigid adhesives like epoxies.

3.3 Fatigue tests

The parameter combinations used in the AS1805 and RTV106 adhesive fatigue experiments are presented in Table 4 and in Table 5 respectively.

For the AS1805 adhesive, specimens F1 to F9 were cyclically loaded up to failure. Experiments F10, F11 and F12 were stopped after 1.2×10^6 , 1.01×10^6 , and 3.1×10^6 cycles, respectively. The specimens that did not failed during the fatigue loading were subsequently statically loaded up to failure in the same fashion as the statically tested specimens. The average lap shear strengths were 0.99, 1.04, and 1.28 MPa, respectively. This is approximately 80%, 84% and 102.5% of the initial strength, indicating that very little damage had accumulated within the joint.

Table 4. Summary of the experimental fatigue program and results for SLJs with the AS1805 RTV adhesive.

Experiment/ Specimen	Load range F (N)	Amplitude ratio R	% of ultimate load	No. of cycles up to failure N_f
F01 – F03	0 - 782.8±51.81	-	100	1
F1	72 - 720	0.1	92	16
F2	55 - 550	0.1	70	973
F3	48-480	0.1	61	15175
F4	48-480	0.1	61	22500
F5	48-480	0.1	61	7197
F6	40 - 400	0.1	51	30235
F7	40 - 400	0.1	51	34549
F8	31 – 312	0.1	40	144915
F9	31 – 312	0.1	40	368415
F10	31 – 312 619 statically	0.1	40	1236002 Not failed
F11	24 – 240 652 statically	0.1	30	1012488 Not failed
F12	24 – 240 801 statically	0.1	30	3189037 Not failed

Table 5. Summary of the experimental fatigue program and results for SLJs with the RTV106 adhesive.

Experiment/ Specimen	Load range F (N)	Amplitude ratio R	% of ultimate load	No. of cycles up to failure N_f
F01 – F03	0 - 1017.66 ± 88.79	-	100	1
F1	81.4 - 814	0.1	80	3522
F2	81.4 - 814	0.1	80	14634
F3	61 – 610.5	0.1	60	16137
F4	61 – 610.5	0.1	60	32461
F5	61 – 610.5	0.1	60	16544
F6	50.8 - 508	0.1	50	86487
F7	40.7 - 407	0.1	40	299187
F8	40.7 - 407	0.1	40	1048268
F9	40.7 – 407 1062 statically	0.1	40	1367023 Not failed
F10	30.5 – 305 1086 statically	0.1	30	1237502 Not failed
F11	30.5 – 305 997 statically	0.1	30	1178092 Not failed

For the adhesive RTV106, specimens F1 to F8 were cyclically loaded up to failure. Experiments F9, F10 and F11 were stopped after 1.04×10^6 , 1.23×10^6 , and 1.17×10^6 cycles,

respectively. The specimens that did not failed during the fatigue loading were subsequently statically loaded up to failure in the same fashion as the statically tested specimens. The average lap shear strength was 1.73, 1.77, and 1.62 MPa. This is approximately 105%, 108% and 99.25% of the initial strength, indicating that very little damage had accumulated within the joint, similarly to the AS1805.

Figure 15 shows the resulting fatigue life curves of the SLJs adhesives AS1805 and RTV106 in the typical logarithmic representation. Fatigue data was normalised with respect to the average static failure load, F_0 . Load is used rather than stress amplitude because an average shear stress may be misleading, considering the non-uniform nature of the shear stresses and the existence of significant peel stresses, which most likely contributes to failure [33,34].

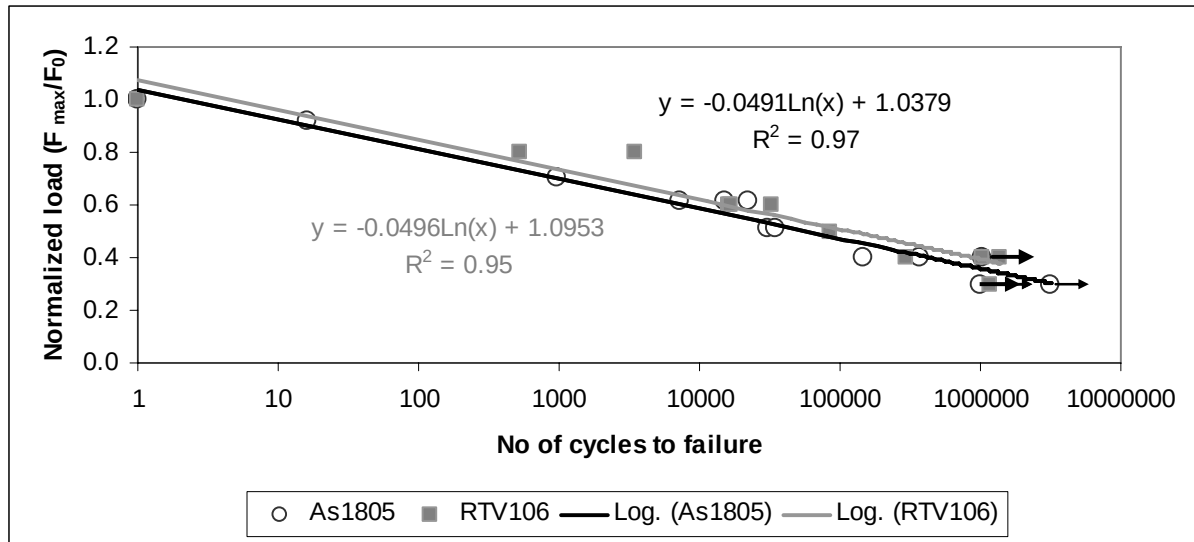


Fig. 15 Fatigue life curve of RTV adhesives/aluminium SLJs.

A straight line was fitted to the measured values (a logarithmic approximation was made). The equation of the fitted straight line for the adhesive AS1805 is

$$F_{max}/F_0 = 1.03 - 0.049 \ln(N_f), \quad (3)$$

while for RTV106 it is

$$F_{max}/F_0 = 1.09 - 0.049 \ln(N_f), \quad (4)$$

where F_{max} is the maximum load reached at regular intervals by the sinusoidal load applied to the specimen, F_0 is the average static failure load and N_f is the number of cycles up to failure. From the comparison of the fatigue results between the two tested adhesives, it can be seen that the slope of the fatigue curves of the adhesives AS1805 RTV and RTV106 SLJs are similar, indicating that the joints have the same fatigue behaviour.

The correlation coefficient of the linear least-squares fit is $R^2 = 0.97$ for AS1805, $R^2 = 0.95$ respectively for RTV106, which indicates a very good fit. This shows that there is a relatively little scatter in the results, indicating that the adhesive is not so sensitive to defects. Generally, the scatter associated with fatigue testing is large [35].

The experiments showed a fatigue limit at about 30 % of the static failure load at $R = 0.1$ for both adhesives (the fatigue limit was defined as the highest maximum load at which a specimen could survive 10^6 cycles with no visible apparent damage). Damage initiation and progression before failure could not be detected with the chosen measurement setup (measurement of loads and displacements).

The fracture surfaces were evaluated visually and the failure modes were identical to the failure modes of the static SLJ experiments (see Fig. 16).

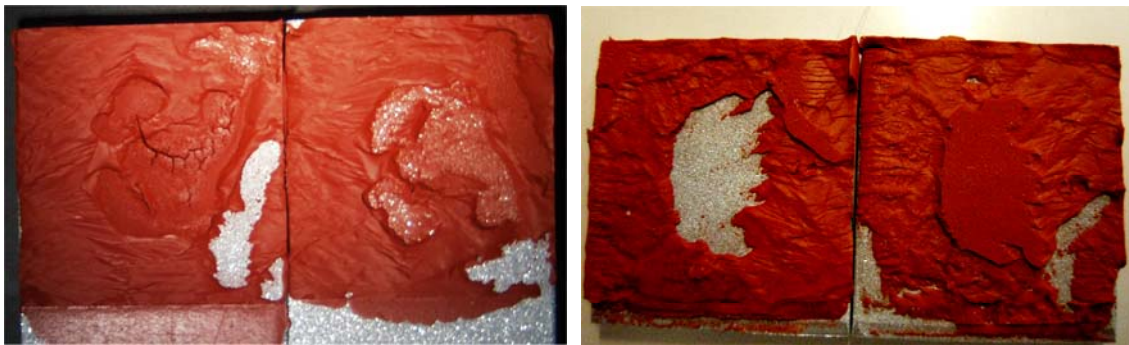


Fig. 16 Failure mode of fatigue: a) AS1805 SLJ specimen F7 (failure after 34549 cycles); b) RTV106 SLJ specimen F6 (failure after 86487 cycles).

4 Conclusions

In this paper, the performances of two different RTV silicone adhesives were investigated. The following conclusions can be drawn:

- The average lap shear stress in the joints at failure in quasi-static loading is very similar to the measured shear strength with the TAST (For RTV106 from TAST, $\tau_r = 1.97$ MPa and from SLJs $\tau_{av} = 1.64$ MPa, whereas for AS1805 from TAST, $\tau_r = 1.47$ MPa and from SLJs $\tau_{av} = 1.25$ MPa). This indicates that the shear stresses in the joints are essentially uniformly distributed whenever a TAST or SLJ specimens are used.
- SLJs of the RTV silicone adhesives were tested at different temperatures. The lap shear strength of the RTV silicone adhesives was affected by the temperature. However, the adhesive RTV106 showed a better behaviour at high temperatures than the adhesive AS1805.
- In contrast to SLJs with brittle adhesives, the strength of ductile joints with flexible adhesives increases almost proportionally with increasing the overlap length.
- The reduction of failure load with increasing the bondline thickness is very common when dealing with structural adhesives. For the low strength RTV silicone adhesives, the failure loads increased as the bondline gets thicker from 0.5 to 1 mm. This might be due to different cure and interfacial chemical reactions as the RTV silicone adhesives thickness varies.
- The fatigue tests on RTV silicone adhesives SLJs showed a fatigue limit of approximately 30% of the static failure load. The slope of the fatigue curves of the AS1805 RTV and RTV106 adhesive bonded joints are similar, indicating that the SLJs have the same fatigue behaviour.

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Failure strength tests of high temperature adhesives for aerospace applications (XN1244)

Effects of Temperature and Loading Rate on the Mechanical Properties of a High Temperature Epoxy Adhesive

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Abstract

The variation of the mechanical properties of adhesives with temperature and strain rate is one of the most important factors to consider when designing a bonded joint due to the polymeric nature of adhesives. It is well known that adhesive strength generally shows temperature dependence. Moreover, in many structural applications, the applied loads can be dynamic and the design of the joint requires the knowledge of the high loading rate mechanical behaviour of the adhesive.

In this study, the combined effect of the temperature and test speed on the tensile properties of a high temperature epoxy adhesive was investigated. Tensile tests were performed at three different test speeds and various temperatures (room temperature (RT) and high temperatures (100°C, 125°C and 150°C)). The glass transition temperature (T_g) of the epoxy adhesive investigated is approximately 155°C. The ultimate tensile stress linearly decreased with temperature (T) while logarithmically increased with the loading rate, which is in the accordance with the Airing's molecular activation model.

Keywords: High temperature adhesives, temperature tests, tensile strength, test speed.

1. Introduction

Adhesively bonded joints are an increasing alternative to mechanical joints in engineering applications and provide many advantages over conventional mechanical fasteners. Among these advantages are lower structural weight, lower fabrication cost, and improved damage tolerance. The influence of temperature and strain rate on the strength of the adhesive joints is an important factor to consider in the design of adhesive joints. The most significant factors that determine the strength of an adhesive joint when used over a wide temperature range are: the cure shrinkage [1], the coefficients of thermal expansion (CTE), (especially when compared to the CTE of the substrates) [2], and different adhesive mechanical properties with temperature [3-7]. However, due to the polymeric nature of adhesives, the variation of the mechanical properties of the adhesives with temperature is one of the most important factor to consider when designing a bonded joint.

Studies that present experimental results of adhesive joints with structural adhesives (especially epoxies) as a function of temperature generally show a decrease in strength with increasing and decreasing temperatures [3, 8-11]. At high temperatures the cause is the lower adhesive strength while at low temperatures the high thermal stresses and the brittleness of the adhesive are the origin of such behaviour.

On the other hand, in many structural applications, the applied loads can be dynamic and the design of the joints requires the knowledge of the high strain-rate mechanical behaviour of the adhesive [12-15].

In literature, most studies concern separately the influence of temperature and strain rate on the adhesives behaviour, while only a few concern the combine effects of temperature and strain rate [16,17]. For example, Richardson *et al.* [16] proposed and evaluated a multiaxial, time and temperature (MATT) model for an epoxy adhesive, which is based on an ellipsoidal failure envelope and is fairly accurate for temperatures between -29°C to 46°C for a wide range of normal and shear load combinations provided that the assembly is loaded at a constant rate. Park and Liechti [17] performed tensile and shear experiments on a urethane structural adhesive. They tested dry and salt-water saturated urethane specimens at four different strain rates and at four different temperature levels. They concluded that the stress-strain behaviour at large strains was rate-dependent and exhibited temperature and solvent dependency. Sharon *et al.* [18] studied the effects of loading rate and temperature on the mechanical properties of four structural adhesives containing a carrier. They found that the

yield stress and modulus decreased with temperature, while the loading rate presented a pronounced effect on the yield stress which increased with increasing loading rates, and had a negligible influence on the modulus. Recently, Carlberger *et al.* [19] investigated in an experimental study the effect of temperature and strain rate on the cohesive properties of an epoxy adhesive. They found that the fracture toughness is fairly unaffected by the temperature, from -40°C to $+80^{\circ}\text{C}$; (the glass transition temperature of the epoxy adhesive investigated is $+90^{\circ}\text{C}$) and the peak stress in peel loading decreased monotonically with increasing temperature in this temperature range, while both cohesive parameters increased with increasing the strain rate.

The effects of testing rate and temperature on the behaviour of adhesively bonded joints have been presented by several authors [8,20,21]. Deb *et al.* [8] investigated the effects of temperature and strain rate on the quasi-static response of double-lap steel joints bonded with an epoxy adhesive. At high temperature (82°C), the joints exhibited a greater degree of strain rate sensitivity with a significant decrease in joint strength compared to joints at ambient temperature. These differences were attributed to the softening of the adhesive at high temperatures. Srivastava [20] investigated the effect of temperature and strain rate on the bond strength of lap joints of Ti-6Al-4V and C/C-SiC composite bonded joints and reported that the bond strength decreases by 40–50% at 300°C , whereas it increases with strain rate.

In this study, the effects of the temperature and test speed on the tensile properties of a high temperature epoxy adhesive were investigated. Tensile tests were performed at three different test speeds (0.1, 1 and 10 mm/min) and various temperatures (RT and high temperatures (100°C , 125°C and 150°C)). An empirical relation based on the Airing's molecular activation model based on the experimental data with variables of temperature and test speed for the high temperature epoxy adhesive has been used. With the obtained models, the mechanical properties of adhesive joints can be predicted as a function of temperature and loading rate.

2. Experimental details

2.1. Adhesive

The adhesive investigated in this study was a one-component high temperature paste epoxy adhesive XN1244, supplied by Nagase ChemteX (Japan).

A key parameter in the testing of adhesive joints is the glass transition temperature (T_g) of the adhesive. When the adhesively bonded joints are tested below this temperature, the adhesive will behave like a low-strain rigid material while above this temperature it will have a more rubber-like behaviour.

The dynamical mechanical thermal analysis (DMTA) method was used to determine the T_g of the adhesive. The test results are plotted in Fig. 1 for three specimens (the dimensions of the T_g specimens were 40 mm x 10 mm, with a thickness of 4 mm, and the fabrication was in the same fashion as presented in section 2.2 for “dogbone” specimens). This graph contains the storage modulus - E' and the loss factor - $\tan \delta$ for each specimen. The glass transition temperature of the XN1244 adhesive, corresponding to the $\tan \delta$ peak, is approximately 155°C.

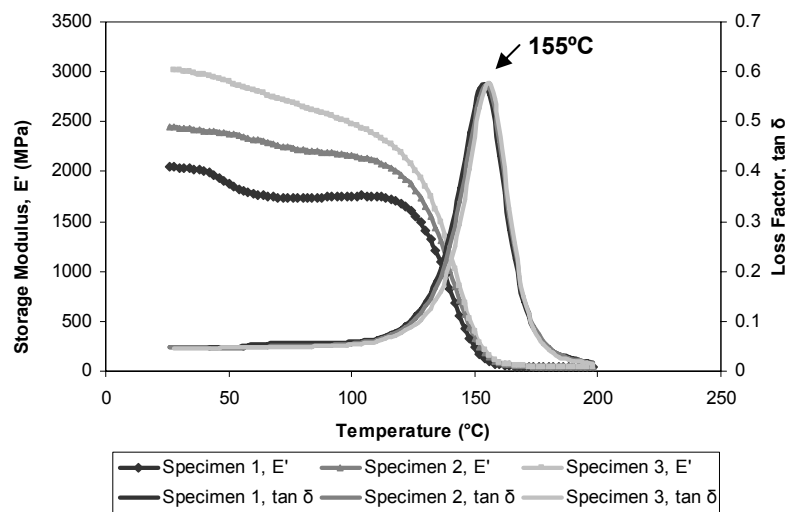


Fig. 1 – Dynamic mechanical analysis results for the epoxy adhesive XN1244.

2.2. Specimen fabrication

Thin sheets of XN1244 adhesive were produced by curing the adhesive between steel plates of a mould with a silicone rubber frame (Fig. 2) according to the French standard NF T 76-142 [6], which were hot pressed (2 MPa) for 60 minutes at 140°C. The silicone rubber frame stops the adhesive from flowing out and the pressure applied creates a good surface finish.

The dimensions of the adhesive plate after cure were, 150 mm x 45 mm, with a thickness of 2 mm, which corresponds to the internal dimensions of the silicone rubber frame. “Dogbone” specimens 2 mm thick and 10 mm wide were machined from the bulk sheet plates. The geometry of the “dogbone” tensile specimens used (BS 2782 standard) is shown in Fig. 3. “Dogbone” tensile specimens, before and after tests, can be seen in Fig. 4.

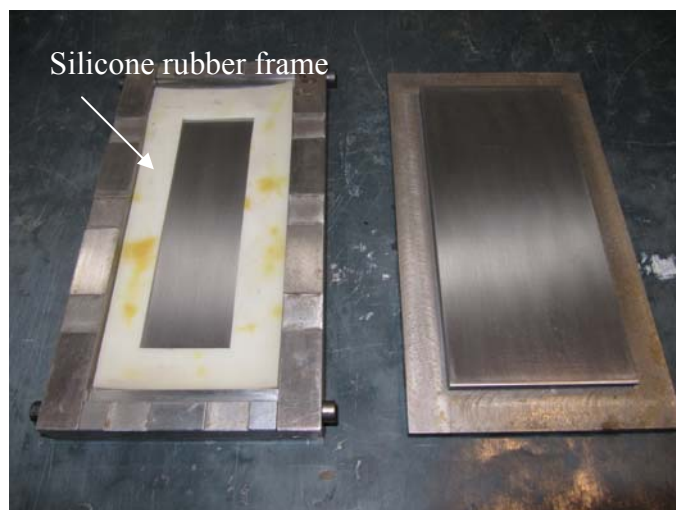


Fig. 2 – Mould for producing bulk specimens.

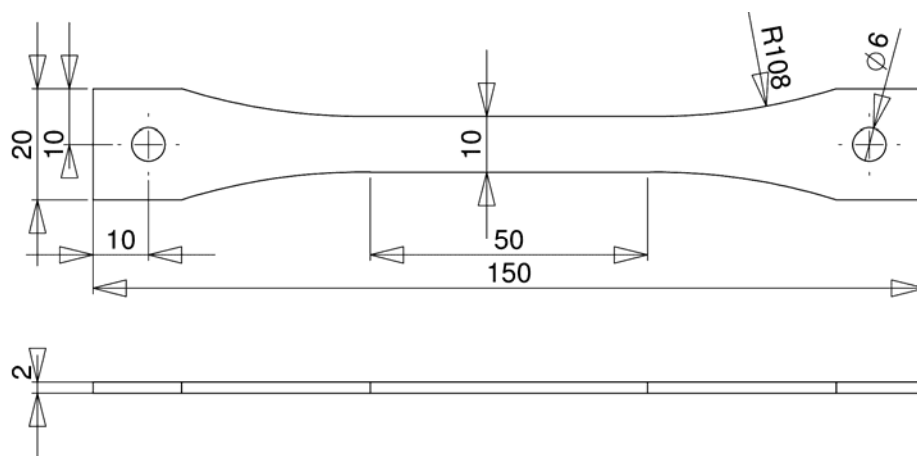


Fig. 3 – Tensile test specimen geometry bulk specimens (dimensions in mm).



Fig. 4 – Bulk specimens before and after tests.

2.3. Test procedure

2.3.1. Tensile tests

The XN1244 specimens were tested in tension using a universal testing machine Shimadzu[®] model AUTOGRAPH. A load cell of 5 kN was used. A video extensometer Messphysik ME46 was used to record the displacement. Two marks (50 mm gauge length) were painted on the specimens for the calibration and the strain measurements by the videoextensometer. For the high temperature tests, an environmental chamber of the machine was used to reach the desired temperature. At least three “dogbone” tensile specimens were tested to failure at three different constant crosshead rates of 0.1, 1 and 10 mm/min for each temperature (RT (23°C), 100°C, 125°C and 150°C).

A thermocouple was applied to the specimen in order to assure that the air temperature inside the chamber was equal to the specimen’s temperature. The tests were always performed after approximately 5 min of achieving the test temperature in the specimens, to ensure a steady-state temperature throughout the specimen prior to testing.

3. Experimental Results and Discussion

The XN1244 “dogbone” specimens were tested at three different test speeds for each temperature. For each specimen, an engineering tensile stress-strain curve was produced based on load, displacement values and specimen dimensions. From the stress-strain curve, the elastic modulus, ultimate tensile strength and maximum tensile strain were calculated.

Representative adhesive tensile stress-strain curves as a function of temperature and test speed can be seen in Fig. 5.

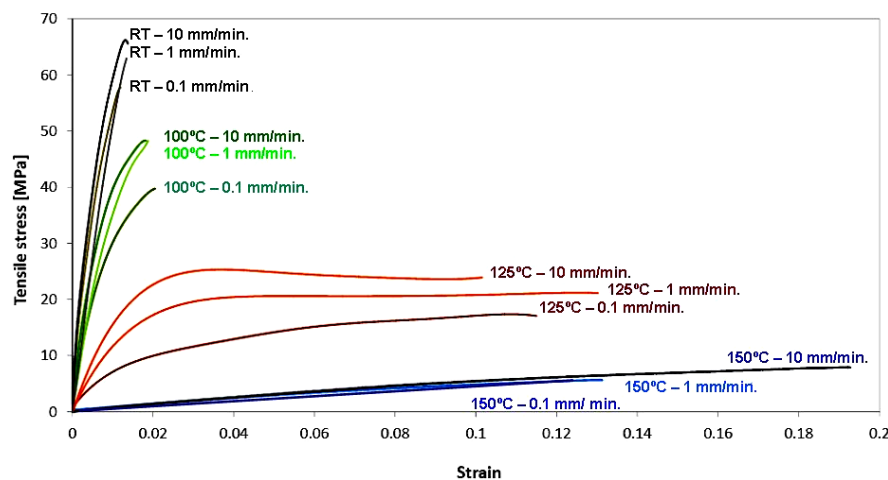


Fig. 5 – Representative XN1244 adhesive tensile stress-strain curves as a function of temperature and test speed.

As expected for polymeric materials, it can be seen that the ultimate tensile stresses were reduced at high temperatures and were higher at a high test speed than at a low test speed. At 125°C and 150°C, the stress-strain curves illustrate considerable softening and plastic type behaviour.

The average values and standard deviations of the tensile strength, Young' modulus and tensile strain to failure as a function of temperature, of the XN1244 adhesive at a test speed of 1 mm/min, 0.1 mm/min and 10 mm/min are presented in Table 1, Table 2 and Table 3, respectively.

Table 1. Tensile properties of the XN1244 adhesive at a test speed of 1 mm/min.

	Young's modulus, E [GPa]	Tensile strength, [MPa]	Tensile failure strain, [%]
RT	5.87 ± 0.33	68.23 ± 5.06	1.46 ± 0.23
100°C	4.46 ± 0.53	45.16 ± 3.44	1.93 ± 0.43
125°C	1.52 ± 0.17	21.55 ± 0.55	13.63 ± 0.29
150°C	0.07 ± 0.01	6.49 ± 0.86	13.71 ± 1.46

Table 2. Tensile properties of the XN1244 adhesive at a test speed of 0.1 mm/min.

	Young's modulus, E [GPa]	Tensile strength, [MPa]	Tensile failure strain, [%]
RT	4.70 ± 0.05	60.50 ± 3.5	1.53 ± 0.4
100°C	4.37 ± 0.03	37.71 ± 4.9	2.82 ± 0.5
125°C	1.21 ± 0.05	18.44 ± 1.5	12.63 ± 0.1
150°C	0.07 ± 0.01	5.25 ± 0.5	13.51 ± 0.9

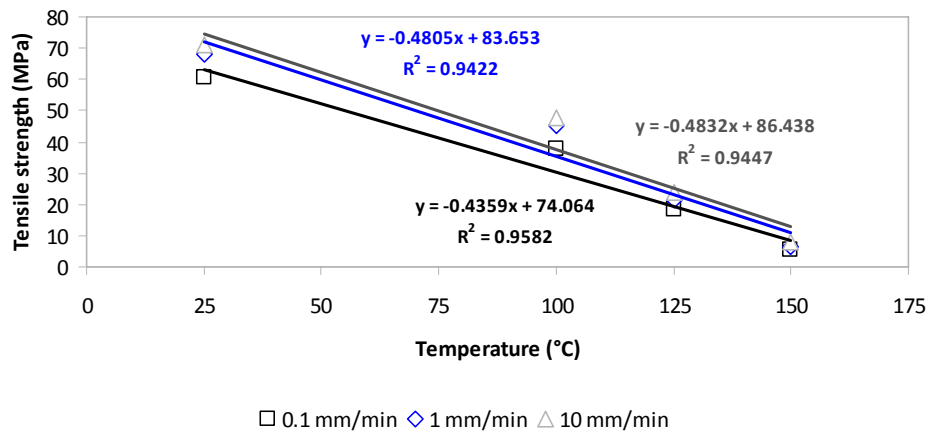
Table 3. Tensile properties of the XN1244 adhesive at a test speed of 10 mm/min.

	Young's modulus, E [GPa]	Tensile strength, [MPa]	Tensile failure strain, [%]
RT	8.17 ± 0.68	71.2 ± 0.8	1.30 ± 0.1
100°C	5.99 ± 0.23	45.7 ± 2.5	1.76 ± 0.1
125°C	2.15 ± 0.20	24.05 ± 1.6	11.28 ± 0.4
150°C	0.08 ± 0.01	7.90 ± 0.9	19.93 ± 0.2

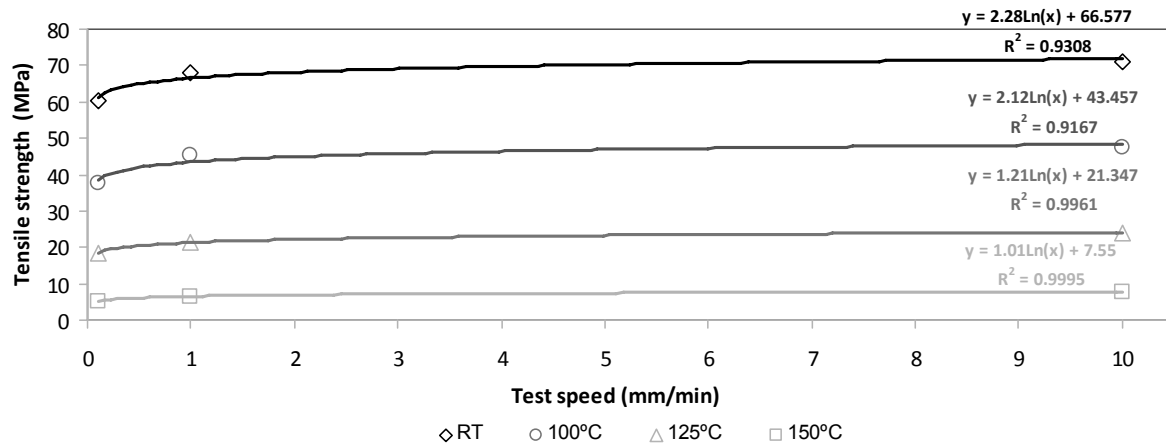
3.1. Effect of temperature on ultimate tensile strength, Young' modulus and tensile strain to failure

The data obtained (Fig. 5) shows a decrease in XN1244 adhesive ultimate tensile strength (UTS) with increasing temperature and an increase in the ductile response of the adhesive. Fig. 6a shows the UTS at different test speeds as a function of temperature with an approximately linear dependence. From RT to 100°C the UTS decreased by approximately

30% (the adhesive is in the glassy state). In the intermediate temperature range (from 125–150°C), the UTS decrease is more pronounced as the temperatures are around the T_g and the epoxy adhesive is in the transition from the glassy state to the rubbery state (see the drop in the storage modulus in Fig. 1). Finally, the UTS decreased by approximately 90% from the RT to 150°C ($\approx 0.97T_g$).



a)



b)

Fig. 6 – Tensile strength as a function of temperature (a) and test speed (b).

Fig. 7a shows the tensile strain to failure at different test speeds as a function of temperature. As can be seen, the tensile strain exhibits a nonlinear relationship with temperature. As the temperature increases, the adhesive becomes more ductile, resulting in higher strain to failure. This tendency of adhesives to have higher elongation or strain to failure is due to the increased mobility of the polymer chains in the adhesive material as temperature increases,

which allows the material to extend more easily. It is noted that at temperatures below T_g , the tensile strain to failure increased slowly with increasing temperature. At temperatures around T_g , the tensile strain to failure increased drastically with increasing temperature.

The variation of the elastic modulus with temperature is shown in Fig. 8a, for each of the three test speeds. It can be seen that the Young's modulus has a nonlinear relationship with the temperature. From RT to 100°C, the Young's modulus decreased by approximately 29%. Since the temperatures are far below the T_g , the material is still in the glassy state, so that a weak temperature dependence appears. The Young's modulus then decreases sharply in the temperature range of 100–125°C. At 150°C, the modulus decreases dramatically as the glass transition temperature of the adhesive, $T_g = 155^\circ\text{C}$, is approached. This type of behaviour is similar to that observed by Shi *et al.* [22].

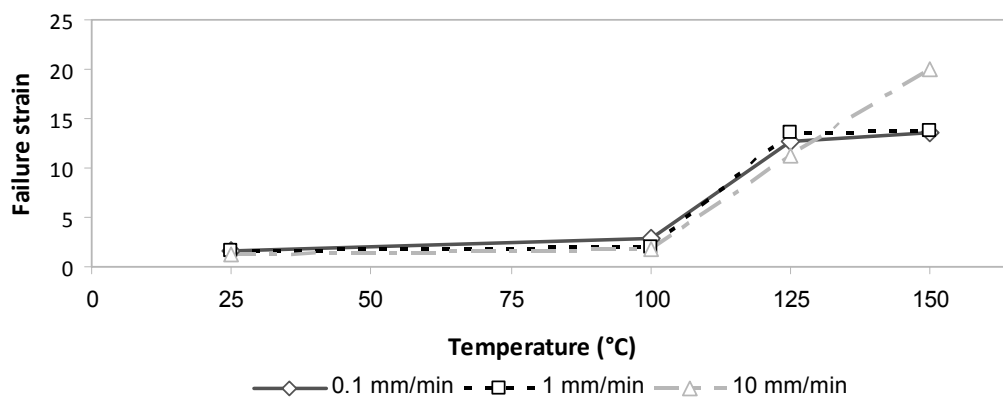
3.2. Effect of the test speed on ultimate tensile strength, Young' modulus and tensile strain to failure

Fig. 6b shows the UTS as a function of different test speeds at each temperature, which logarithmically increased with the test speed. It can be seen, that the slope for the RT and 100°C, which is below the T_g , is slightly higher than those for the high temperatures (125 and 150°C), which are around the T_g . This means that at the temperatures well below the T_g the test speed has a slightly stronger effect on the ultimate tensile stress. The Young's modulus shows a similar behaviour (Fig. 8b).

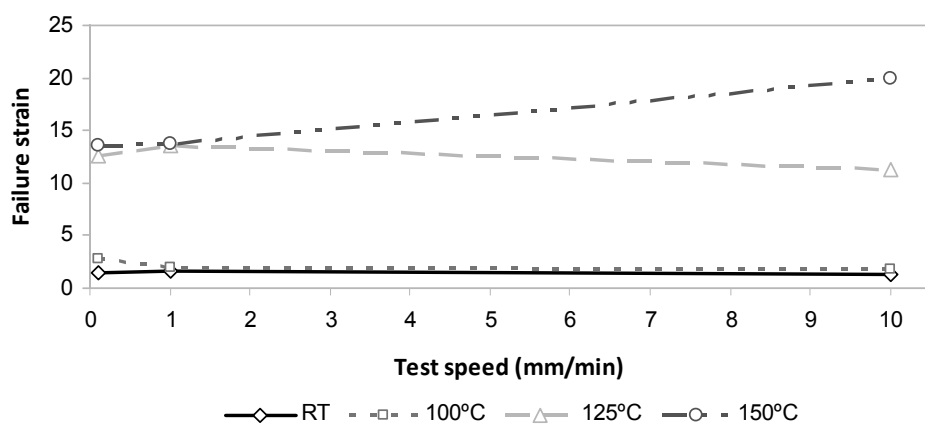
Fig. 7b shows the tensile strain to failure as a function of different test speeds for each temperature. The tensile strain to failure exhibits an approximately linear relationship with test speed. The slopes for the RT and 100°C are substantially smaller (flatter) than those for the high temperatures (125 and 150°C) indicating a reduced sensitivity of the tensile strain to failure with the test speed at these temperatures (RT and 100°C). At 150°C, the tensile failure strain linearly increased with the test speed (the correlation coefficient of the linear least-squares fit was $R^2 = 0.997$). However, the ultimate tensile strain has a more complex behaviour in relation to the test speed at 125°C (decreased with increasing and decreasing of test speed). This can be explained by the fact that at this temperature (see Fig. 5), plastic effects begin to dominate and viscous creep effects are higher.

Liang and Liechti [23] found that the ultimate shear strain is affected by the strain rate for a cross-linked epoxy resin. On the other hand, Cady *et al.* [24] found little rate sensitivity of failure strain from tests on the bulk material of PEEK or polyamideimide under compression.

In summary, at temperatures below the T_g , the test speed has a smaller effect on tensile strain to failure as compared to the effect at temperatures around the T_g . This means that at temperatures below T_g , there is little viscous creep effect.

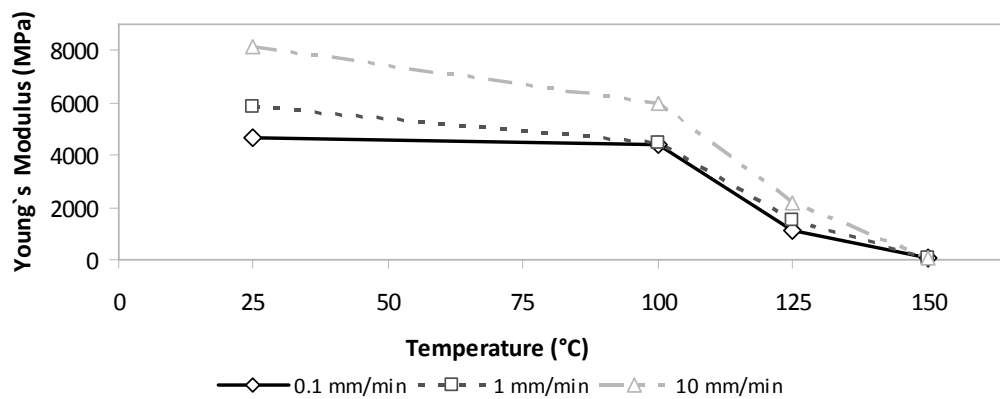


a)

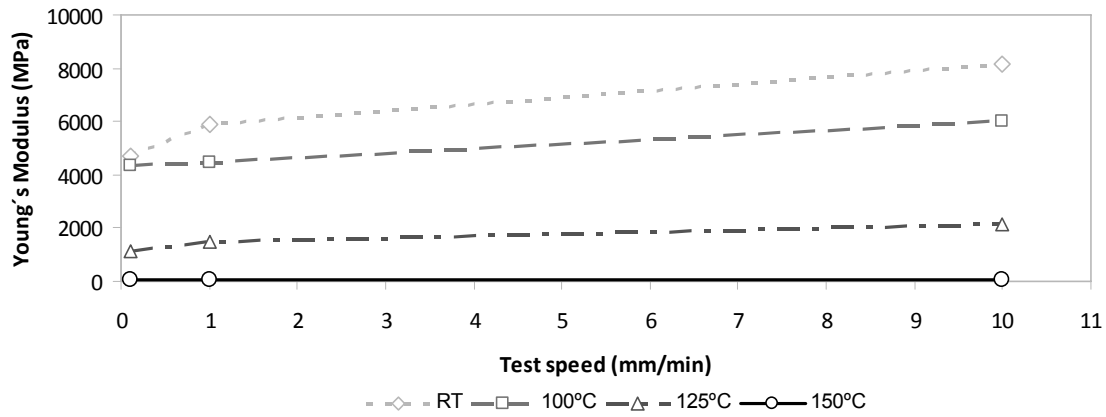


b)

Fig. 7 – Tensile failure strain as a function of temperature (a) and test speed (b).



a)



b)

Fig. 8 – Young's modulus as a function of temperature (a) and test speed (b).

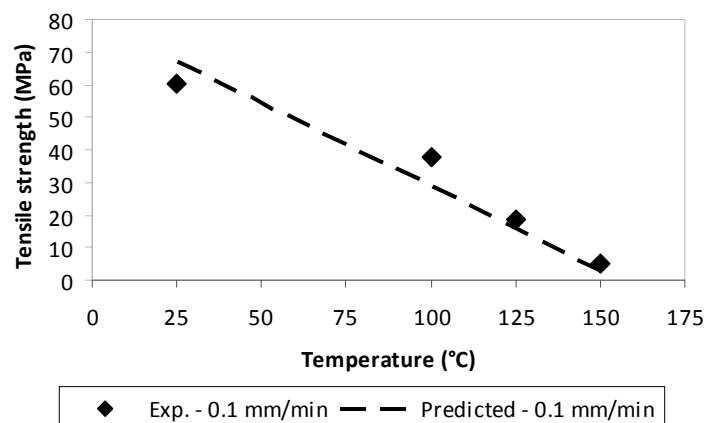
3.3. Analytical Considerations

Analytical relations can be useful for practical applications. From Fig. 6a and Fig. 6b it can be seen that the ultimate tensile stress varies linearly with the temperature and logarithmically with the test speed. Thus, the following relation can be written [25]:

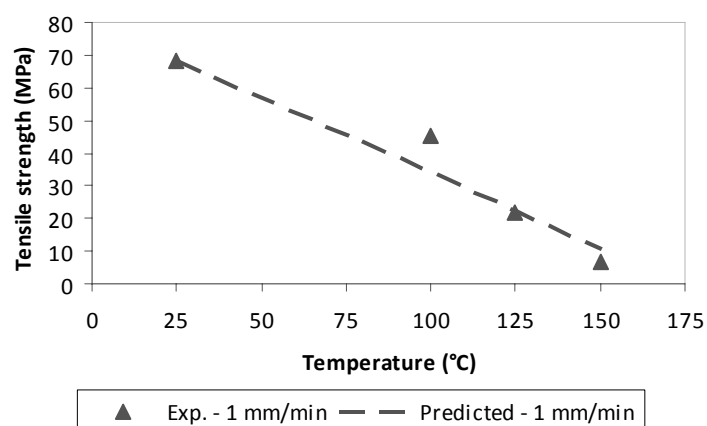
$$\sigma = A_1 \left[1 + A_2 \left(\frac{T}{T_g} \right) \log \left(\frac{\gamma}{\gamma_0} \right) \right] \quad (1)$$

Where σ is the ultimate tensile stress, in MPa, T is the temperature, in °C and γ is the test speed, in mm/min. A_1 , A_2 , γ_0 are the material constants, which were determined using the MAPLE software by solving the equations system obtained by introducing in equation (1) the values of temperature tests, experimental tensile stresses and test speeds, making the analytical curves as close as possible to the experimental curves.

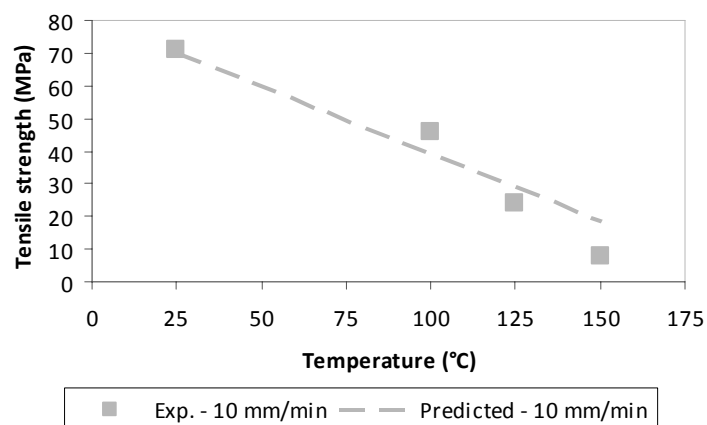
In Fig. 9, experimental and predicted ultimate tensile stresses are shown. It can be seen that the simple equation adopted gives stresses that compare satisfactory with the experimental results, provided that the coefficients (A_1 , A_2 , γ_0) are chosen as 80 MPa, 0.044, and 6.50E+08, respectively. A more fundamental understanding of the dependence of the stress on temperature and rate is offered by Airing's theory of molecular activation, where the coefficients A_1 and A_2 are interpreted in terms of activation energy and activation volume [25].



a)



b)



c)

Fig. 9 – Comparison of experimental tensile data and predictions.

3.4. Microscopic examination

A scanning electron microscopy (SEM) analysis was performed on the fracture surfaces of the “dogbone” specimens. Samples were coated with an Au layer prior to examination. The

fracture surfaces of specimens tested at RT and different temperatures were examined. Fig. 10 presents the SEM micrographs of a “dogbone” specimen tested at RT and 125°C. As expected in case of the specimen fractured at 125°C (Fig. 10b) it can be seen that the surface is more irregular indicating a higher deformation, i.e. the material is more ductile (at higher temperatures the yield strength is lowered and the fracture is more ductile in nature).

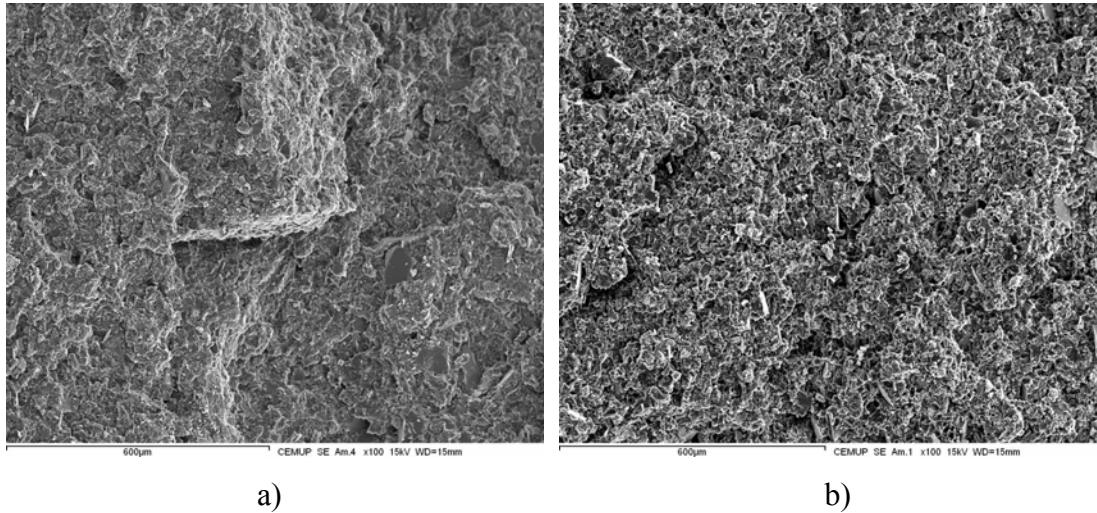


Fig. 10 – SEM micrograph of fracture surfaces of “dogbone” specimens tested in tension at 1 mm/min: a) at RT; b) at 125°C (x100).

Similarly, typical SEM micrographs are presented in Fig. 11 to show the effect of test speed on the fracture behaviour of the tested “dogbone” specimens. It can be noted that the differences of SEM micrographs for different test speed are very small. This observation verifies the fact that the test speed has a smaller effect on the tensile properties of the epoxy adhesive as compared with the effect of temperature.

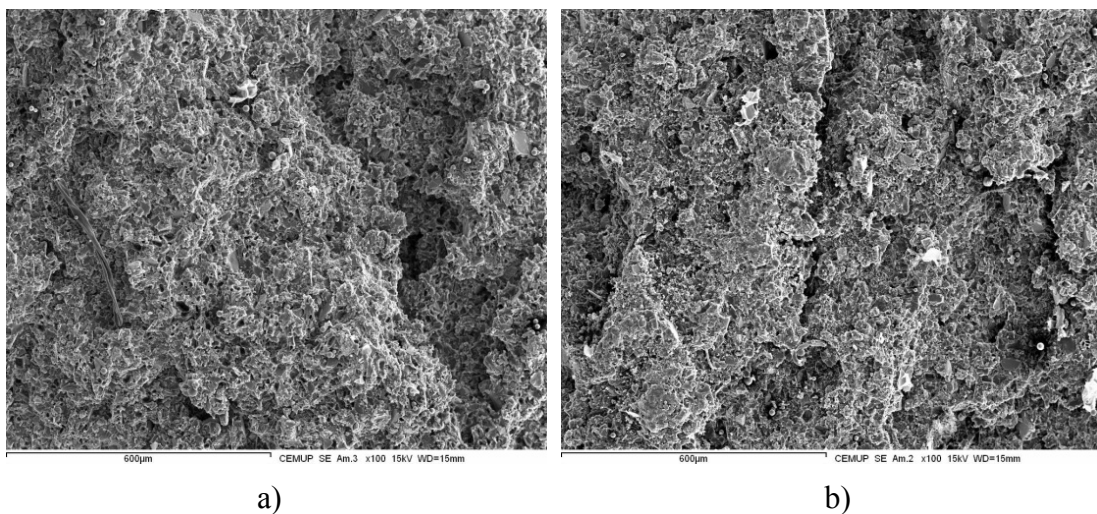


Fig. 11 – SEM micrograph of fracture surfaces of “dogbone” specimens tested in tension: a) at 0.1mm/min; b) at 10 mm/min (x100).

4. Conclusions

Tensile data obtained from tests at RT showed that the XN1244 adhesive has a high strength and high stiffness. Large increases in ductility (strain to failure) and reductions in ultimate strength were measured at elevated temperatures (100°C, 125°C and 150°C).

The ultimate tensile stress linearly decreased with temperature while logarithmically increased with the test speed, which is in accordance with the Airing's molecular activation model. The Young's modulus nonlinearly decreased with the temperature, while shows a similar behaviour as the UTS as a function of test speed. The ultimate tensile strain nonlinearly increased with temperature, while has a more complex behaviour in relation to the test speed. At RT and 100°C (when the temperatures are far below the T_g), reduced rate sensitivity of tensile strain with the test speed was found as compared to the effect at temperatures around the T_g . At 150°C, the tensile failure strain linearly increased with the test speed, while at 125°C the ultimate tensile strain decreased at a higher test speed. As expected, the temperature effect on the properties of the adhesive is more significant than that of the strain rate.

A database of mechanical properties of the XN1244 high temperature epoxy adhesive was established which can be used in finite element analysis to simulate the deformations at different testing conditions.

Acknowledgements

The authors would like to thank the Portuguese Foundation for Science and Technology for supporting the work presented here, through the individual grant SFRH/BD/61880/2009 and through the research project PTDC/EME-PME/67022/2006, and Nagase ChemteX (Japan) for supplying the adhesive.

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**Mode I Fracture Toughness of Adhesively Bonded Joints as a
Function of Temperature (RTV106)**

Temperature Dependence of the Fracture Toughness of Adhesively Bonded Joints

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Abstract

Adhesives used in structural high temperature space and aerospace applications must operate in extreme environments. They need to exhibit high-temperature capabilities in order to maintain their mechanical properties and their structural integrity at the intended service temperature. One of the adhesives which are able to withstand the temperature extremes that are experienced in the space environment and are able to maintain a good degree of flexibility at very low temperatures are the room temperature vulcanizing (RTV) silicone adhesives.

As is known, adhesive strength generally shows temperature dependence. Similarly, the fracture toughness is expected to show temperature dependence. In this study, the pure mode I fracture toughness for adhesive joints bonded with a high temperature RTV silicone adhesive was measured over a wide range of temperatures. Double cantilever beam (DCB) tests were performed on specimens at room temperature (RT), 100 and 200°C. Mode I traction–separation laws were obtained as a function of temperature, directly from the experiments, by differentiation of simultaneously measured data (the J -integral and the end-opening displacement). Results showed that the fracture toughness, the peak cohesive stress and the respective end-opening displacement all decreased with the temperature rise.

Keywords: RTV adhesives; fracture toughness, traction–separation laws, temperature tests.

1. Introduction

RTV silicone adhesives are being increasingly used in a variety of space applications (e.g. in satellites construction and spacecrafts) as they are able to withstand the temperature extremes that are experienced in the space environment and to maintain a good degree of flexibility at very low temperatures. They cure from the moisture in the air and form low strength structural joints. They require access of moisture for curing, and, therefore, cannot be cured in deep sections. Typically a bondline width or depth might not be greater than 10-15 mm so that moisture can diffuse and react with the material in the centre of the joint. If greater depths are required, two-component RTV curing silicones are generally used. RTV silicone adhesives retain excellent properties over a temperature range from nearly 300°C down to cryogenic values, providing low-temperature flexibility and very high temperature resistance. Also, they are used where organic materials (based on carbon) cannot withstand harsh environmental conditions. The excellent peel strength properties of silicones are more important in joints design than the tensile or lap-shear properties [1]. However, the RT mechanical properties of RTV silicone adhesives are quite low compared to typical polymers. Moreover, silicones have not found broad use as adhesives, relatively to the total consumption volume, because of their high cost.

Relatively only limited data are available relative to the mechanical properties of silicone adhesives. For example, the stress-strain characteristics of two moisture curing RTV elastomeric adhesives were investigated in the tensile mode and in the lap shear configuration by Geiss and Vogt [2]. They found that ageing in a humid atmosphere significantly influenced the mechanical properties of the adhesives. Banea and da Silva [3,4] studied the performance of two high temperature RTV silicone adhesives through bulk and adhesive joint tests, concluding that the RTV adhesives retain their flexibility and a substantial part of their strength in the temperature range from RT to 300°C.

As is known, adhesive strength generally shows temperature dependence. Studies that present experimental results on adhesive joints with structural adhesives (especially epoxies) as a function of temperature generally show a decrease in strength at high and low temperatures [5-7]. At high temperatures this is due to the low adhesive strength, while at low temperatures the high thermal stresses and the brittleness of the adhesive are the origin of such behaviour. Similarly, the fracture toughness is expected to show temperature dependence.

The majority of fracture characterization of adhesively-bonded assemblies under pure mode I have been performed using the DCB specimen [8-11]. The main advantages of this test method include its simplicity and the possibility to obtain the fracture toughness mathematically using the beam theory for brittle materials [12].

Several techniques can be used to derive the fracture toughness of structural adhesives from fracture characterization tests. The most common methodologies for analysis are based on Linear-Elastic Fracture Mechanics (LEFM). The Compliance Calibration Method (CCM) is based on the Irwin-Kies equation [13,14], requiring the calculation of the compliance, C , ($C=\delta/P$, where δ is the displacement and P is the applied load) relative to the crack length during crack growth. The Direct Beam Theory (DBT), based on elementary beam theory [15], and the Corrected Beam Theory (CBT), including the effects of crack tip rotation and deflection [16], are also available within the scope of LEFM. However, the use of LEFM is not recommended when the fracture process zone is large, as the assumed stress fields are not rigorously correct on account of the large-scale plasticity [17]. In some structures, the Fracture Process Zone (FPZ) is long in one dimension but small in the perpendicular direction. This type of process zone is sometimes due to large scale bridging (LSB) [18]. These two problems should not be analysed by the LEFM either [19].

As explained by Suo et al. [20], LEFM solutions for stress intensity factors or energy release rates generally differ from the J -integral solution in the case of large-scale plasticity. For most specimens, closed form analytical solutions for J -integral cannot be obtained, since the J -integral solution depends on the details of the stress-separation law. Nevertheless, analytical solutions for the J -integral are available for a few fracture test specimens [20]. Li and Ward [21] used two compact tension specimens having different notch sizes to obtain the J -integral from the area between the two load–displacement curves. Another example is the DCB specimen loaded under pure bending moments (for pure mode I stress-separation laws) [18, 20, 22-24]. However, the standardised DCB tests do not provide a pure bending moment in the specimen arms. It is thus necessary to include the contribution of the rotation of the beams near the crack front for the J -integral calculation [25, 26].

Under the circumstances of large-scale plasticity or LSB, fracture can be modelled by a stress-separation law [19, 20], which represents the local normal stress as a function of the local crack opening.

When the FPZ in a structure is comparable to or larger than some length dimensions (e.g. the crack size), strength prediction requires modelling of the failure process zone. This can be

done by simulating crack growth with cohesive zone models (CZMs). The key concept of a CZM is that the FPZ can be described by a traction–separation law relating stresses and displacements between the crack faces, thus simulating the gradual degradation of the material properties. As a result, a second fracture parameter, the cohesive strength, is introduced in addition to the fracture toughness. The shape of the softening law can also be adjusted to conform to the behaviour of the material being simulated.

A few number of techniques have been used in the past to define the traction-separation laws for adhesive layers, both brittle and ductile. The most common one relies on iterative comparisons between experimentally measured data and the respective numerical predictions, considering a precise description of the experimental geometry and approximate cohesive laws with a parameterized shape, established based on the typical behaviour of the material that the law simulates. Using this technique, the value of fracture toughness, which corresponds to the plateau value of the respective R -curve of the fracture characterization test, is used as an input parameter in the numerical model. These models also include typical values for the cohesive strength. A few numerical iterations must be performed until a good accuracy between the experimental and numerical data is obtained. Examples of the experimental data used for fitting are the R -curve [27], the crack opening profile [28], and most typically the P - δ curve [29]. The direct method is also available, allowing extraction of the cohesive law of an adhesive layer directly from the measured data from the experiments without requiring such a laborious numerical analysis [30]. In this method, the damage evolution across the width of the specimens has to be uniform, which is typically difficult to achieve. The direct method is based on the simultaneous measurement of the J -integral and the end-opening (normal or shear) of the cohesive zone. This has been successfully employed in the extraction of traction–separation laws for adhesive bonds [24, 26] and fiber-reinforced composites [18].

In this study, the pure mode I fracture toughness of adhesive joints bonded with a high temperature RTV silicone adhesive was measured over a wide range of temperatures. DCB tests were performed at RT, 100 and 200°C. Pure mode I traction–separation laws were obtained as a function of temperature, directly from the experiments by differentiation of simultaneously measured data (the J -integral and the end-opening displacement).

2. Experimental details

2.1. Adhesive

The adhesive investigated in this study was a 1-part high temperature acetoxy adhesive sealant, RTV106 silicone rubber, supplied by GE Bayer Silicones. The working temperature of the adhesive RTV106 is from -60°C to 260°C .

A key parameter in the testing of adhesive joints is the glass transition temperature (T_g) of the adhesive. When the adhesively bonded joints are tested below this temperature, the adhesive will behave like a low-strain rigid material while above this temperature it will have a more rubber-like behaviour. For example, the elastomeric adhesives (the case of the RTV silicone studied here) have a T_g below 0°C and are in a rubbery state at room temperature, so that they have different behaviour in adhesively bonded joints when tested at different temperatures.

The glass transition temperature (T_g) of the RTV106 adhesive is -60°C (data provided by supplier).

2.2. Specimen fabrication

Steel substrates were used for the DCB specimens. The joint surfaces were grit blasted and degreased with acetone prior to the application of the adhesive. The bondline thickness was nominally 1 mm. Spacers were inserted between the adherends before the application of the adhesive in order to control the bondline thickness. These spacers were removed after the adhesive was cured. The specimen geometry and the loading are shown in Figure 1. A sharp pre-crack in the adhesive layer mid-thickness was assured using a razor blade. A mould with spacers for the correct alignment of the adherends was used and is shown in Figure 2. The DCB joints were cured at room temperature for a week.

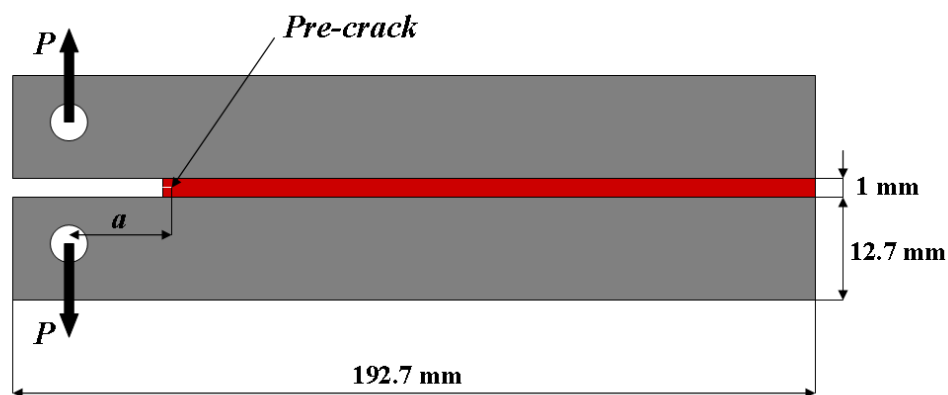


Figure 1. DCB specimen geometry.

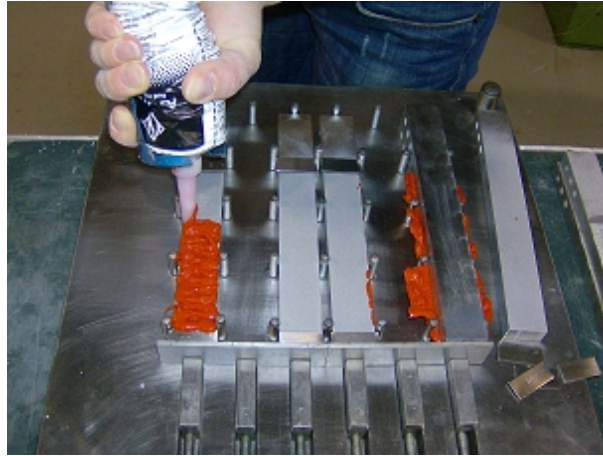


Figure 2. Mould for DCB specimens fabrication.

2.3. Test procedure

The DCB specimens were tested at RT and high temperatures (100°C and 200°C) using a universal testing machine Instron[®] model 8801 (Instron Co., USA), under a constant crosshead rate of 1 mm/min. For the high temperature tests, the environmental chamber of the machine was used to reach the desired test temperatures. Three joints were tested to failure at each temperature. The DCB specimens setup is shown in Figure 3.

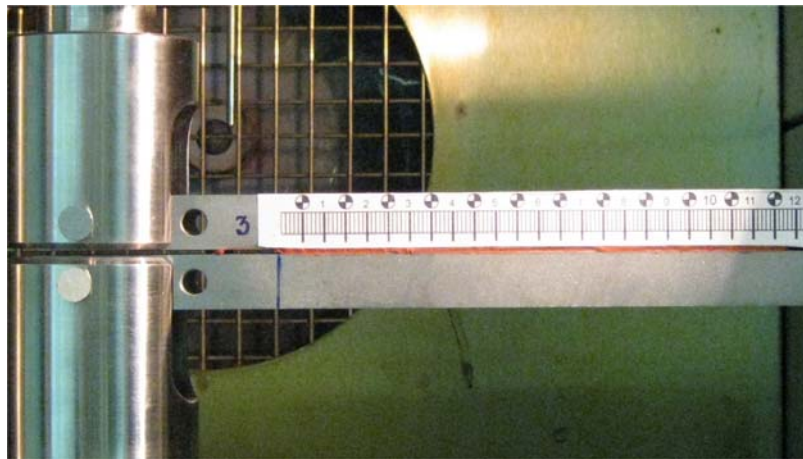


Figure 3. DCB specimen setup.

A thermocouple was applied to the specimen in order to assure that the air temperature inside the chamber was equal to the specimen's temperature. The tests were always performed after approximately 10 min of achieving the test temperature in the specimens, to ensure a steady-state temperature throughout the specimen prior to testing.

As shown by Nairn [10], the effect of residual stresses on adhesively bonded joints may be significant but is often ignored. In this study, the presented fracture toughness of the adhesive

was determined by an expression that ignores the effects of residual stresses due to heating from RT to the test temperatures. If the grips of the loading machine are kept at a fixed position during the heating process (to 100° and 200°C) a compression loading on the specimens is expected. In fact, heating of the loading apparatus (e.g. specimen holders and tension rods) causes their extension, and thereby induces a compression mode I loading on the specimen. On the other hand, the DCB arms cannot be held fixed at the neutral position (zero load at RT). To provide consistency in the high temperature test results, the crosshead of the test frame was adjusted to provide zero load on the specimen during heating and before testing. Therefore, the reference position for zero displacement for all tests corresponds to the position for zero applied load on the specimen at the test temperature.

A digital camera was used to monitor the separation of the adherends. The digital camera was set to take pictures every 10 s. recording the changes in separation of the two beams throughout the test. The digital images were then analysed using an image processing and analysis software, and the end-opening displacement (δ_h) and the rotation of the beams at the tip of the crack were extracted for each specimen.

2.4. Data analysis

As mentioned previously, if considerable plasticity is present in the adhesive, the fracture toughness cannot be determined from the elastic stress-field and LEFM techniques are, therefore, inappropriate, since the fracture toughness is significantly affected by the plastic zone at the tip of the crack [31]. The J -integral method is an effective tool to determine the fracture toughness, regardless of the shape of the bond as well as the linear or nonlinear behaviour of the adhesive, given generally as [32]:

$$J = \int_{\Gamma} \left(W dy - T \frac{\partial u}{\partial x} ds \right), \text{ with } W = W(x, y) = W(\varepsilon) = \int_0^\varepsilon \sigma_{ij} d\varepsilon_{ij}, \quad (1)$$

where W is the strain energy density, σ is the stress tensor and ε the strain tensor. Γ is a closed contour counter clockwise around the crack tip beginning and ending at the crack faces, T is the tension vector perpendicular to Γ pointing towards the outside of the contour, u is the displacement in the x -direction and ds is an element of Γ .

The J -integral, calculated with Equation (1), may lose its path independence from Γ and no longer be representative of crack tip stresses in the case of large scale plasticity and extensive crack propagation. However, to determine the cohesive law in pure mode I of the adhesive

layer of silicone/steel DCB specimens as a function of temperature, the J -integral was determined by Equation (2) that is valid in any situation.

For a DCB specimen loaded by point loads at the cracked edge (see Figure 4), the J -integral can be determined by eq. 2 [26]:

$$J = 12 \frac{(Pa)^2}{Eh^3} + P(w_1' - w_2'), \quad (2)$$

where P is the applied load per unit width, a is the crack length, h is the adherend thickness, E is the elastic modulus of the steel adherends, and w_1' and w_2' are, respectively, the rotations of the upper and lower adherends at the crack tip (see Figure 4).

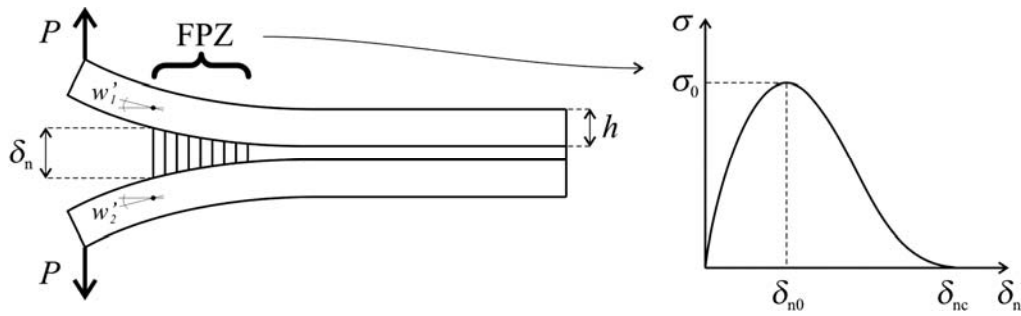


Figure 4. Scheme of the DCB specimen under loading.

Evaluating the J -integral along a path just outside of the FPZ yields:

$$J = \int_0^{\delta_{nc}} \sigma(\delta_n) d\delta_n, \quad (3)$$

where δ_n and δ_{nc} are the normal opening and the normal end-opening of the cohesive law at the tip of the initial crack, respectively, and σ is the normal traction. J -integral reaches a steady-state value, J_{ss} , when δ_n attains δ_{nc} , the critical normal end-opening. Due to its path independence, the J -integral in eq. (3) is equal to that obtained by eq. (2).

Since the normal end-opening δ_n can be recorded in digital images, the normal traction can be obtained by differentiation of Eq. (3) with respect to δ_n :

$$\sigma(\delta_n) = \frac{\partial J}{\partial \delta_n} \quad (4)$$

The pure mode I traction–separation law can then be obtained by measuring simultaneously the J -integral and the normal end-opening δ_n up to J_{SS} , followed by the polynomial fitting and differentiation of the respective J - δ_n curve.

3. Results and discussion

Representative load – time curves as a function of temperature can be seen in Figure 5. At RT the crack starts at approx. 200s of testing (when the maximum load is reached). At 100°C the behaviour was identical (the crack appears at approx. 200s) but at a slightly lower load, while at 200°C the maximum load and onset of crack propagation substantially diminish (the crack appears at approx. 120s), which anticipates a major reduction in the adhesive properties at 200°C.

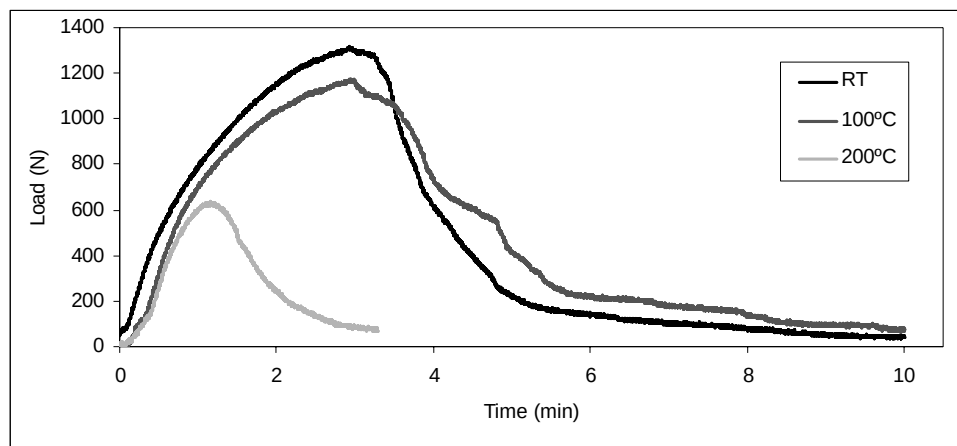


Figure 5. Representative load – time curves as a function of temperature.

3.1. Determination of J - integral values

The J -integral, the maximum cohesive stress (σ_0), the respective normal end-opening (δ_{n0}) and the critical normal end-opening (δ_{nc}) as a function of temperature are presented in Table 1. The traction–separation laws clearly exhibited temperature dependence, with the values of J and σ_0 decreasing with temperature rise.

Table 1. J -integral, maximum cohesive stress (σ_0), and respective normal end-opening (δ_{n0}) and critical normal end-opening (δ_{nc}) as a function of temperature.

Temperature °C	J -integral (N/m)	Max. Cohesive Stress σ_0 (MPa)	δ_{n0} (mm)	δ_{nc} (mm)
RT	2731 ± 129	2.08 ± 0.11	0.96 ± 0.25	2.12 ± 0.29
100	2050 ± 68	1.30 ± 0.17	0.84 ± 0.20	2.24 ± 0.19
200	387 ± 48	0.85 ± 0.15	0.28 ± 0.10	0.70 ± 0.10

The variation of the J -integral with the normal end-opening displacement at RT can be seen in Figure 6 for the three specimens tested. A cubic polynomial was fitted to the J - δ_n response, showing an accurate fit. This figure represents J as a function of δ_n , in the interval $[0, \delta_{nc}]$, where δ_{nc} is defined by the point when the maximum J , J_{SS} , is reached. Testing always continued after J_{SS} was attained, but only the region up to J_{SS} is shown, since this fully defines the cohesive law [25]. The J - δ_n curves for 100°C and 200°C are shown in Figure 7 and Figure 8, respectively.

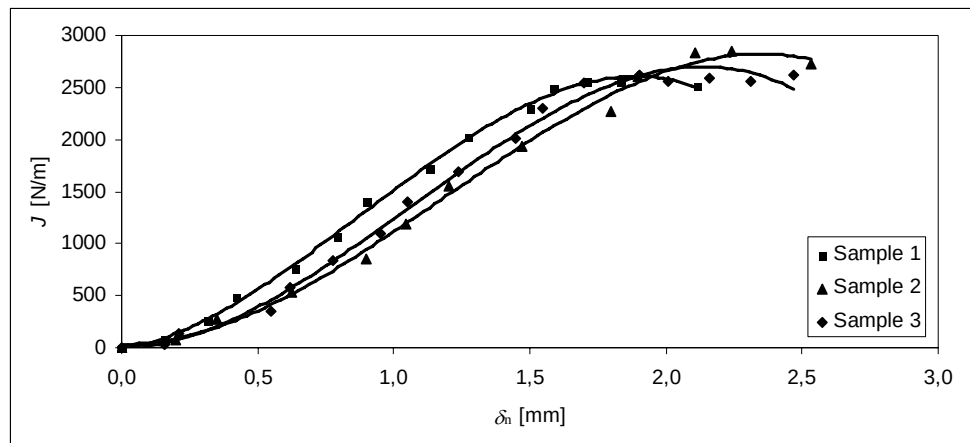


Figure 6. J - δ_n response at RT (three specimens tested).

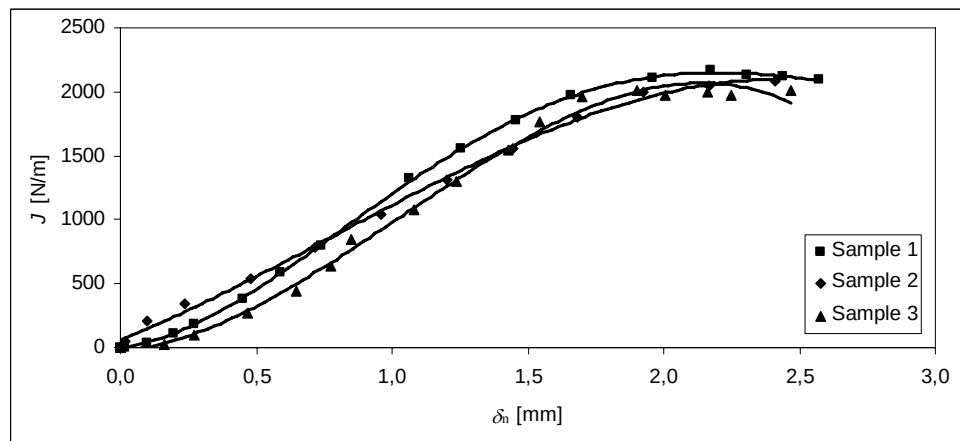


Figure 7. J - δ_n response at 100°C (three specimens tested).

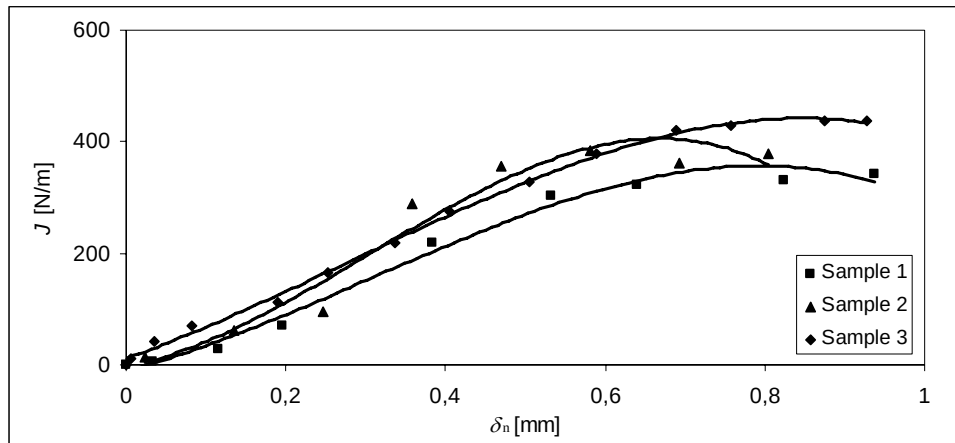


Figure 8. J - δ_n response at 200°C (three specimens tested).

Representative J - δ_n response curves for the three temperatures tested can be seen in Figure 9. It is clear that the adhesive exhibited temperature dependence. J -integral decreased with temperature rise, due to degradation of the mechanical properties of the adhesive induced by the temperature [4].

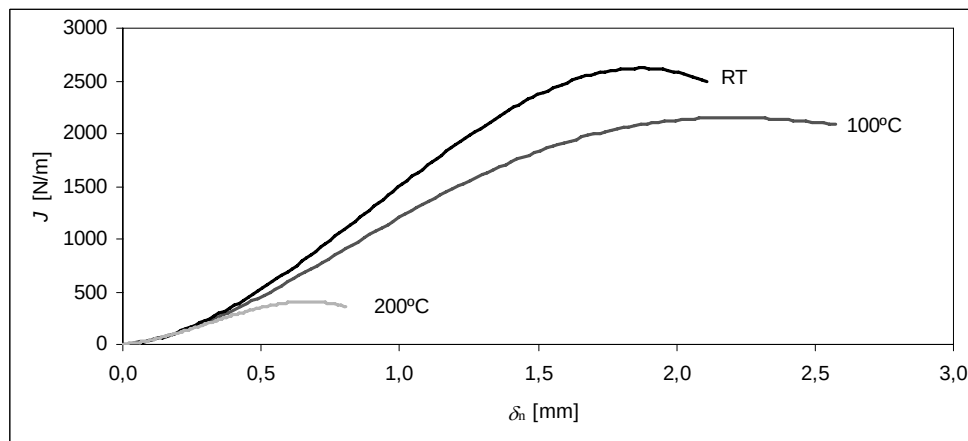


Figure 9. Representative J - δ_n response as a function of temperature.

3.2. Cohesive laws

The cohesive laws of the tested adhesive in pure mode I were calculated by differentiation of the J -integral with respect to the normal end-opening δ_n (Eq. 4). Representative cohesive laws for the three temperatures tested (RT, 100°C and 200°C) are shown in Figure 10.

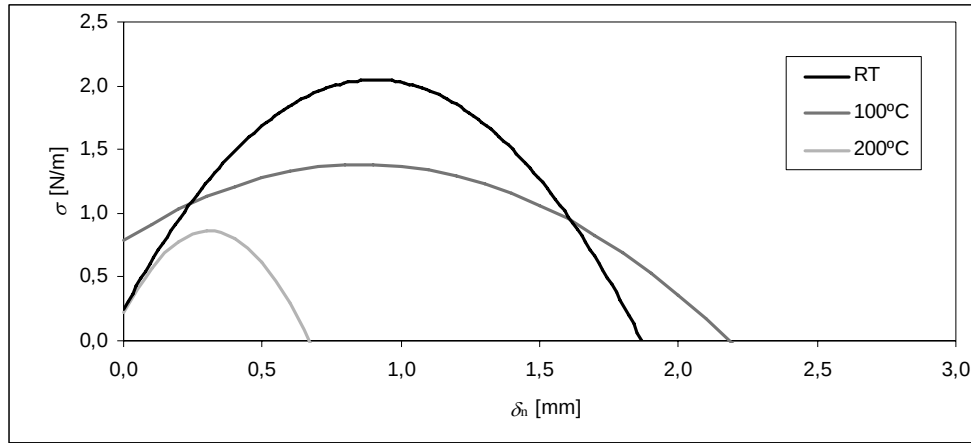


Figure 10. Representative cohesive laws as a function of temperature.

For all the curves, the value of σ is not nil at the onset of loading ($\delta_n=0$), and increases with δ_n up to a peak value. At this point, σ starts to decrease up to complete failure. Figure 10 shows that the shape of the cohesive law substantially varies with the test temperature, although the reduction of δ_{n0} from RT to 100°C is negligible. The values of σ_0 and δ_{n0} decreased with the increase of the test temperature. The value of δ_{nc} increased at 100°C and decreased at 200°C from the RT.

prominent feature of the traction–separation laws relates to the nonzero value for σ at the initiation of loading. This behaviour was previously reported by Sørensen [24] who also extracted the traction–separation laws of an adhesive layer of polyurethane using the direct method. Identical tendencies were equally found in traction–separation laws extracted by inverse modelling techniques [28,33]. Theoretical reasoning for this behaviour is provided in the work of Jin and Sun [34], where it is argued that this particular shape is required to remove the singularity at the crack tip. In fact, by considering the cohesive zone development and crack extension in an infinite elastic background medium, it is found that the stress singularity at the tip of the cohesive zone cannot be removed unless the cohesive law has a nonzero traction at the initial zero opening displacement. However, it seems unlikely that a real material would have a steep increase in σ at $\delta_n=0$, and such behaviour would then be related to an actual steep increase of the σ - δ_n curve in this region, added to a typically long period of time between recorded images and lack of accuracy of the polynomial fit near $\delta_n=0$ [26]. Actually, as shown by Zhu *et al.* [26], the derivation of the fitting polynomial of the J - δ_n can increase the data scatter and also yield some smearing of the real curve details, especially in the initial stages of loading.

Concerning the influence of using the exact shape of the cohesive law in the numerical calculations of strength prediction, it is more or less accepted amongst researchers that the effect of the shape on the fracture process is relatively unimportant for most materials [26,35]. Despite this fact, Gu [36] described in his work that the shape of the cohesive law actually affected the mechanics of crack growth in fiber-reinforced ceramics, while Jacobsen and Sørensen [37] made similar observations regarding mode I intralaminar crack growth in fibre-reinforced polymers. These are exceptions to the usual behaviour of materials.

3.3. Failure modes

For high strength adhesives there are four typical mechanisms of crack growth, which are represented in Figure 11 [38]. The most representative one is the cohesive failure (a1 and b1), characterized by a proper surface preparation that leads to a stronger interface with the adherends than the cohesive properties of the adhesive. Interfacial failures (c1) are frequent when bonding adherends with low surface energy or as a result of a poor preparation of the bonding surfaces. Alternative crack propagation between interfaces can also appear (d1), due to opening of micro-cracks at opposite interfaces alternatively ahead of the crack tip. The tensile stresses within the bond plane can also play an important role in this behaviour, causing the crack to oscillate within the adhesive layer or alternate from one adherend to the other. This effect is controlled by the T-stress, a non-singular stress that is parallel to the local crack path. If the tensile magnitude of the T-stress is sufficiently large, the crack path is not stable and so will continuously change direction as it propagates [39].

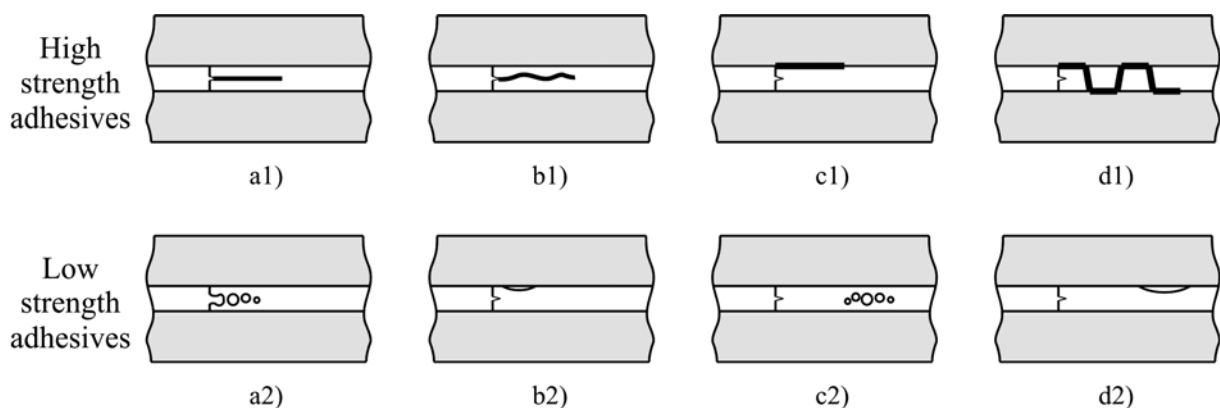


Figure 11. Typical failure modes for high strength structural adhesives and ductile low strength adhesives.

For ductile low strength adhesives, a number of typical failure mechanisms have been reported in the literature: (a2) near-tip void growth and coalescence, (b2) interface debonding near the crack tip, (c2) high triaxiality cavitation ahead of the crack tip and subsequent coalescence, and (d2) interfacial debonding ahead of the crack tip [40-42]. Schematics of these mechanisms are also shown in Figure 11. The RTV silicone adhesive studied here belongs to ductile group of adhesives, since RTV silicone adhesives can be stretched to several hundred percent.

The failure mechanism a2) is related to ductile fracture of homogeneous materials. Under these conditions, the crack tip typically blunts to identical dimensions of the average spacing between voids. Since these voids are under high strains, their coalescence will lead to propagation of the crack. The failure mechanism b2) can develop when the through-thickness normal stresses near the tip of the crack exceed the bonding strength of a weak interface region. However, with a strong bond between the adhesive and adherends, the adhesive layer will endure extensive plasticization and the maximum normal stresses will occur far from the tip of the crack. This is the case of the following mechanisms (c2 and d2). The failure mechanism c2) is triggered by stress triaxiality ahead of the crack tip which develops when the interfacial bond is sufficiently strong to allow the ductile layer to undergo substantial plastic deformation. The failure mechanism d2) has similarities with c2), occurring due to stress triaxiality ahead of the crack tip, but in presence of a weaker interface than the cohesive properties of the adhesive. The difference between these mechanisms is caused by issues such as the high stress triaxiality induced by constrained plastic flow and stress relaxation caused by extensive voiding.

Figure 12 shows examples of typical crack initiation for pure mode I fracture testing at RT, 100°C and 200°C. The failure mechanism b2) was observed at RT and 200°C, while at 100°C the damage initiation was according to mechanism d2) and then propagated mostly as a cohesive failure (see Figure 12). The crack initiation was sometimes accompanied by a small drop in the load (see Figure 5). Further increasing the load, the crack propagated through the specimen.

In some of the specimens (particularly for testing at RT), the decohesion shifted to the interface after a debond length of approximately 10 mm (somewhat different for different specimens), creating a bridge of adhesive between the fracture surfaces (see Figure 13a). Whether this behaviour is caused by a local variation of the bond strength or whether it is related to the material non-linearity is still unclear. From a structural point of view, crack bridging can be considered as beneficial, since the formation of a crack bridging zone results

in an apparently increasing fracture toughness of the materials at the crack tip, due to the increasing load required to propagate the crack (mentioned in the literature as *R*-curve behaviour [20]).

As observed in Figure 12 for testing at RT, in the DCB specimens the separation occurred by a mixed cohesive and interfacial failure (along the steel/adhesive interface), but mostly as a cohesive failure (Figure 14a). This indicates that at some locations the interface was weaker than the cohesive strength of the silicone adhesive. This phenomenon was also observed in failure of single lap joints (SLJs) with the same adhesive tested at RT [4].

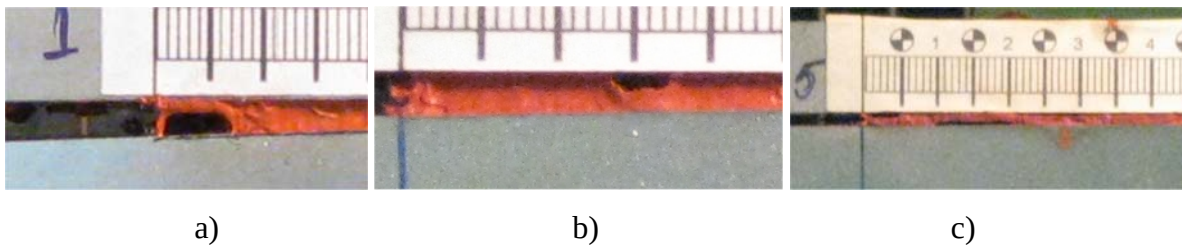


Figure 12. Initiation of failure at RT (a), at 100°C (b) and at 200°C (c).

At 100°C, the initial fracture occurred by interfacial debonding ahead of the crack tip (see Figure 12b), but as the load was further increased, fracture developed typically cohesively (Figure 14b). Bridging was also detected at 100°C, but was much less pronounced than at the RT (see Figure 13b), which can be the origin of the small reduction of J from the RT (Table 1). The onset of blunting at the tip of the crack for RT and 100°C (Figure 12a and b, respectively) is evident, as the cohesive stresses increased, but small cavitations could be seen developing ahead of this region (initiation and growth of debonding at the upper or lower interface slightly ahead of the pre-crack). Although the crack opening was large, the crack did not propagate prior to cavitation. The existing decohesion regions increased in dimensions as the specimens were loaded and more cavitations appeared along the interfaces, leading to a reduction of the cohesive stress. The final stage of the separation process corresponded to zero cohesive stress and the growth of the crack.

At 200°C, the plastic deformation and the rotation of the beams are much smaller than at RT and 100°C, due to the degradation of the adhesive properties, and the crack propagation takes place at smaller normal displacements. Moreover, the mixed failure observed in all of the specimens was predominantly interfacial (Figure 14c).

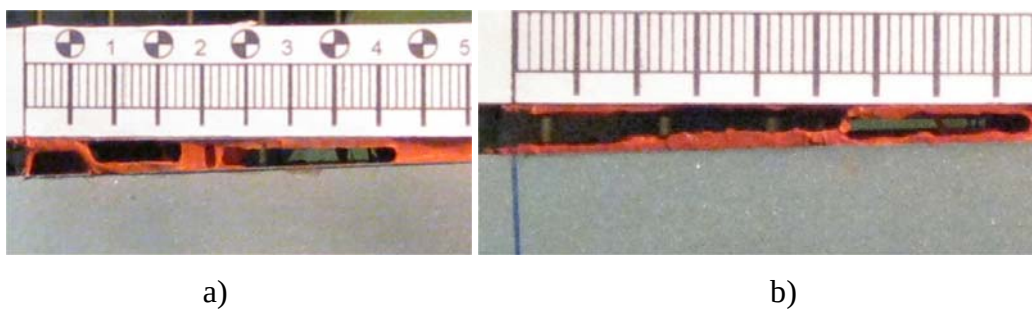


Figure 13. Propagation of failure at RT (a) and at 100°C (b).



a) RT



b) 100°C



c) 200°C

Figure 14. Typical failure modes at RT (a), 100°C (b) and 200°C (c).

4. Conclusions

Pure mode I traction–separation laws were obtained for silicone/steel DCB specimens. The cohesive laws were obtained directly from the experiments by differentiation of simultaneously measured data (the J -integral and the end-opening displacement).

Several important observations on the traction–separation laws and the associated fracture features are summarized in the following:

- The traction–separation laws can be suitably fitted with a cubic polynomial.
- The fracture toughness and the traction–separation laws exhibited temperature dependence. The fracture toughness, the peak cohesive stress and the respective end-opening displacement decreased with the temperature rise, while the critical opening displacement increased at 100°C and decreased at 200°C from the RT.
- The initial failure of the specimens occurred by interfacial debonding near or ahead of the original cohesive crack-tip, but mixed propagations were observed, including regions of cohesive failure and spots of interfacial failure.

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**Mode I Fracture Toughness of Adhesively Bonded Joints as a
Function of Temperature – Experimental Study (XN1244)**

Effect of Temperature on Tensile Strength and Mode I Fracture Toughness of a High Temperature Epoxy Adhesive

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Abstract

There has been a growing requirement in the last years, particularly in the aerospace industry, for adhesives to withstand high temperatures. The most important factor to consider when studying the effect of temperature on adhesively-bonded joints is the variation of adhesive mechanical properties with temperature such as the stress-strain curve and the toughness. Adhesive strength and strain show temperature dependence, especially near the glass transition temperature (T_g) of the adhesive. Similarly, the fracture toughness is expected to show temperature dependence.

In this work, an experimental study is performed to evaluate the effect of the temperature on the adhesive strength and fracture toughness of a high temperature epoxy adhesive. Bulk specimens of cured adhesive were produced and tested in tension at room temperature (RT), 100°C, 150°C and 200°C, in order to obtain a strength profile of the adhesive over this temperature range. Results showed that as the temperature increases the adhesive tensile strength reduces but the ductility increases. Pure mode I adhesive fracture toughness (G_{Ic}) tests were performed on double cantilever beam (DCB) specimens at room (RT) and high temperatures (100°C, 150°C and 200°C) and the fracture toughness G_{Ic} as a function of temperature was obtained. It is shown that at temperatures below T_g , the fracture toughness, G_{Ic} , is relatively insensitive to temperature, while above T_g (at 200°C) a drastic decrease in fracture toughness was observed.

Keywords: High temperature adhesives, fracture toughness, tensile strength, temperature tests.

1. Introduction

In the last years, there has been a growing requirement, particularly in the aerospace industry, for adhesives to withstand high temperatures. The adhesives used in structural high temperature aerospace applications must operate in extreme environments. These adhesives have to maintain their mechanical properties at the intended service temperature and to maintain their structural integrity (resist thermal breakdown at elevated temperature). Thus, it is fundamental for an efficient and quick design process to understand the fracture behavior of these adhesives at high temperatures and the existence of damage models/laws that allow a given bonding solution to be quickly analyzed by numerical methods such as the FEM.

The influence of temperature on the strength of adhesive joints is an important factor to consider in the design of adhesive joints. The most significant factors that determine the strength of an adhesive joint when used over a wide temperature range are the coefficients of thermal expansion (CTE), (especially when compared to the CTE of the substrates) [1], and different adhesive mechanical properties with temperature [2-6]. Studies that present experimental results of adhesive joints with structural adhesives (especially epoxies) as a function of temperature generally show a decrease in strength with increasing and decreasing temperatures [2, 7-9]. At high temperatures the cause is the low adhesive strength while at low temperatures the high thermal stresses and the brittleness of the adhesive are the origin of such behaviour. Adams *et al.* [7] studied the performance of single lap joints with epoxy adhesives at low and room temperatures. They investigated the effects of adherend mismatch, shrinkage and adhesive properties on the stress state of lap joints and found that the stresses caused by adhesive shrinkage have much less effect on the lap joint strength than those generated by the adherend thermal mismatch. Owens and Lee-Sullivan [10] tested single lap joints with a rigid and a flexible epoxy adhesive at room temperature and at -40°C in quasi-static conditions. They studied stiffness loss due to crack growth in composite-to-aluminium joints. Results showed that the joint stiffness is more affected by the response of the adherends to the test temperature than by the modulus of the thin adhesive layer.

The fracture behaviour of the adhesive bonded joints is also expected to be affected by temperature. Spingarn [11] evaluated the value of G_{Ic} of a nylon-modified epoxy adhesive in Chevron-notched specimens with aluminium alloy 2024 adherends as a function of the testing rate, adhesive thickness and temperature, using developed data reduction schemes. The experimental results showed that G_{Ic} is practically insensitive to the testing temperature up to the value of T_g of the adhesive, drastically reducing above this limit. Chai [12] investigated the influence of temperature on the fracture energy in shear and showed that the mode II

fracture energy decreased in the region $0.7 < T/T_g < 1.0$. Melcher and Johnson [13] studied the Mode I, G_{Ic} , fracture toughness of an adhesively bonded composite-composite joint in a cryogenic environment. They observed a substantial decrease in the fracture toughness at cryogenic temperature compared to room temperature. Banea *et al.* [14] used the DCB test to evaluate the temperature dependence of G_{Ic} for adhesive joints bonded with a high temperature Room Temperature Vulcanizing (RTV) silicone adhesive, covering a range of temperatures between RT to 260°C. The pure mode I cohesive laws were obtained by a direct method that consisted on the differentiation of the J -integral vs. end-opening displacement (J - w) curve. They concluded that the fracture toughness and the traction–separation laws exhibit temperature dependence. Recently, Carlberger *et al.* [15] showed in an experimental study that the fracture toughness is fairly unaffected by the temperature, from -40°C to $+80^\circ\text{C}$; (the glass transition temperature of the epoxy adhesive investigated is $+90^\circ\text{C}$) and the peak stress in peel loading decreased monotonically with increasing temperature in this temperature range. However, relatively only limited data are available relative to the critical strain energy release rate at low or high temperatures.

The majority of adhesively-bonded assemblies fracture characterization under pure mode I is performed using the DCB specimen [16-19]. In a fracture mechanics analysis of this specimen, the crack is predicted to propagate when the energy release rate for mode I crack growth (G_I) becomes equal to the toughness of the adhesive or the adhesive's critical energy release rate (G_{Ic}). The main advantages of this test method include its simplicity and the possibility to obtain the fracture toughness mathematically using the beam theory for brittle materials [20].

Several techniques can be used to derive the fracture toughness of structural adhesives from fracture characterization tests. The most common methodologies for analysis are based on Linear-Elastic Fracture Mechanics (LEFM). The Compliance Calibration Method (CCM) is based on the Irwin-Kies equation [21], requiring the calculation of the compliance, C , ($C = \delta/P$, where δ is the displacement and P is the applied load) relative to the crack length during crack growth. The Direct Beam Theory (DBT), based on elementary beam theory [22], and the Corrected Beam Theory (CBT), including the effects of crack tip rotation and deflection [23], are also available within the scope of LEFM. The Compliance-Based Beam Method (CBBM) was recently developed by de Moura *et al.* [24, 25] and is based on the crack equivalent concept, depending only on the specimen's compliance during the test.

However, the use of LEFM is not recommended when the fracture process zone is large, as the assumed stress fields are not rigorously correct on account of the large-scale plasticity

[26]. In some structures, the Fracture Process Zone (FPZ) is long in one dimension but small in the perpendicular direction. This type of process zone is sometimes due to large scale bridging (LSB) [27]. These two problems should not be analysed by the LEFM [28].

An accurate strength prediction of the adhesively bonded joints is essential to decrease the amount of expensive testing at the design stage. Since the strength and fracture toughness of adhesives are particularly sensitive to temperature, the characterization of any structural adhesive to be used in these conditions should be performed. Furthermore, it is essential the availability of damage laws of adhesives to be used under different temperatures for the design of bonded structures. The parameters that define the traction law are a fracture toughness (G_c) and a maximum stress (σ_c^0) in each fracture mode.

In this work, an experimental study is performed to evaluate the effect of the temperature on the adhesive strength and fracture toughness of a high temperature epoxy adhesive. Tensile tests and pure mode I DCB adhesive fracture toughness (G_{Ic}) tests were performed at room and high temperatures (100°C, 150°C and 200°C). The experimental determination of the cohesive parameters (the fracture toughness in pure mode I (G_{Ic}) and maximum tensile stress (σ_n^0) at different temperatures allowed the definition of cohesive laws that could be subsequently used in FEM simulations to model the fracture of bonded structures [29].

2. Experimental details

2.1. Materials

2.1.1. Adhesive

The adhesive investigated in this study was a one-component high temperature paste epoxy adhesive XN1244, supplied by Nagase ChemteX (Japan).

A key parameter in the testing of adhesive joints is the T_g of the adhesive. When the adhesively bonded joints are tested below this temperature, the adhesive will behave like a low-strain rigid material while above this temperature it will have a more rubber-like behaviour.

Dynamical mechanical thermal analysis (DMTA) method was used to determine the T_g of the adhesive. The test results are plotted in Fig. 1 for three specimens. This graph contains the storage modulus - E' and the loss factor - $\tan \delta$ for each specimen. The glass transition temperature of the XN1244 adhesive, corresponding to the $\tan \delta$ peak, is approximately 155°C.

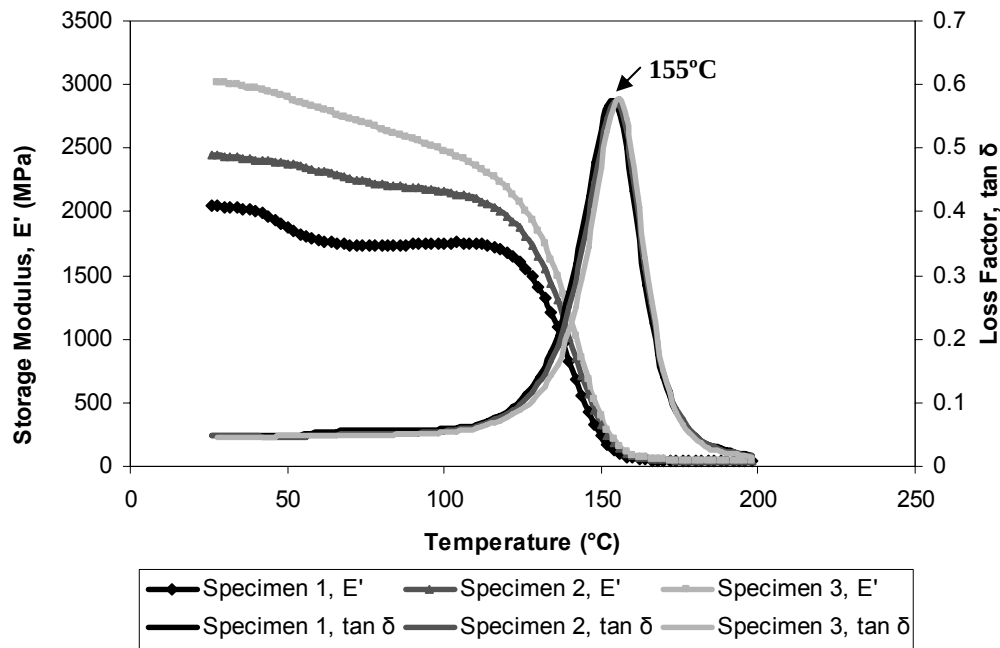


Fig. 1 – Dynamic mechanical thermal analysis results for the epoxy adhesive XN1244.

2.1.2. Substrates

Hard tool steel DIN 40CrMnMo7 substrates were used for the DCB specimens, in order to assure an elastic behaviour of the adherends. The mechanical and physical properties of the tool steel DIN 40CrMnMo7 are given in Table 1 (data provided by supplier).

Table 1. Mechanical and physical properties of steel adherends.

Tensile failure strength [MPa]	1000-1068		
Yield Stress [MPa]	861-930		
Elongation %	14-17		
	Temperature		
	20°C	200°C	400°C
Young's modulus, E [MPa]	205 000	200 000	185 000

2.2. Specimen fabrication

2.2.1. Bulk tensile specimens

Thin sheets of XN1244 adhesive were produced by curing the adhesive between steel plates of a mould with a silicone rubber frame (Fig. 2) according to the French standard NF T 76-142 [6], which were hot pressed (2MPa) for 60 minutes at 140°C. The silicone rubber frame stops the adhesive from flowing out and the pressure applied creates a good surface finish. The dimensions of the adhesive plate after cure were, 150 mm x 45 mm, with a thickness of 2 mm, which corresponds to the internal dimensions of the silicone rubber frame. “Dogbone”

specimens 2 mm thick and 10 mm wide were machined from the bulk sheet plates. The geometry of the dogbone tensile specimens used (BS 2782 standard) is shown in Fig. 3. “Dogbone” tensile specimens, before and after tests, can be seen in Fig. 4.

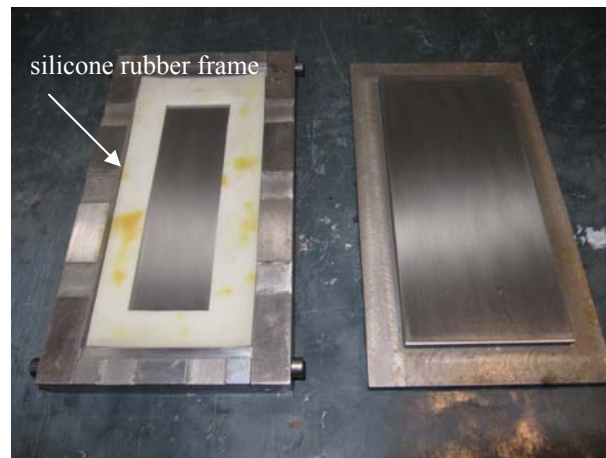


Fig. 2 – Mould for producing bulk specimens.

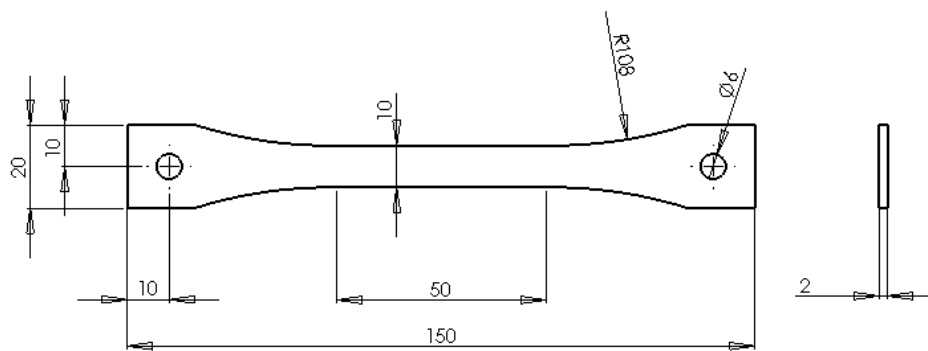


Fig. 3 – Tensile test specimen geometry bulk specimens (dimensions in mm).



Fig. 4 – Bulk specimens before and after tests.

2.2.2. DCB specimens

The DCB joint surfaces were grit blasted and degreased with acetone prior to the application of the adhesive. The specimen geometry and the loading are shown in Fig. 5.

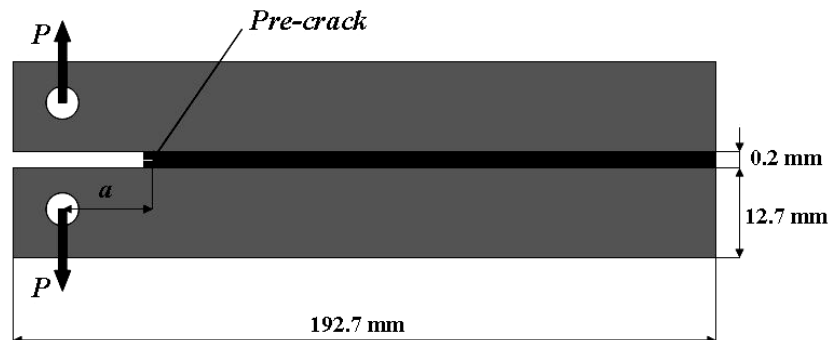


Fig. 5 – DCB specimen geometry.

The bondline thickness was nominally 0.2 mm. Spacers (calibrated steel bars of 0.20 mm) were inserted between the adherends before the application of the adhesive in order to control the bondline thickness. These spacers were removed after the adhesive was cured. A sharp pre-crack in the adhesive layer mid-thickness was assured using a razor blade. To guarantee the correct pre-crack position at the adhesive layer middle plane, a simple set with a razor blade glued in between two feeler gauges was introduced in the gap between the upper and lower adherends to promote the pre-crack. This set was done with a 0.1 mm thick razor blade glued in between two feeler gauges of 0.05 mm. A thin layer of mould release agent was applied on the surface of the feeler gauges so that the inserts could be easily removed after the adhesive curing. A mould with spacers for the correct alignment of the adherends was used and is shown in Fig. 6. The DCB joints were cured at 140°C for 1 hour.

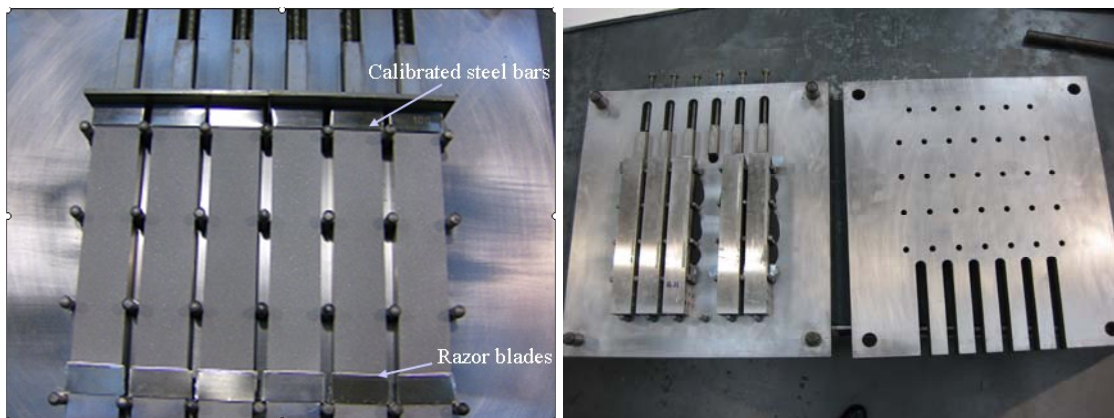


Fig. 6 – Mould with DCB specimens.

2.3. Test procedure

2.3.1. Tensile tests

The XN1244 specimens were tested in tension using a universal testing machine Shimadzu® model AUTOGRAPH, under a constant crosshead rate of 1 mm/min. A load cell of 5 kN was used. A video extensometer Messphysik ME46 was used to record the displacement. Two marks (50 mm gauge length) were painted on the specimens for the calibration and the strain measurements by the videoextensometer. For the high temperature tests, an environmental chamber of the machine was used to reach the desired temperature. At least three “dogbone” tensile specimens were tested to failure at each temperature.

2.3.2. DCB tests

The DCB specimens were tested at RT and high temperatures (100°C, 150°C and 200°C) using a universal testing machine Instron® model 8801 (Instron Co., USA), under a constant crosshead rate of 0.5 mm/min. For the high temperature tests, the environmental chamber of the machine was used to reach the desired test temperatures.

Before the testing was initiated, in order to avoid a blunt crack, all specimens were slightly loaded to ensure 2-3 mm of crack propagation, after which a_0 was measured. The load–displacement (P – δ) curve was registered during the test. Pictures were recorded during the specimens testing with 5 s intervals using a 10 MPixel digital camera.

This procedure allows measuring the crack length during its growth and afterwards collecting the P – δ – a parameters. This was performed correlating the time elapsed since the beginning of each test between the P – δ curve and each picture (the testing time of each P – δ curve point is obtained accurately with the absolute displacement and the established loading rate).

Fig. 7 shows the crack tip propagation during testing, allowing the crack length measurement. Four joints were tested to failure at each temperature. The DCB specimens set-up is shown in Fig. 8.

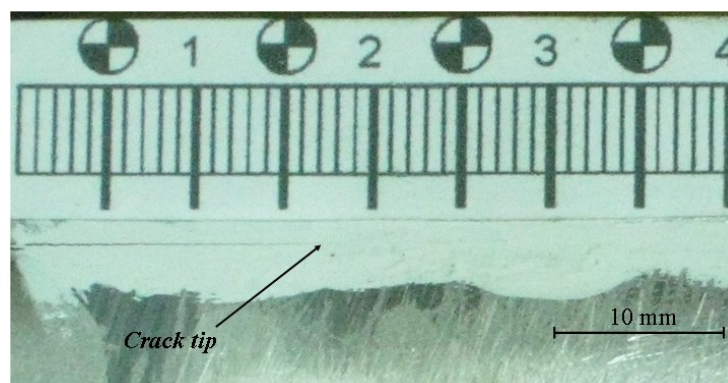


Fig. 7 – Crack length measurement during propagation in the DCB test.

A thermocouple was applied to the specimen in order to assure that the air temperature inside the chamber was equal to the specimen's temperature. The tests were always performed after approximately 10 min. of achieving the test temperature in the specimens, to ensure a steady-state temperature throughout the specimen prior to testing.



Fig. 8 – DCB specimens set-up.

2.4. DCB data analysis

Different methods were employed to evaluate the critical fracture energy in pure mode I, G_{Ic} . The Compliance Calibration Method (CCM) is based on the Irwin-Kies equation [21]:

$$G_{Ic} = \frac{P^2}{2b} \frac{dC}{da} \quad (1)$$

where P represents the load, b the specimen width and $C=\delta/P$ the compliance (δ is the specimen displacement). Cubic polynomials ($C=C_3a^3+C_2a^2+C_1a+C_0$) are used to fit the $C=f(a)$ curves, leading to:

$$G_{Ic} = \frac{P^2}{2b} (3C_3a^2 + 2C_2a + C_1) \quad (2)$$

By the Corrected Beam Theory (CBT), G_{Ic} is obtained using [23]:

$$G_{Ic} = \frac{3P\delta}{2b(a+|\Delta|)} \quad (3)$$

where Δ is a crack length correction for crack tip rotation and deflection. Δ is determined from a linear regression analysis of $(C)^{1/3}$ versus a data.

The Compliance-Based Beam Method (CBBM) was recently developed by de Moura *et al.* [24, 25] and is based on the crack equivalent concept, depending only on the specimen's compliance during the test. G_{Ic} can be obtained by the following expression:

$$G_{Ic} = \frac{6P^2}{b^2h} \left(\frac{2a_{eq}^2}{h^2E_f} + \frac{1}{5G} \right) \quad (4)$$

$a_{eq} = a + |\Delta| + \Delta a_{FPZ}$ is an equivalent crack length obtained from the experimental compliance and accounting for the fracture process zone (FPZ) at the crack tip, where a is the real crack length, Δ is the root rotation correction for the initial crack length, obtained from the linear regression of $C^{1/3} = f(a_0)$ and Δa_{FPZ} is the correction induced by the presence of the FPZ; h is the specimen height; E_f is a corrected flexural modulus to account for all phenomena affecting the P - δ curve, such as stress concentrations at the crack tip and stiffness variability between specimens, and G is the shear modulus of the adherends.

3. Experimental results and discussion

3.1. Tensile tests

The XN1244 “dogbone” specimens were tested at RT (23°C), 100°C, 150°C and 200°C. For each specimen, a tensile stress-strain curve was produced based on load, displacement values and specimen dimensions. From the stress-strain curve, the elastic modulus, ultimate tensile strength and maximum tensile strain were calculated and the results are presented in Table 2. The values for Young's modulus were calculated from the tangent to the tensile stress-strain curve at the origin (a polynomial approximation of the curve was made).

Table 2. Properties of the XN1244 adhesive.

	Young's modulus, E [GPa]	Tensile strength, [MPa]	Tensile failure strain, [%]
RT	5.87 ± 0.33	68.23 ± 5.06	1.46 ± 0.23
100 °C	4.17 ± 0.89	45.16 ± 3.44	1.93 ± 0.43
150 °C	0.07 ± 0.01	6.49 ± 0.86	13.71 ± 1.46
200 °C	0.04 ± 0.02	1.44 ± 0.17	3.33 ± 0.2

The data obtained shows a decrease in XN1244 adhesive strength and Young's modulus with increasing temperature and an increase in the ductile response of the adhesive (Table 1). As the temperature increases, the adhesive becomes more ductile, resulting in higher strain to failure and smaller strength (Fig. 9). From RT to 100°C the tensile strength moderately decreased (the adhesive is in the glassy state). At 150°C, the tensile strength decrease is more pronounced as the temperature is very close to the T_g and the epoxy adhesive is in the transition from the glassy state to the rubbery state (see the drop in the storage modulus in Fig. 1). As expected, at 200°C, when the testing temperature overpasses the T_g , a drastic fall in strength and strain was observed (the epoxy adhesive is in the rubbery state).

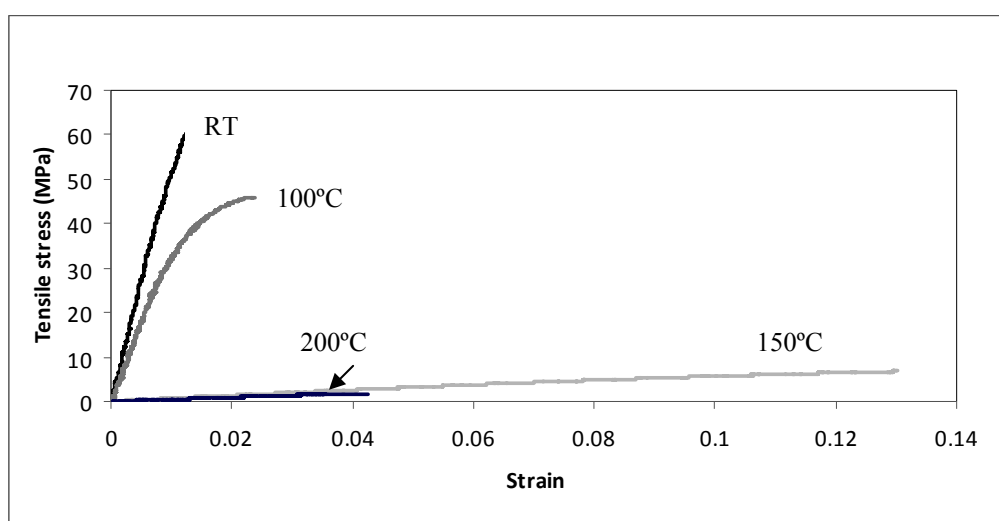


Fig. 9 – Representative XN1244 adhesive tensile stress-strain curves as a function of temperature.

3.2. Determination of G_{Ic} values as a function of temperature

Representative experimental $P-\delta$ curves of the DCB specimens at each temperature are presented in Fig. 10. The slopes and maximum loads were almost the same at RT and 100°C, but a slightly increase in displacement can be seen at 100°C. The maximum load slightly decreased at 150°C, while at 200°C, a dramatic drop in maximum load and displacement was observed.

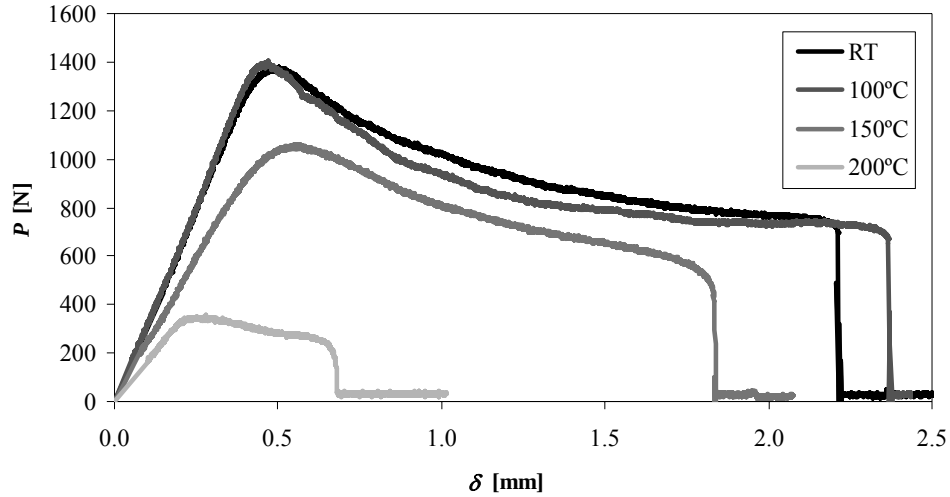


Fig. 10 – Representative experimental P – δ curves of the DCB specimens as a function of temperature.

The critical fracture energy in mode I, G_{Ic} , was evaluated using the methods presented in Section 2.4. It is known that the LEFM does not apply if the crack-tip FPZ is not small in comparison with the specimen size. The steel adherends thickness was designed to be relatively thick to prevent any plastic deformation. Also, the adherends properties at high temperatures do not significantly vary. Thus, all the nonlinear behaviour is limited in the adhesive interlayer. In the case of thin brittle adhesive layers, there is minor plastic deformation in peel [30, 31] and the fracture process length is small which enables the use of CCM, CBT and CBBM. However, the results predicted by the beam theory should be carefully examined when the adhesive is more ductile at high temperatures and the FPZ size is not too small compared to the crack length. The CZM method, with parameters carefully calibrated from the experimental load displacement curves can play the role of judge. In fact, recent results by the same authors [29] in which the cohesive properties derived in this paper were applied to numerical models of DCB joints showed that the techniques used are sufficiently accurate. To further reinforce the applicability of these techniques to the particular combination of materials and geometrical conditions investigated in this work, it should be noted that the agreement between the numerical results and the experimental results for the DCB geometry are essentially controlled by the value of toughness [32, 33].

The effect of temperature on the fracture toughness, G_{Ic} , is presented in Fig. 11. At 100°C the fracture toughness, G_{Ic} , of the adhesive slightly increased (by approximately 10%). This can be explained by the fact that, as the temperature increases, the strength decreases but the ductility increases giving an additional plastic deformation at the crack tip, hence an increase

in toughness. At 150°C, G_{Ic} is slightly lower indicating the vicinity to the T_g . However, it can be concluded that at temperatures below T_g , the fracture toughness, G_{Ic} , seems to be insensitive to temperature. Moreover, a drastic drop in fracture toughness was observed at 200°C. This was expected as the testing temperature overpasses the T_g of the adhesive.

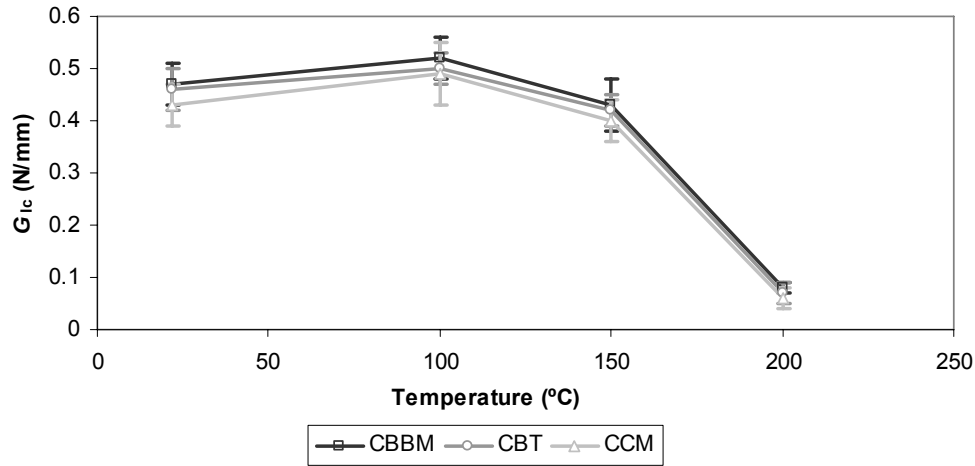
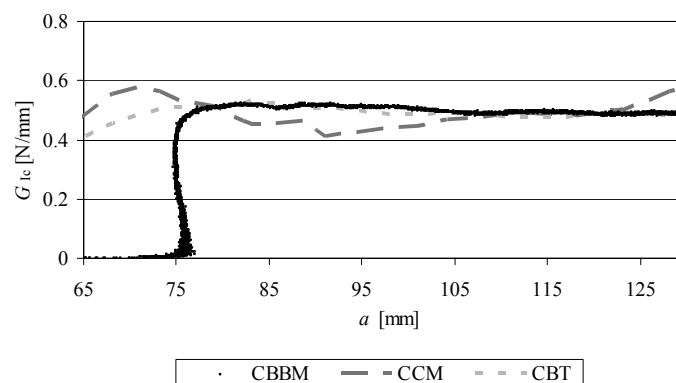


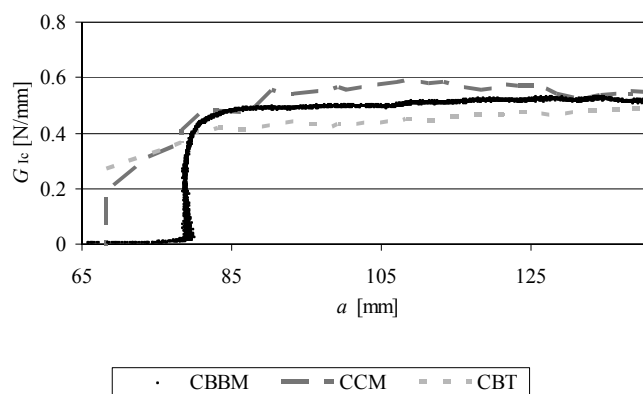
Fig. 11 – Fracture toughness G_{Ic} as a function of temperature.

Experimental R -curves obtained by the different methods for one specimen at RT are shown in Fig. 12a. Similar results were obtained by CBT and CBBM. The CCM presents a slight difference, which is explained by polynomial fitting difficulties. It should be noted that the CBBM R -curve is out of phase to the right relatively to the remaining ones, since the equivalent crack used in this method is higher than the real crack length measured during the tests and used in the other two methods.

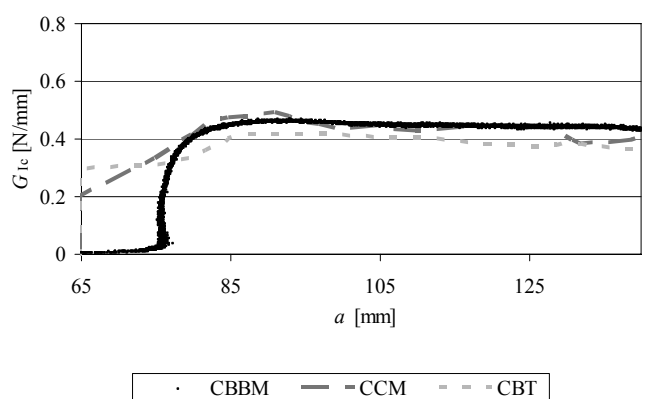
Experimental R -curves obtained by the different methods for one specimen at 100°C, 150°C and 200°C are presented in Fig. 12b, c and d. A similar degree of consistency between the three methods was found.



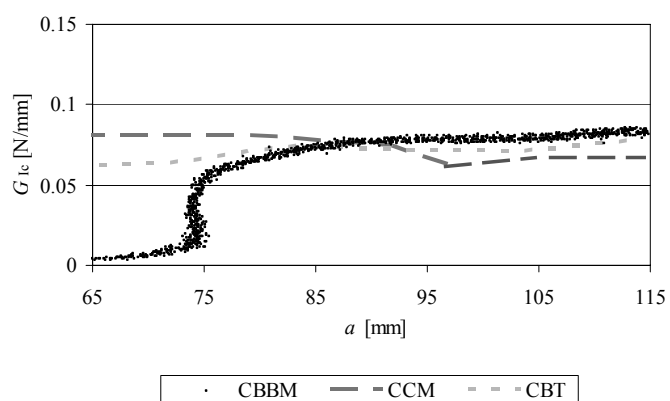
a)



b)



c)



d)

Fig. 12 – Experimental *R*-curves for one specimen at RT (a), 100°C (b), 150°C (c) and 200°C (d).

3.3 Correlation between G_{Ic} and the tensile data

As is known, the toughness is related to strength and ductility. Thus, a correlation of the mode I fracture test results with the results from tensile tests was done, in order to examine the feasibility of deducing the fracture energy of the joints from tensile material properties.

The area under the stress-strain curves from, U_T , Fig. 9 (area under the curves represents the fracture energy) was calculated for each temperature using the following formula:

$$U_T = \int_0^{\varepsilon_f} \sigma \cdot d\varepsilon \quad (5)$$

Where σ is the tensile strength, ε is the failure tensile strain and ε_f is the failure tensile strain.

Although the results show some scatter (see Fig. 13), the trends of G_{Ic} and U_T are similar, that is, at 100°C the fracture energy increased initially in relation to RT and then decreases with the increase of temperature close to T_g .

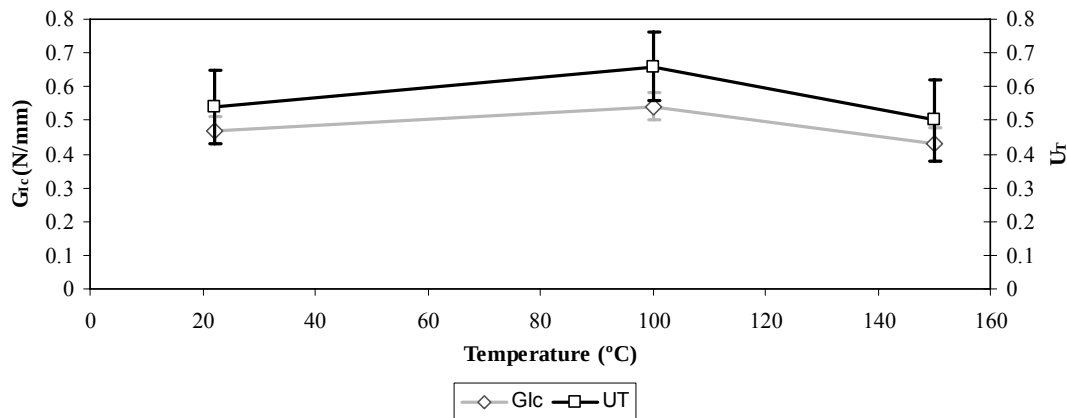


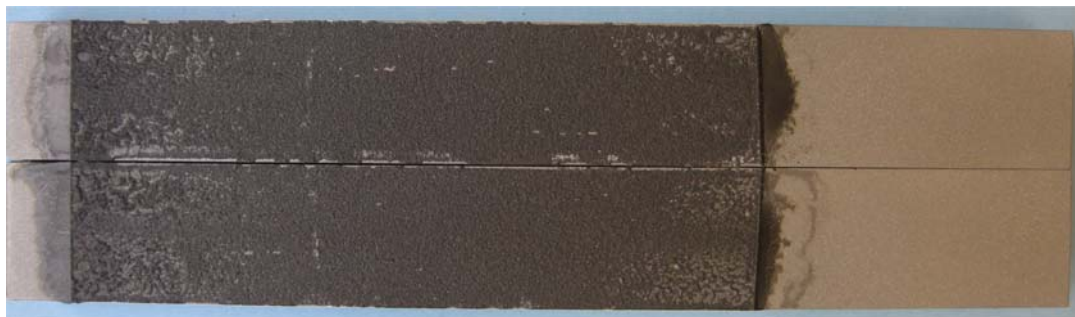
Fig. 13 – Fracture toughness vs. estimate fracture energy from the tensile stress-strain curve.

This difference might be due to the different manufacturing technique of the specimens used for these tests. As is known, different properties of adhesives from bulk form are obtained than from bonded joints. In bulk form, the adhesives have a higher sensitivity to defects such as voids and micro cracks, while in bond form the influence of the geometry of the joint must be considered. On the other hand, studying the material in adhesive bond form is advantageous in that the plastic deformation is contained.

Also, it should be noted that the results from bulk tests (the values of U_T) shows a higher scatter than the values of G_{Ic} . This can be explained by the fact that the true strain to failure is difficult to measure due to the brittleness of the adhesive in tensile tests.

3.4. DCB Failure mode

After the tests, the failure modes of the specimens were evaluated visually. As can be observed in Fig. 14 the failure in the DCB specimens was cohesive for all temperatures.



a)



b)



c)



d)

Fig. 14 – Typical failure modes at RT (a), 100°C (b), 150°C (c) and 200°C (d).

4. Conclusions

This work aimed to determine the cohesive parameters (mode I adhesive fracture toughness (G_{Ic}) and maximum tensile stress (σ_n^0)) for subsequent use in building damage laws to model crack propagation of a thin layer of a structural epoxy adhesive in bonded structures at different temperatures (RT, 100°C, 150°C and 200°C).

Tensile tests and pure mode I DCB adhesive fracture toughness (G_{Ic}) tests were performed at room and high temperatures (100°C, 150°C and 200°C). The results of the tensile tests showed that the ultimate tensile stress decreased monotonically with the temperature (showing a steep decrease between 100 and 150°C). At 200°C, as the temperature increased above the T_g of the adhesive, the adhesive strength and strain drastically reduces.

The value of G_{Ic} was found to be relatively insensitive to temperature up to T_g , while above T_g (at 200°C) a drastic decrease in G_{Ic} was found. Similar results were obtained by CBT and CBBM. The CCM presents a slight difference, which is explained by polynomial fitting difficulties.

The experimental determination of the cohesive parameters at different temperatures allows the definition of cohesive laws that could be subsequently used in FEM simulations to model the fracture of bonded structures as a function of temperature.

Acknowledgements

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**Mode I Fracture Toughness of Adhesively Bonded Joints as a
Function of Temperature – Numerical Study (XN1244)**

Mode I Fracture Toughness of Adhesively Bonded Joints as a Function of Temperature: Experimental and Numerical Study

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Abstract

Adhesives used in structural high temperature aerospace applications must operate in extreme environments. They need to exhibit high-temperature capabilities in order to maintain their mechanical properties and their structural integrity at the intended service temperature. One of the main problems caused by high temperature conditions is the fact that the adhesives have different mechanical properties with temperature. As is known, adhesive strength generally shows temperature dependence. Similarly, the fracture toughness is expected to show temperature dependence.

In this work, the Double Cantilever Beam (DCB) test is analysed in order to evaluate the effect of the temperature on the adhesive mode I fracture toughness of a high temperature epoxy adhesive. Cohesive zone models, in which the failure behaviour is expressed by a bilinear traction–separation law, have been used to define the adhesive behaviour and to predict the adhesive P – δ curves as a function of temperature. The simulation response for various temperatures matched the experimental results very well. The sensitivity of the various cohesive zone parameters in predicting the overall mechanical response as a function of temperature was examined as well for a deeper understanding of this predictive method. Also, issues of mesh sensitivity were explored to ensure that the results obtained were mesh independent.

Keywords: A. High temperature adhesives, C. fracture mechanics, D. mechanical properties of adhesives, temperature tests, cohesive zone models.

1. Introduction

In the last years, there has been a growing requirement, particularly in the aerospace industry, for adhesives to withstand high temperatures. The adhesives used in structural high temperature aerospace applications must operate in extreme environments. These adhesives have to maintain their mechanical properties at the intended service temperature and to maintain their structural integrity (resist thermal breakdown at elevated temperature). Adhesive systems that meet some of these requirements include: epoxies (having high strength and temperature resistance), silicones (excellent sealant for low stress applications, high degree of flexibility and very high temperature resistance), phenolics, polyimides, bismaleimides and ceramic adhesives.

As is known, adhesive strength generally shows temperature dependence. Studies that present experimental results of adhesive joints with structural adhesives (especially epoxies) as a function of temperature generally show a decrease in strength with increasing and decreasing temperatures [1-4]. At high temperatures this is due to the low adhesive strength, while at low temperatures the high thermal stresses and the brittleness of the adhesive are the origin of such behaviour. Similarly, the fracture toughness is expected to show temperature dependence.

Several investigators addressed the determination of the fracture toughness in tension or shear of thin adhesive layers in adhesively-bonded assemblies, but these studies are often limited to room temperature testing. Melcher and Johnson [5] studied the Mode I fracture toughness of an adhesively bonded composite-composite joint in a cryogenic environment. They observed a substantial decrease in the fracture toughness at cryogenic temperature compared to room temperature. Banea *et al.* [6] studied the pure mode I fracture toughness for adhesive joints bonded with a high temperature RTV silicone adhesive, over a wide range of temperatures (from RT to +260°C) showing that the fracture toughness and the traction–separation laws exhibit temperature dependence. Recently, Carlberger *et al.* [7] showed in an experimental study that the fracture toughness is fairly unaffected by the temperature, from –40°C to +80°C, (the T_g of the epoxy adhesive investigated is +90°C) and the peak stress in peel loading decreased monotonically with increasing temperature in this temperature range. However, relatively only limited data are available relative to the critical strain energy release rate at low or high temperatures.

Cohesive zone models (CZMs) are now widely used to simulate fracture processes of different material systems and numerous CZM models have been presented in the literature [8-10]. The key concept of CZM is that the fracture process zone (FPZ) can be described by a

traction–separation law relating stresses and relative displacements between the crack faces, thus simulating the gradual degradation of the material properties. As a result, a second fracture parameter, the cohesive strength, is introduced in addition to the fracture toughness. The shape of the softening law can also be adjusted to conform to the behaviour of the material it is simulating. Therefore, different shapes of the softening law have been employed in the literature. For example, Needleman suggested a number of different functions such as polynomial [8] and exponential [11] for traction–separation relationship, Tvergaard and Hutchinson [9] the trapezoidal model and Liljedhal et al. [12] the bilinear model. However, some researchers [9, 13] assumed that the influence of the softening law on results can be disregarded, while other researchers [14-16] emphasised that the shape of the softening law can significantly influence the response. A general review of various forms of CZM is presented in Chandra et al. [14].

A few number of techniques have been used in the past to define the traction separation laws for adhesive layers, either brittle or ductile. The most common one relies on iterative comparisons between experimentally measured data and the respective numerical predictions considering a precise description of the experimental geometry and approximate cohesive laws with a parameterized shape, established based on the typical behaviour of the material that the law is simulating. Using this technique, the value of fracture toughness, which corresponds to the plateau value of the respective *R*-curve of the fracture characterization test, is used as an input parameter in the numerical model. These models also include typical values for the cohesive strength. A few numerical iterations must be performed until a good accuracy between the experimental and numerical data is obtained. Examples of the experimental data used for fitting are the *R*-curves [17], the crack opening profile [18], and most typically the *P*- δ curve [19]. The direct method is also available, allowing the extraction of the cohesive law of an adhesive layer directly from the measured data from the experiments without requiring such a laborious numerical analysis [20]. In this method, the damage evolution across the width of the specimens has to be uniform, which is typically difficult to achieve. The direct method is based on the simultaneous measurement of the *J*-integral and the end-opening (normal or shear) of the cohesive zone. This has been successfully employed in the extraction of traction–separation laws for adhesive bonds [21,22] and fiber-reinforced composites [15].

In this work, the Double Cantilever Beam (DCB) test is analysed in order to evaluate the effect of the temperature on the adhesive mode I fracture toughness of a high temperature

epoxy adhesive. Cohesive zone models, in which the failure behaviour is expressed by a bilinear traction–separation law, have been used to define the adhesive behaviour and to predict the adhesive P – δ curves as a function of temperature. From the numerical simulations, the parameters that describe the bilinear CZM are determined. The sensitivity of the various cohesive zone parameters in predicting the overall mechanical response as a function of temperature was examined. Also, issues of mesh sensitivity were explored to ensure that the results obtained were mesh independent.

2. Experimental results

In this section, the DCB experiments results from [23] are briefly presented. The adhesive used was a one-component high temperature paste epoxy adhesive XN1244, supplied by Nagase ChemteX (Japan). The DCB specimen geometry and the loading are shown in Fig. 1. The length of the specimens was limited to 192.7 mm in order to accommodate the geometric constraints of the environmental chamber of the INSTRON[®] machine used for testing.

A key parameter in the testing of adhesive joints is the T_g of the adhesive. When the adhesively bonded joints are tested below this temperature, the adhesive will behave like a low-strain rigid material while above this temperature it will have a more rubber-like behaviour. The glass transition temperature of the XN1244 adhesive, obtained by dynamical mechanical analysis (DMA) method is approximately 155°C [23].

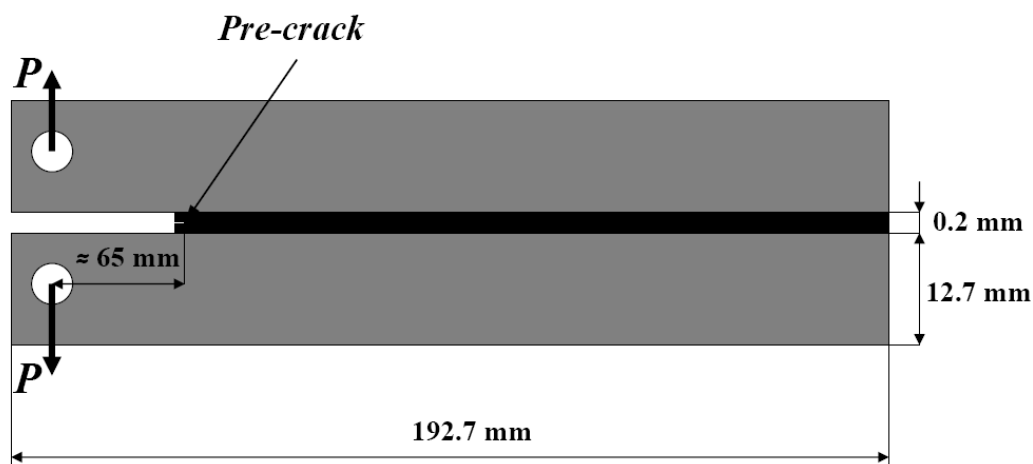


Fig. 1 – DCB specimen geometry.

Testing has been undertaken to determine the material properties of the XN1244 adhesive. The tensile properties were determined using “dogbone” specimens, produced from bulk adhesive plates cured in a steel mould, using a silicone rubber frame, according to the French standard NF T 76-142 [24]. Curing of the bulk plates was carried out in a hot press (1h at 140°C). The XN1244 specimens were tested in tension using a universal testing machine Shimadzu® model AUTOGRAPH, under a constant crosshead rate of 1 mm/min. Strains were measured by a video extensometer Messphysik ME46, along a length of 50 mm between hand-painted marks. For the high temperatures, the environmental chamber of the machine was used to reach the desired temperature. Table 1 summarizes the mechanical properties, which will be subsequently used to define the failure parameters for the finite element simulations. Fig. 2 illustrates representative XN1244 adhesive tensile stress-strain curves as a function of temperature obtained from tensile tests, which show a decrease in XN1244 adhesive strength with increasing temperature and an increase in the ductile response of the adhesive. The characterization of the mechanical properties of XN1244 adhesive has been described in more detail elsewhere [23].

Table 1. Properties of the adhesive XN1244.

Property	RT	100°C	150°C	200°C
Young's modulus, E [GPa]	5.87 ± 0.33	4.17 ± 0.89	0.07 ± 0.01	0.04 ± 0.02
Poisson's ratio, γ^*	0.35	0.35	0.35	0.35
Tensile strength, [MPa]	68.23 ± 5.06	45.16 ± 3.48	6.49 ± 0.86	1.44 ± 0.17
Tensile failure strain, [%]	1.56 ± 0.22	1.92 ± 0.42	13.71 ± 1.46	3.33 ± 0.2

* Manufacturer's data

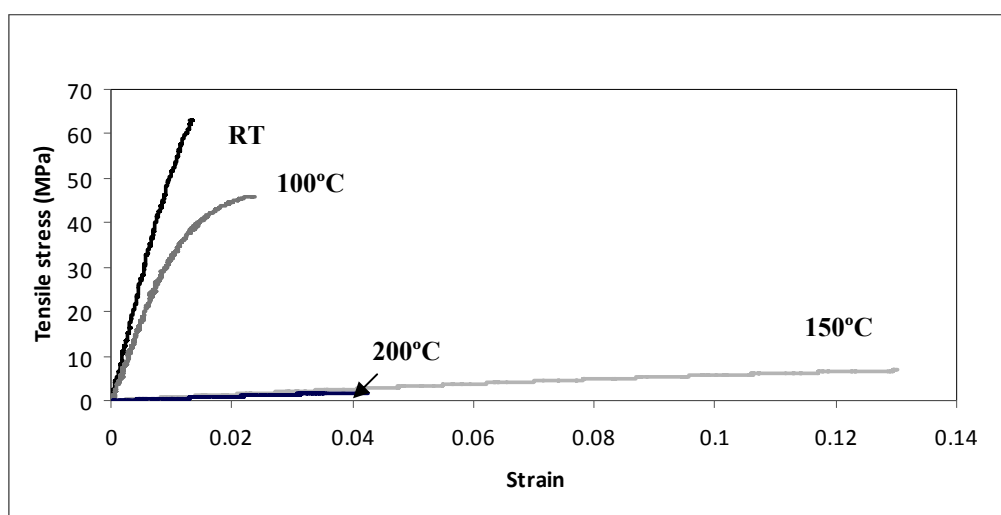


Fig. 2 – Representative XN1244 adhesive tensile stress-strain curves as a function of temperature.

Hard tool steel DIN 40CrMnMo7 substrates were used for the DCB specimens, in order to assure an elastic behaviour of the adherends during the tests. The mechanical and physical properties of the tool steel DIN 40CrMnMo7 are given in Table 2 (data provided by supplier). For the preparation of the DCB specimens, the bonding surfaces of the steel adherends were initially subjected to grit blasting and degreasing with acetone. Calibrated steel spacers of 0.20 mm were used to control the bondline thickness at 0.2 mm. The DCB specimens were cured in a hot press at 140°C for 1 hour, and subsequently were tested at RT and high temperatures using a universal testing machine Instron[®] model 8801 (Instron Co., USA), under a constant crosshead rate of 0.5 mm/min.

Table 2. Mechanical and physical properties of steel adherends.

Tensile failure strength [MPa]	1000-1068		
Yield stress [MPa]	861-930		
Elongation %	14-17		
	Temperature		
	20°C	200°C	400°C
Young's modulus, E [MPa]	205 000	200 000	185 000

Representative experimental P - δ curves of the DCB specimens at each temperature are presented in Fig. 3 [23]. The slopes and maximum loads were almost identical between RT and 100°C, but a slightly increase in displacement can be seen at 100°C. The maximum load slightly decreased at 150°C, while at 200°C, a dramatic drop in maximum load and displacement was observed.

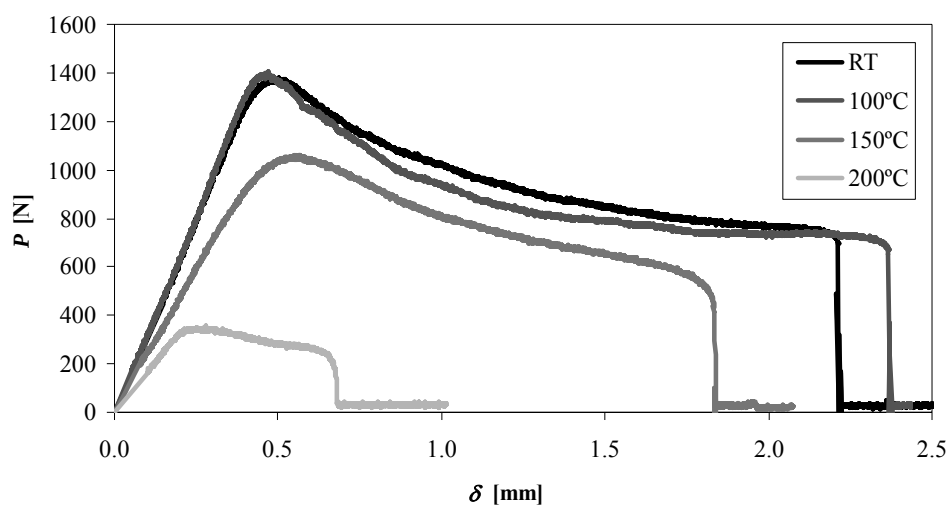


Fig. 3 – Representative experimental P - δ curves of the DCB specimens as a function of temperature.

The effect of temperature on the fracture toughness, G_{Ic} , is presented in Fig. 4 [23]. At 100°C the fracture toughness, G_{Ic} , of the adhesive slightly increased (by approximately 10%). This can be explained by the fact that, as the temperature increases, the strength decreases but the ductility increases giving an additional plastic deformation at the crack tip, hence an increase in toughness. At 150°C, G_{Ic} is slightly lower indicating the vicinity to the T_g . However, a drastic drop in fracture toughness was observed at 200°C. This was expected as the testing temperature overpasses the T_g of the adhesive.

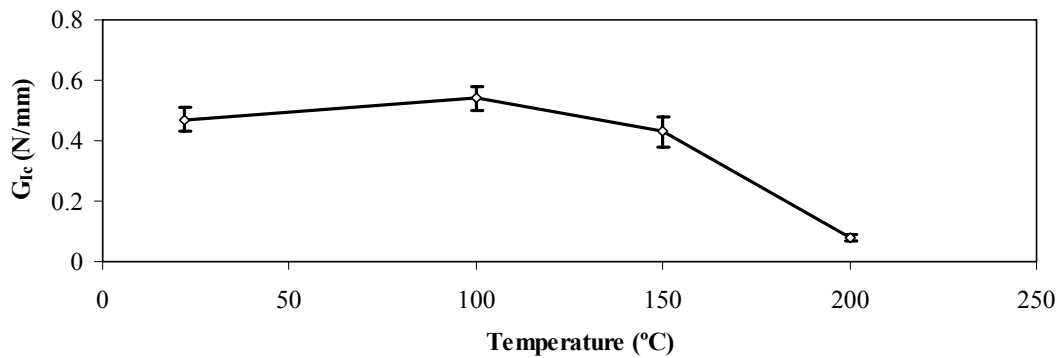


Fig. 4 – Fracture toughness G_{Ic} as a function of temperature.

3. Numerical modelling

A numerical analysis of the DCB joints as a function of temperature was performed in the commercial FEM package ABAQUS® to assess the viability of its CZM formulation, discussed further in terms of generic principles, in reproducing the experimental tests performed. The numerical analysis was carried out by two-dimensional models reproducing the whole specimen using the material properties defined in Table 3, considering geometrical non-linearities for an accurate representation of the deformation behaviour of the specimens, especially the transverse deflection and rotation of the arms. The deformed shape of the DCB specimen during damage propagation and the applied boundary and loading conditions are shown in. The specimen arms were modelled with plane-strain 8-node quadrilateral solid finite elements (CPE8 from ABAQUS®) and the adhesive was modelled with 6-node cohesive elements, including the bilinear mixed mode CZM, compatible with the 8-node quadrilateral elements. Twelve solid finite elements were used in the thickness direction for each arm, with a more refined mesh near the adhesive region [25,26]. The meshes were constructed taking advantage of the automatic meshing capabilities of ABAQUS®, using a manual seeding

procedure that included biasing effects in the horizontal direction from the loading points towards the crack tip, where large stress gradients are expected. In the damage propagation region a more refined mesh was used, considering a length of 0.2 mm for the elements in the horizontal direction. The applied boundary conditions consisted on clamping the lower edge node of the lower arm, and applying a vertical displacement and horizontally restraining the upper edge node of the upper arm (Fig. 5). This solution was adopted for consistency with the experimental tests, performed under displacement control, considering for location of the boundary and loading conditions the axis of the loading pins used for the experimental tests.

Table 3. The adhesive material properties used for simulations.

Temperature (°C)	G_n^c [N/mm]	t_n^c [MPa]	E [MPa]
RT	0.47	68	5872
100	0.50	45	4173
150	0.42	6.5	72
200	0.07	1.4	40

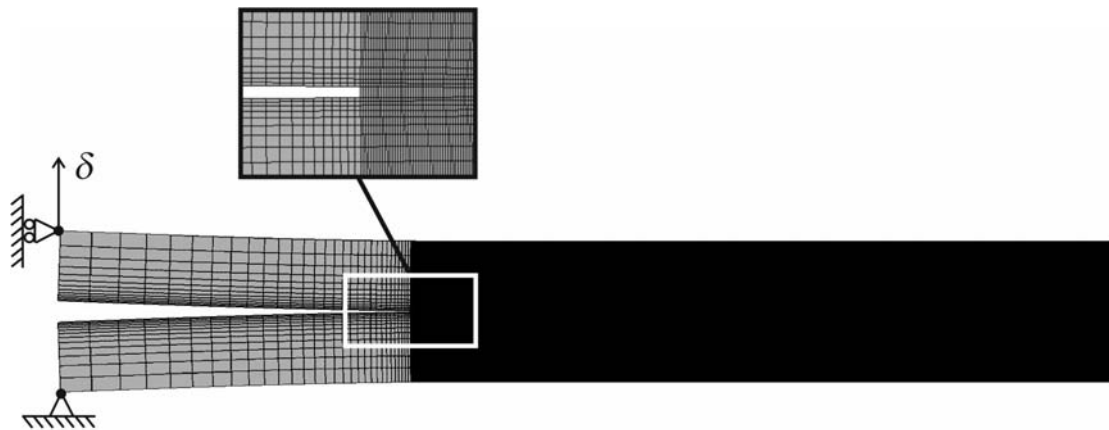


Fig. 5 – Deformed shape of the DCB specimen during propagation, with boundary and loading conditions.

3.1. Cohesive zone model

A CZM models the fracture process extending the concept of continuum mechanics by including a zone of discontinuity modelled by cohesive zones, thus using both strength and energy parameters to characterize the debonding process [27]. The choice of these parameters and how they are measured depends on the problem that is being investigated. In general, the CZM parameters are related to the material system that is being studied. The fracture

toughness can be measured from tests and then in conjunction with a FEA can be used to obtain the cohesive strength. Alternatively, both the toughness and strength can be measured from tests for subsequent use in prediction of failure of other structural configurations.

CZM's model the elastic loading, initiation of damage and propagation due to local failure within a material predicting the full description of the structures damage process up to failure. CZM's are based on a relationship between stresses and relative displacements between initially superimposed nodes of the cohesive elements (Fig. 6), simulating the elastic behaviour up to a peak load and subsequent softening, to simulate a gradual degradation of material properties. Generically speaking, the shape of the softening laws can be adjusted to conform to the behaviour of the material or interface they are simulating [28, 29].

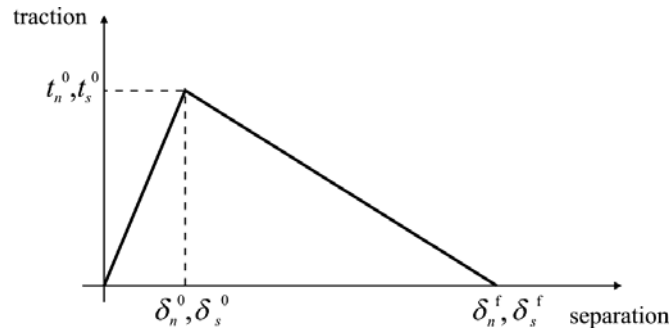


Fig. 6 – Traction-separation law with linear softening available in ABAQUS®.

The areas under the traction-separation laws in each mode of loading (tension and shear) are equalled to the respective fracture energy. Under pure mode, damage propagation occurs at a specific integration point when the stresses are released in the respective traction-separation law. Under mixed mode, energetic criteria are often used to combine tension and shear [28], thus simulating the typical mixed mode behaviour inherent to bonded assemblies. In this work, a continuum-based approach was considered to model the finite thickness of the adhesive layer. The cohesive layer is assumed to be under one direct component of strain (through-thickness) and one transverse shear strain, which are computed directly from the element kinematics. The membrane strains are assumed as zero, which is appropriate for thin and compliant layers between stiff adherends. The traction-separation law assumes an initial linear elastic behaviour followed by linear evolution of damage. Elasticity is defined by an elastic constitutive matrix relating stresses and strains across the interface [30]:

$$\mathbf{t} = \begin{Bmatrix} t_n \\ t_s \end{Bmatrix} = \begin{bmatrix} K_{nn} & K_{ns} \\ K_{ns} & K_{ss} \end{bmatrix} \cdot \begin{Bmatrix} \varepsilon_n \\ \varepsilon_s \end{Bmatrix} = \mathbf{K} \boldsymbol{\varepsilon} . \quad (1)$$

The matrix $\mathbf{K}$ contains the stiffness parameters of the adhesive layer, given by the quotient between the relevant elastic modulus and the adhesive thickness. A suitable approximation for thin adhesive layers is provided with $K_{nn}=E$, $K_{ss}=G$, $K_{ns}=0$; E and G are the longitudinal and transverse elastic moduli [28]. Damage initiation can be specified by different criteria. In this work, the quadratic nominal stress criterion was considered for the initiation of damage, already shown to give accurate results [29], expressed as [30]:

$$\left\{ \frac{\langle t_n \rangle}{t_n^0} \right\}^2 + \left\{ \frac{t_s}{t_s^0} \right\}^2 = 1. \quad (2)$$

t_n^0 and t_s^0 represent the pure mode (normal or shear, respectively) peak values of the nominal stress. $\langle \rangle$ are the Macaulay brackets, emphasizing that a purely compressive stress state does not initiate damage. After the peak value in Fig. 6 is attained, the material stiffness is degraded under different possible laws, depending on the material to be simulated. For brittle materials such as the XN1244, a linear softening law is sufficiently appropriate [16]. Complete separation is predicted by a linear power law form of the required energies for failure in the pure modes [30]:

$$\frac{G_n}{G_n^c} + \frac{G_s}{G_s^c} = 1. \quad (3)$$

The quantities G_n and G_s relate to the work done by the traction and corresponding relative displacements in the normal and shear directions, whilst the relating critical fracture energies required for pure mode failure are given by G_n^c and G_s^c for normal and shear loadings, respectively.

In this work, the shape of the traction–separation law is defined as a bilinear model. Therefore, only two parameters need to be determined (G_n^c and t_n^0), typically determined experimentally.

The t_n^0 can also be extracted by comparing numerical predictions from cohesive-zone simulations to the results of experimental tests.

The maximum relative displacement, δ_n^f , for which complete failure occurs, is obtained by equating the area under the softening curve (the triangle of Fig. 6) to the respective critical fracture energy (Eq.4).

$$G_n^c = \frac{1}{2} t_n^0 \delta_n^f \quad (4)$$

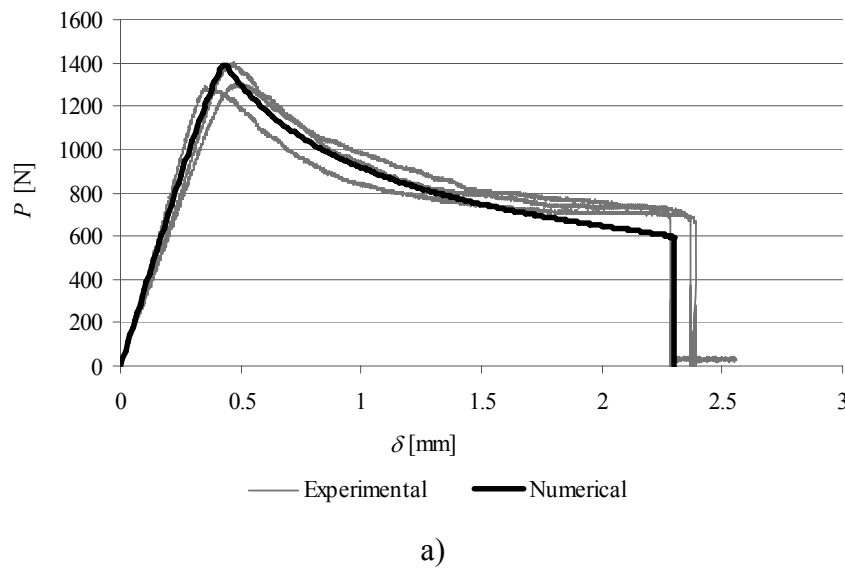
The values introduced in ABAQUS® for the simulation of damage in the adhesive layer are listed in Table 3.

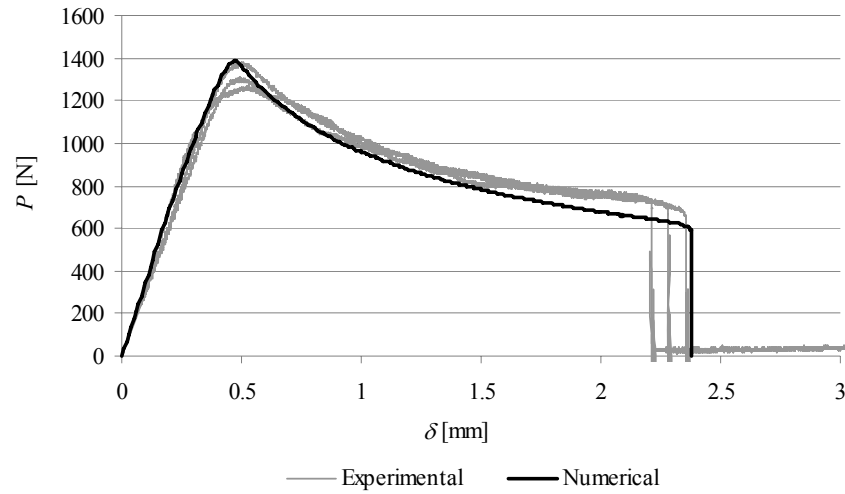
4. Numerical results and discussion

4.1. Comparison with the experiments

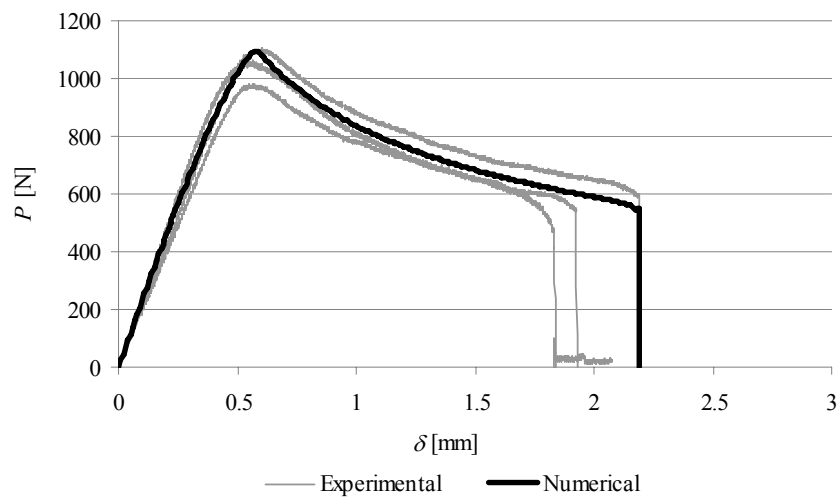
The parameters G_n^c (the fracture toughness in pure mode I, G_{Ic}) and t_n^0 (cohesive stress, σ_n^0) were defined by testing at each temperature, with the DCB tests and bulk tensile tests, respectively. The remaining cohesive parameters are obtained with an inverse method, fitting the numerical P – δ curves with the experiments and allowing complete fracture characterization of the adhesive as a function of temperature in pure mode I. This method consisted of inputting each G_n^c and t_n^0 in the respective DCB model, as a function of temperature.

In Fig. 7, experimental and numerical P – δ curves of the DCB specimens at RT (a), 100°C (b), 150°C (c) and 200°C (d) can be seen. The simulation response for various temperatures matches the experimental results very well.

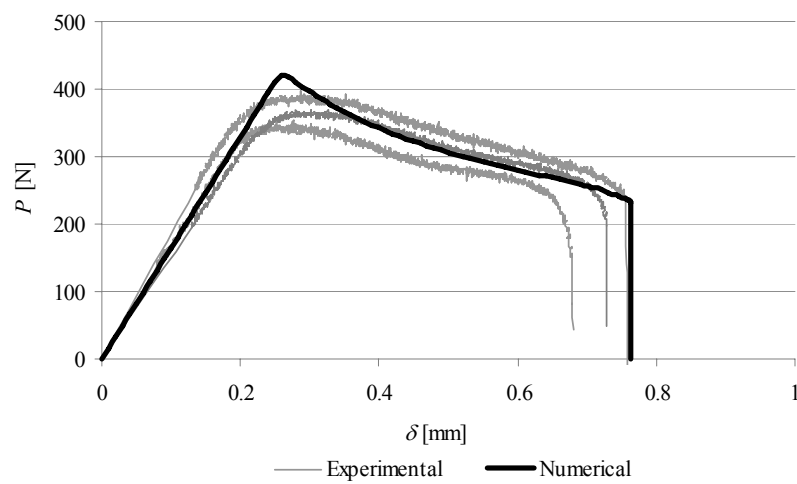




b)



c)



d)

Fig. 7 – Experimental and numerical P - δ curves of the DCB specimens at RT (a), 100°C (b), 150°C (c) and 200°C (d).

Fig. 8 presents the experimental and numerical R -curves (obtained by the CBBM method [31]) for one specimen at RT, 100°C, 150°C and 200°C, showing crack growth at a nearly constant value of G_{Ic} . An excellent agreement was found between the experimental and numerical R -curves. In Fig. 8, a_{eq} is an equivalent crack length obtained from the experimental compliance and accounting for the fracture process zone (FPZ) at the crack tip [23].

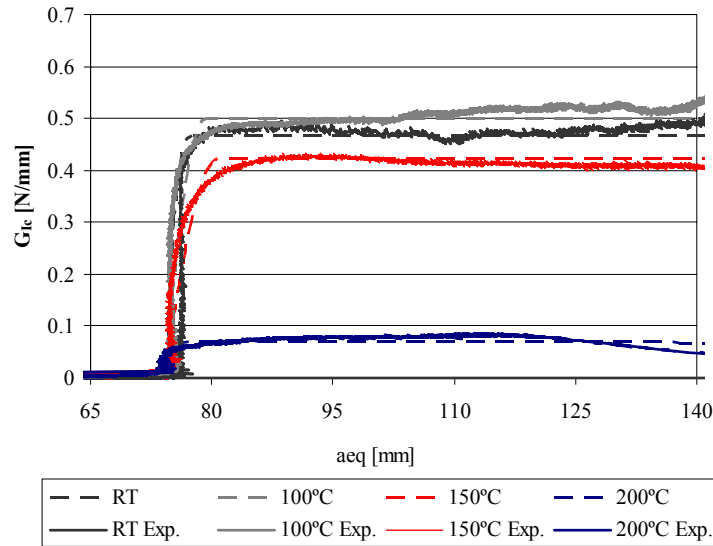


Fig. 8 – Experimental and numerical R -curves of one DCB specimen as a function of temperature.

4.2. Mesh sensitivity

In Fig. 9 a mesh dependency study is performed, to evaluate the influence of the mesh refinement for the cohesive elements representative of the adhesive layer in the global results. The DCB joint tested at RT was investigated with this purpose. In all the simulations the length of the elements were kept constant between 0.05 and 0.4 mm. $P_m/P_{m \text{ avg}}$ refers to the maximum load of the joint normalized to the average maximum load between all element sizes. Also, $G_{Ic}/G_{Ic \text{ input}}$ refers to the rate of the critical fracture energy in mode I, G_{Ic} , and the input values of fracture toughness from the numerical models, $G_{Ic \text{ input}}$. As can be seen from the Fig. 9, there are very small influences of the mesh size at the adhesive bond, which is characteristic of CZM modelling [32] as an energetic criterion, based on the fracture toughness of the material, is used for the damage growth. Since the energy required for propagation is based on the damaged area rather than a discrete value of maximum stress/strain, results are mesh independent provided that a minimum refinement is used [28].

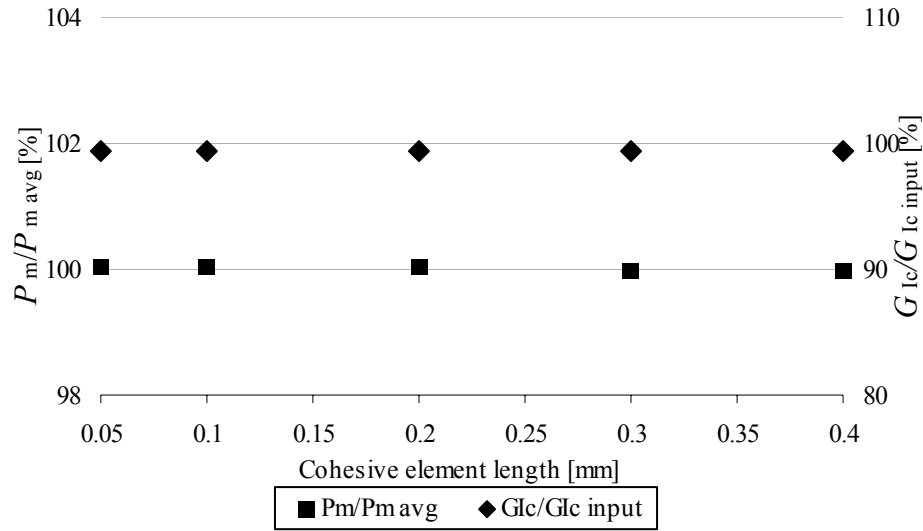


Fig. 9 – Mesh dependency study.

4.3. Sensitivity of the cohesive parameters study

It is known that the numerical predictions for different geometries and material combinations can have different sensitivities to the cohesive parameters.

With the intent of understanding the sensitivity of the DCB output to the test parameters, it is recognized that all of the DCB data reduction methods presented in [23] find that G_{Ic} is dependent on the square of the load (P^2) or maximum load P_m . As a result, the FE sensitivity study presented here is motivated by the form of Eq. 1 to 4 from [23], and uses the maximum predicted load as the output. Thus, different values of toughness (approximately $\pm 20\%$ variation of the experimental value) and strength (in the range of -90% up to $+100\%$ of the experimental value) were used for the bilinear traction–separation law shown in Fig. 6, and the resultant numerical load–displacement curves were compared to the experimental results. As can be seen from the Fig. 10a, the P – δ curve is sensitive to G_{Ic} . This is not surprising, as G_{Ic} is expected to be proportional to P (from LEFM).

Figure 10b shows that the numerical results were relatively insensitive to the value chosen for the cohesive strength of the adhesive layer, except at very low values of cohesive strength (6.8 MPa). Consequently, the cohesive strength was fixed at 68 MPa, which is the average value of all the tensile specimens tested at RT.

In summary, the agreement between the numerical results and the experimental results for the DCB geometry are essentially controlled by the value of toughness, which is consistent with the literature results [33,34].

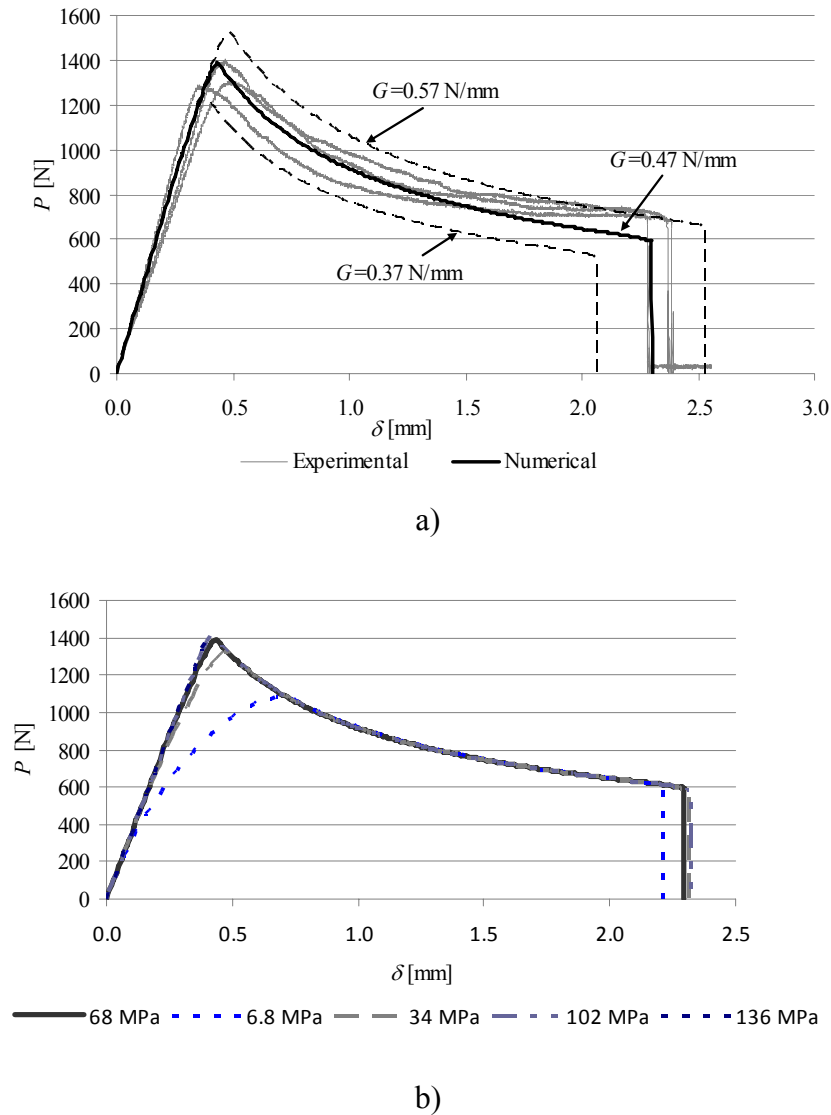


Fig. 10 – Sensitivity of the cohesive parameters study: a) toughness; b) cohesive strength.

5. Conclusions

The goal of this study was to determine the appropriate cohesive laws for pure mode I, as the first step in developing a methodology for design-level predictions of the effect of temperature on the performance of structural adhesive joints for high temperature applications.

Cohesive zone models have been used to characterize the adhesive behaviour and to predict the adhesive P – δ curves as a function of temperature. The simulation response for various temperatures matches the experimental results very well. From the numerical simulations, the parameters that describe the bilinear CZM were determined. The fracture toughness was found to be essentially temperature independent below T_g . Moreover, a drastic drop in

fracture toughness was observed at 200°C, when the T_g of the adhesive was overpassed. The cohesive strength decreases as the temperature increase.

The agreement between the numerical results and the experimental results for the DCB geometry are essentially controlled by the value of toughness and are relatively insensitive to the value chosen for the cohesive strength of the adhesive layer, except at very low values of cohesive strength.

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**Mode II Fracture Toughness of Adhesively Bonded Joints as a
Function of Temperature (XN1244)**

Mode II Fracture Toughness of Adhesively Bonded Joints as a Function of Temperature: Experimental and Numerical Study

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Abstract

In this work, an experimental and numerical study is performed to evaluate the effect of temperature on the mode II fracture toughness of a high temperature epoxy adhesive. Mode-II fracture testing on End Notch Flexure (ENF) test specimens was performed at room temperature (RT) and high temperatures (100°C, 150°C and 200°C) and the mode II fracture toughness G_{IIc} as a function of temperature was obtained. It is shown that at temperatures well below the glass transition temperature, the fracture toughness, G_{IIc} , increases with temperature and decreases as the temperature approaches T_g , while above T_g a drastic decrease in fracture toughness was observed. Furthermore, cohesive zone models, in which the failure behaviour is expressed by a bilinear traction–separation law, were used to define the adhesive behaviour in mode II and to predict the adhesive load-displacement (P – δ) curves as a function of temperature. The simulation response matched the experimental results very well at temperatures below T_g . The sensitivity of the various cohesive zone parameters in predicting the overall mechanical response of the joint was examined as well for a deeper understanding of this predictive method.

Keywords: High temperature adhesives, mode II fracture toughness, temperature tests, End Notch Flexure (ENF) test specimen.

1. Introduction

It is fundamental for the design of adhesively bonded joints the availability of reliable damage models to predict their fracture behaviour. Therefore, in order to apply a fracture mechanics or damage mechanics approach, it is essential to have the fracture toughness of the material. The fracture toughness varies with the type of loading, i.e., modes I, II, III, and mixed. In addition, the adhesives used in structural high temperature aerospace applications must operate in extreme environments and have to maintain their mechanical properties at elevated temperatures. Hence, their high-temperature mechanical performances, especially fracture toughness, should be taken into consideration.

It was reported that the adhesive strength generally shows temperature dependence [1-6]. Similarly the fracture toughness is expected to show temperature dependence. Several investigators addressed the determination of the fracture toughness in tension (pure mode I) of thin adhesive layers in adhesively-bonded assemblies as a function of temperature [7-12]. Melcher and Johnson [7] studied the mode I fracture toughness, G_{Ic} , of an adhesively bonded composite-composite joint in a cryogenic environment. They observed a substantial decrease in the fracture toughness at cryogenic temperature compared to room temperature (RT). Banea *et al.* [8] used the double cantilever beam (DCB) test to evaluate the temperature dependence of the mode I fracture toughness G_{Ic} for adhesive joints bonded with a high temperature Room Temperature Vulcanizing (RTV) silicone adhesive, covering a range of temperatures between RT to 200°C. The pure mode I cohesive laws were obtained by a direct method that consisted on the differentiation of the J -integral vs. end-opening displacement (J - w) curve. They concluded that the fracture toughness and the traction–separation laws exhibit temperature dependence. Carlberger *et al.* [9] showed in an experimental study that the mode I fracture toughness is fairly unaffected by temperature, from -40°C to $+80^{\circ}\text{C}$; (the glass transition temperature of the epoxy adhesive investigated was $+90^{\circ}\text{C}$) and the peak stress in peel loading decreased monotonically with increasing temperature in this temperature range. More recently, Banea *et al.* [11,12] performed an experimental and numerical study to evaluate the effect of temperature on the mode I fracture toughness of a high temperature epoxy adhesive. They found that the fracture toughness of the epoxy adhesive investigated is essentially temperature independent below T_g ($T_g \approx 155^{\circ}\text{C}$), while above the T_g of the adhesive (at 200°C), a drastic drop in fracture toughness was observed.

Chai [13] investigated the influence of temperature on the fracture energy in shear and showed that the mode II fracture energy decreased in the region $0.7 < T/T_g < 1.0$. However, only limited data are available relative to the effect of the temperature on the fracture toughness for pure mode II. One of the reasons is that no joint geometry has been standardised for mode II testing. On the other hand, in mode II tests crack length monitoring is difficult, as propagation occurs rapidly and without a clear opening. In addition, identification of the crack tip can be ambiguous, due to micro-cracks in the relatively large fracture process zone (FPZ) [14]. Therefore, there is growing interest in equivalent or effective crack methods [15-18], which consists in using the experimental compliance and a beam theory based relation to compute the crack length.

For the determination of the toughness in mode II there are various test methods available: the end notched flexure (ENF) test, the end loaded split (ELS) test and the four-point notched flexure (4ENF) test. The ELS test presents large displacements and is sensitive to the clamping device [15]. The 4ENF is more sophisticated but has problems of friction due to the loading mode in the pre-crack region [19]. Therefore, the most suitable testing method for mode II appears to be the ENF test [16,20]. The ENF test is essentially a three-point flexure test on a pre-cracked specimen causing a shear mode loading in the adhesive.

Most of the results in the literature concerning the effect of temperature on the fracture toughness are for mode I. However, adhesive joints are also loaded in mode II and under mixed mode. In this work, an experimental and numerical study is performed to evaluate the effect of the temperature on the mode II fracture toughness of a high temperature epoxy adhesive. Mode-II fracture testing on ENF test specimens were performed at RT and high temperatures (100°C, 150°C and 200°C) and the mode II fracture toughness G_{IIc} as a function of temperature was obtained. Furthermore, cohesive zone models, in which the failure behaviour is expressed by a bilinear traction–separation law, were used to define the adhesive behaviour in pure mode II and to predict the adhesive load-displacement ($P-\delta$) curves as a function of temperature. The sensitivity of the various cohesive zone parameters in predicting the overall mechanical response as a function of temperature was examined as well for a deeper understanding of this predictive method.

2. Experimental details

2.1. Materials

The adhesive investigated in this study was a one-component high temperature paste epoxy adhesive XN1244, supplied by Nagase ChemteX (Japan).

A key parameter in the testing of adhesive joints is the T_g of the adhesive. When adhesively bonded joints are tested below this temperature, the adhesive behaves like a low-strain rigid material while above this temperature it has a more rubber-like behaviour. The T_g of XN1244 adhesive is approximately 155°C, obtained by dynamical mechanical thermal analysis (DMTA) method [21].

The characterization tests for XN1244 adhesive were carried out under tension and shear considering three specimens for each condition, which allowed the determination of the strengths and moduli in both loadings. The tensile properties (mode I loading) of XN1244 adhesive were determined using “dogbone” tensile specimens in a previous study [21].

The Thick Adherend Shear Test (TAST) was performed in order to measure the shear properties (mode II loading) of the adhesive according to standard ISO 11003-2:1999 [22], using steel substrates of dimensions 110 x 25 x 12 mm³ (Fig. 1). The joint surfaces were grit blasted and degreased with acetone prior to the application of the adhesive. The bondline thickness was nominally 0.7 mm and the length of the overlap test section was 5 mm. Two spacers (1.5 mm thick) were inserted in the gaps between the adherends after the application of the adhesive and prior to curing in order to provide the necessary spacing between the two adherends. These spacers were removed after the adhesive was cured. The joints were cured in a hot press following the manufacturer’s suggested curing conditions (1h at 140°C). TAST tests were performed at RT on a MTS servo-hydraulic machine, model 312.31, at a constant crosshead rate of 0.1 mm/min. The displacement was measured with two methods: a 25 mm length MTS extensometer and a non-contact method (video microscopy). As the extensometer is mounted in the metallic substrate, the extensometer measures not only the displacement of the adhesive, but also the displacement of the adherend. Therefore, it is necessary to apply a correction to the measured displacements. At the same time, a video microscopy was used to record the displacements, which gives only the adhesive displacement. The strains were calculated using the spatial correlation method developed by Chousal [23].

A characteristic shear stress-strain curve of XN1244 adhesive measured by the two methods (MTS extensometer and video microscopy) at RT is shown in Fig. 2. The XN1244 adhesive has a shear modulus of 2.15 ± 0.10 , shear failure strength of 31.61 ± 2.83 and a maximum shear

strain of 8.05 ± 1.2 .

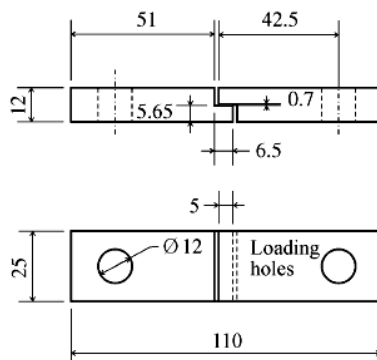


Fig. 1 Standard TAST specimen (dimensions in mm).

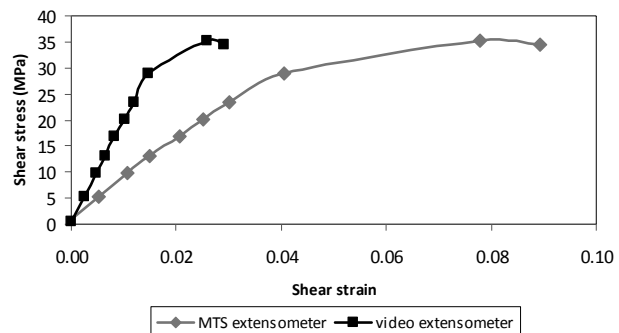


Fig. 2 Typical XN1244 adhesive shear stress-strain curve measured by the two methods (MTS extensometer and video microscopy).

Hard tool steel DIN 40CrMnMo7 substrates were used for the ENF specimens, in order to assure an elastic behaviour of the adherends. The mechanical and physical properties of the tool steel DIN 40CrMnMo7 are given in Table 1 (data provided by supplier).

Table 1. Mechanical and physical properties of steel adherends.

Tensile failure strength [MPa]	1000-1068		
Yield Stress [MPa]	861-930		
Elongation %	14-17		
	Temperature		
	20°C	200°C	400°C
Young's modulus, E [MPa]	205 000	200 000	185 000

2.2. Specimen fabrication

The ENF joint surfaces were grit blasted and degreased with acetone prior to the application of the adhesive. The specimen geometry and the loading are shown in Fig. 3. The bondline thickness was nominally 0.2 mm. Spacers (calibrated steel bars of 0.20 mm) were inserted between the adherends before the application of the adhesive in order to control the bondline thickness. These spacers were removed after the adhesive was cured. A sharp pre-crack in the adhesive layer mid-thickness was assured using a razor blade. A mould with spacers for the correct alignment of the adherends was used and is shown in Fig. 4. The ENF joints were cured at 140°C for 1 hour.

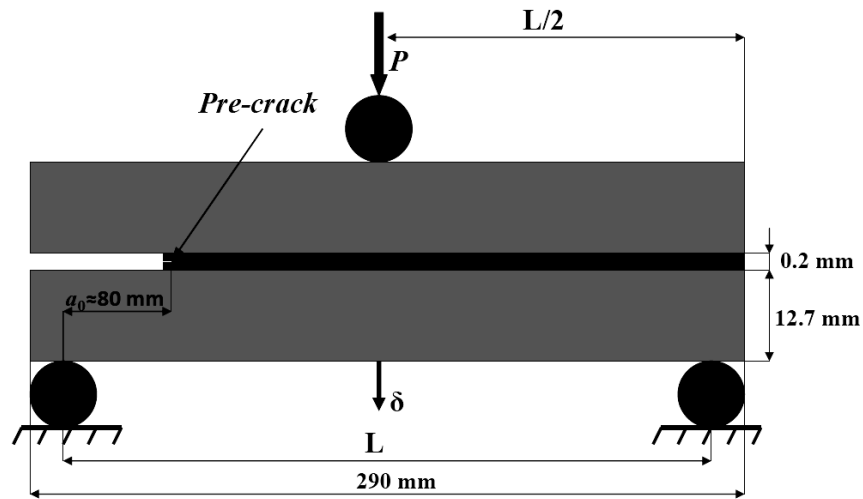


Fig. 3– ENF specimen geometry.

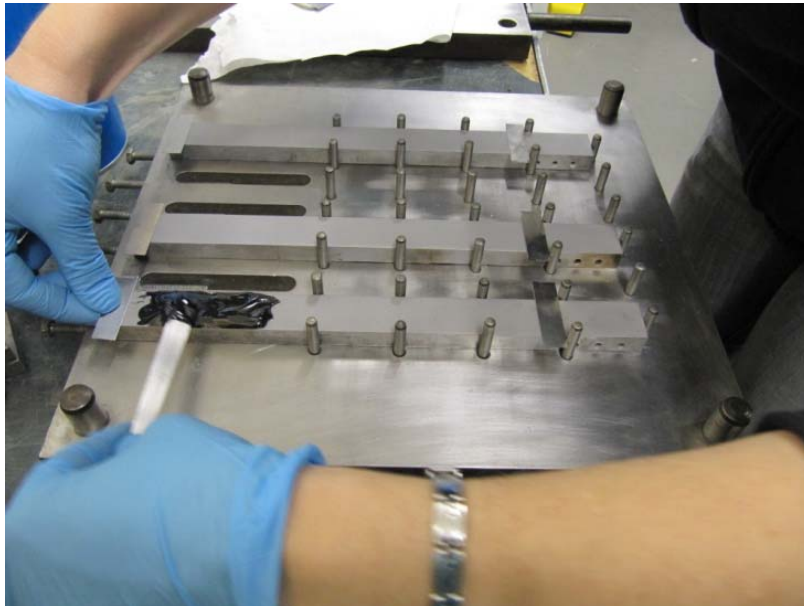


Fig. 4– Mould with ENF specimens.

2.3. Test procedure

The ENF specimens were tested at RT (approximately 30°C) and high temperatures (100°C, 150°C and 200°C) using a universal testing machine Instron® model 8801 (Instron Co., USA), under a constant crosshead rate of 0.5 mm/min. For the high temperature tests, the environmental chamber of the machine was used to reach the desired test temperatures.

Before the testing was initiated, in order to avoid a blunt crack, all specimens were slightly loaded to ensure 2-3 mm of crack propagation, after which a_0 was measured. The P – δ curve was registered during the test.

Three joints were tested to failure at each temperature. The ENF specimens set-up is shown in Fig. 5.

A thermocouple was applied to the specimen in order to assure that the air temperature inside the chamber was equal to the specimen's temperature. The tests were always performed after approximately 5 min of achieving the test temperature in the specimens, to ensure a steady-state temperature throughout the specimen prior to testing.



Fig. 5 – ENF specimens set-up.

2.4. ENF data analysis

The Compliance-Based Beam Method (CBBM) which was developed by de Moura *et al.* [18,24] was used to evaluate G_{IIc} . This method is based on the crack equivalent concept, depending only on the specimen's compliance during the test. Thus, it does not require crack length monitoring during crack growth which was observed to be very difficult to perform with accuracy in the ENF test. In addition, the equivalent crack length, a_{eq} , accounts for the FPZ effects at the crack tip, which is not taken into account when the real crack length is considered. G_{IIc} can be obtained by the following expression:

$$G_{IIc} = \frac{9P^2 a_{eq}^2}{16b^2 E_f h^3} \quad (1)$$

where P is applied load, E_f is an equivalent flexural modulus calculated from the initial compliance and initial crack length, thus avoiding the influence of specimen variability on the results; b is the specimen width; a_{eq} is the equivalent crack length and h is the adherend thickness. The readers are referred to Ref. [18] for the detailed formulations of the CBBM method.

3. Experimental results and discussion

3.1. ENF Failure mode

After the tests, the failure modes of the specimens were evaluated visually. As can be observed in Fig. 6, the failure in the ENF specimens was cohesive for all temperatures.

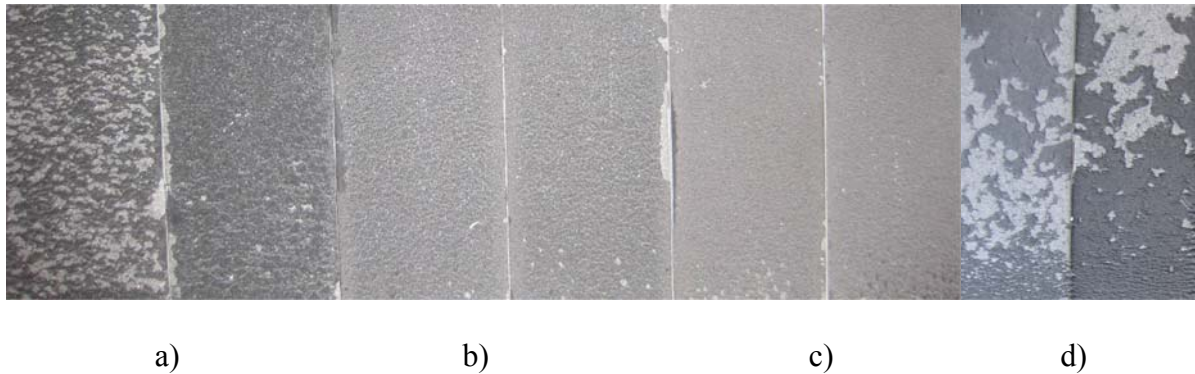


Fig. 6 – Typical failure modes at RT (a), 100°C (b), 150°C (c) and 200°C (d).

3.2. Determination of G_{IIC} values as a function of temperature

Representative experimental P – δ curves of the ENF specimens at each temperature are presented in Fig. 7. The load increases until the crack initiation process is completed. After that, the crack tip starts propagating. A continuous drop in the force is seen as the crack continuously propagates until the crack reaches the ENF specimen's middle (at the time the crack approaches the loading cylinder, the load starts to increase due to the compression near the crack tip, which obstruct propagation because of friction effects).

The maximum loads increased at 100°C and decreased at 150°C, while a significant increase in displacement can be seen at both temperatures. At 200°C, a dramatic drop in maximum load and displacement was observed.

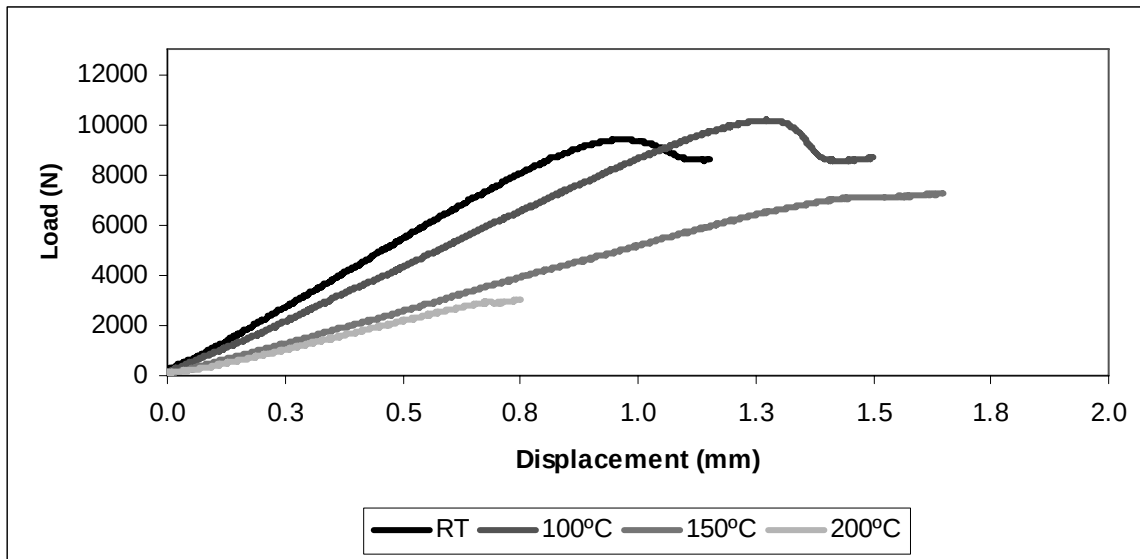


Fig. 7 – Representative experimental P – δ curves of the ENF specimens as a function of temperature.

It is known that crack propagation is controlled by the plastic deformation processes taking place ahead of the crack tip [25]. From Fig. 7 it can be seen that at 150°C and 200°C, the post-peak load reduction is significantly smaller than at 100°C and RT, which indicate an increasing plastic zone and a reduced yield stress of the adhesive.

The critical fracture energy in mode II, G_{IIc} , was estimated using the CBBM method presented in Section 2.4.

Representative experimental R -curves for one specimen at RT, 100°C, 150°C and 200°C are presented in Fig. 8. At RT and 100°C, it can be seen that after the crack initiation, as crack advances, the R -curve rise rapidly in the initial stages of crack growth and gradually reach the steady state, (a more or less stable plateau region of approximately constant G_{IIc}) after some 10 mm of crack propagation. These results are in agreement with previous studies [25,26].

At 150°C, after the crack initiation, the R -curve rises and reaches a steady state region less stable than at RT and 100°C. This can be explained by an increasing plastic zone at this temperature. After approximately 15 mm of crack propagation the R -curve starts to rise, which signifies that the crack will not propagate, but will be embedded in an increasing plastic zone.

At 200°C, the crack propagated on a very short distance (approximately 10 mm) and no initial rise of the R -curve was observed. This is explained by the fact that if the yield stress of the adhesive is very low, it will flow easily at the crack tip, offering little resistance to crack initiation.

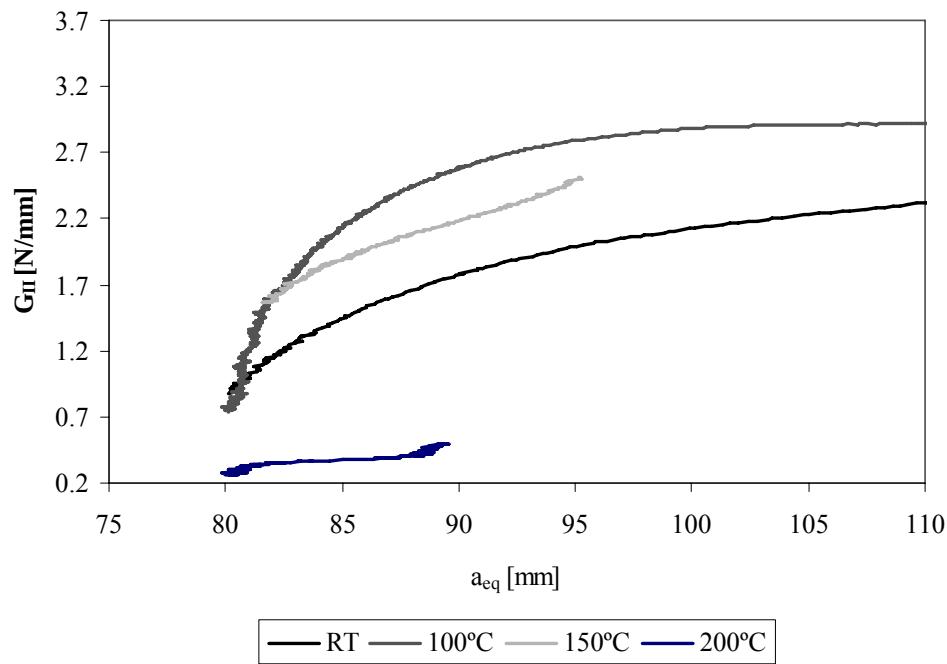


Fig. 8 – Representative experimental R -curves for one specimen at RT, 100°C, 150°C and 200°C.

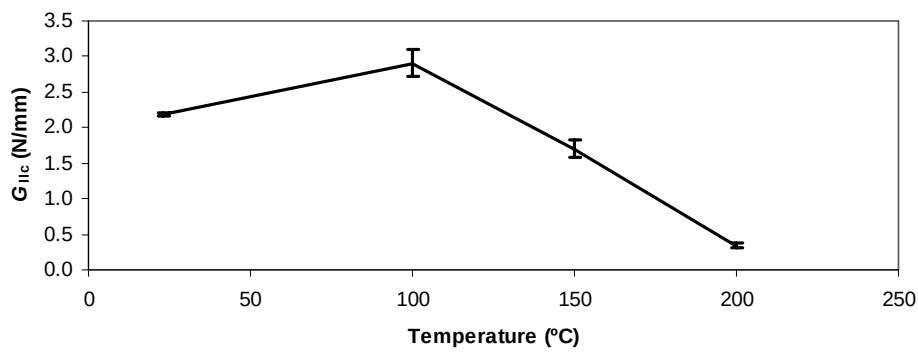


Fig. 9 – Fracture toughness G_{IIc} as a function of temperature.

The effect of temperature on the fracture toughness, G_{IIc} , is presented in Fig. 9. At 100°C the fracture toughness, G_{IIc} , of the adhesive increased by approximately 33% in relation to the value at RT. This can be explained by the fact that, as the temperature increases, the strength decreases but the ductility increases giving an additional plastic deformation at the crack tip, hence an increase in toughness. At 150°C, G_{IIc} decreased by approximately 22%, indicating

the vicinity to the T_g . These results are consistent with the results from Chai [13] which found that the mode II fracture energy decreased in the region $0.7 < T/T_g < 1.0$.

Nevertheless, a drastic drop in G_{IIc} was observed at 200°C. As is known, the value of G_{IIc} of a bonded joint is directly related to the material's ability to resist shear flow at the crack tip [27]. Thus, if the yield stress of the adhesive is very low, it will flow easily at the crack tip, offering little resistance to crack initiation. This was expected as the testing temperature overpasses the T_g of the adhesive.

3.3. Comparison of the mode I and mode II fracture characteristics

The fracture toughness measured here as a function of temperature in mode II can be compared with that measured in mode I by the same authors in a previous work [11,12]. The fracture toughness in mode I was measured using the DCB method under a test speed similar to that used in the present study. The adhesive thickness that was used is 0.2 mm, similar to the present study. The values are presented in Table 2 along with the mode II values and the relation G_{IIc}/G_{Ic} . It can be seen that the G_{IIc} follows the same trend as G_{Ic} as a function of temperature, but the increase in ductility with the increase of temperature affects more G_{IIc} as shear deformation involves more plastic work of the adhesive.

In the literature, the most common is to assume a value of 2 for G_{IIc}/G_{Ic} when the value of G_{IIc} is unknown [18,28]. On the other hand, there are studies that found higher values for G_{IIc}/G_{Ic} , such as, 14 for a brittle epoxy adhesive and 22 for a ductile adhesive [29], while in [30] the value found is approximately 10. Therefore, it is important to test not only in mode I but also in mode II for a complete and accurate characterization of an adhesive bond.

Table 2. Comparison of the fracture toughness in mode I (G_{Ic}) and mode II (G_{IIc}) as a function of temperature (average values).

Temperature (°C)	Mode I G_{Ic} (N/mm)	Mode II G_{IIc} (N/mm)	G_{IIc}/G_{Ic}
RT	0.47	2.18	4.68
100	0.50	2.90	5.80
150	0.42	1.70	4.04
200	0.07	0.34	4.25

4. Numerical modelling

A numerical analysis of the ENF joints as a function of temperature was performed in the commercial finite element (FE) package ABAQUS® to assess the viability of its cohesive zone model (CZM) formulation, discussed further in terms of generic principles, in reproducing the experimental tests performed. The numerical analysis was carried out using non-linear geometrical considerations using the material properties defined in Table 3. The deformed shape of the ENF specimen during damage propagation and the applied boundary and loading conditions are shown in Fig. 10. The specimen arms were modelled with plane-strain 4-node quadrilateral solid finite elements (CPE4 from ABAQUS®) and the adhesive was modelled with 4-node cohesive elements, including the bilinear CZM. Twelve solid finite elements were used through-thickness in each arm, with a more refined mesh near the adhesive region [31-33]. The meshes were constructed taking advantage of the automatic meshing capabilities of ABAQUS®, from a manual seeding procedure that included biasing effects from the loading points towards the crack tip, where large stress gradients are expected. In the damage propagation region and near the cylinders a more refined mesh was used, considering 0.20 mm length elements. The applied boundary conditions consisted in fixing the supporting cylinders in the directions x and y and restraining the loading cylinder in the direction x . The lowest node at the specimen mid-section was restrained in the direction x (Fig. 10). Also, at the initial crack (a_0), contact conditions were imposed to prevent interpenetration of the cracked parts.

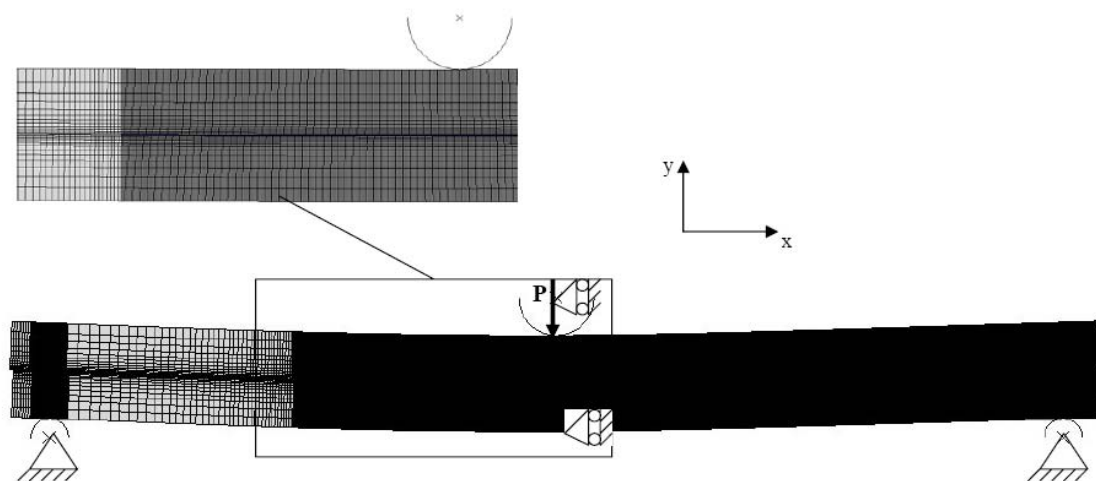


Fig. 10 – Deformed shape of the ENF specimen during damage propagation and the applied boundary and loading conditions.

4.1. Cohesive zone model

A CZM models the fracture process extending the concept of continuum mechanics by including a zone of discontinuity modelled by cohesive zones, thus using both strength and energy parameters to characterize the debonding process [34]. The choice of these parameters and how they are measured depends on the problem that is being investigated. In general, the CZM parameters are related to the material system that is being studied.

The areas under the traction-separation laws in each mode of loading (tension and shear) are equalled to the respective fracture energy. Under pure mode, damage propagation occurs at a specific integration point when the stresses are released in the respective traction-separation law. Under mixed mode, energetic criteria are often used to combine tension and shear [35], thus simulating the typical mixed mode behaviour inherent to bonded assemblies. In this work, a continuum-based approach was considered to model the finite thickness of the adhesive layer. The cohesive layer was assumed to be under one direct component of strain (through-thickness) and one transverse shear strain, which are computed directly from the element kinematics. The membrane strains are assumed as zero, which is appropriate for thin and compliant layers between stiff adherends.

In this work, the shape of the traction–separation law was defined as a bilinear model (Fig. 11). The subscripts n and s relate to pure normal (tension) and shear behaviours, respectively. t_n and t_s are the corresponding current stresses, and δ_n and δ_s the current values of δ . For pure mode II, only the “s” parameters are considered. t_s^0 can be extracted by comparing numerical predictions from cohesive-zone simulations to the results of experimental tests. The maximum relative displacement, δ_s^f , for which complete failure occurs, is obtained by equating the area under the softening curve (the triangle of Fig. 11) to the respective critical fracture energy (Eq.2).

$$G_s^c = \frac{1}{2} t_s^0 \delta_s^f \quad (2)$$

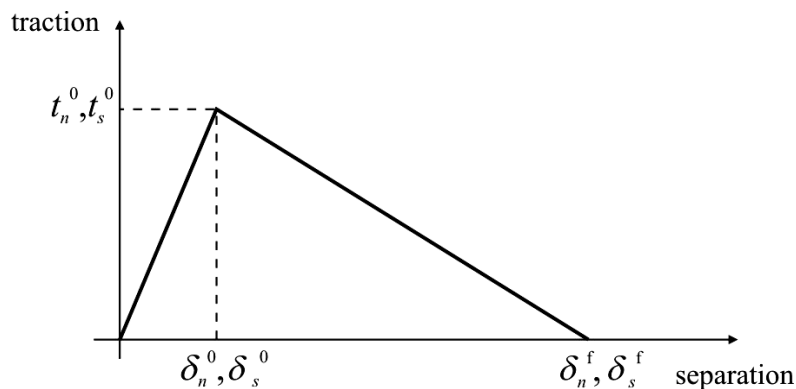


Fig. 11 – Traction-separation law with linear softening available in ABAQUS®.

5. Numerical results and discussion

5.1. Comparison with the experiments

The cohesive parameter G_s^c (the fracture toughness in pure mode II) for various temperatures was obtained experimentally from the ENF tests. The remaining cohesive parameters were obtained with an inverse method, fitting the numerical P - δ curves with the experiments and allowing complete fracture characterization of the adhesive as a function of temperature in pure mode II. This method consisted of inputting each G_s^c in the respective ENF model as a function of temperature.

The value of the Young's modulus, E , was obtained from bulk tensile tests as a function of temperature [21], while the value of shear modulus G was obtained from Thick Adherend Shear Test (TAST) at RT, and at high temperatures it was obtained analytically from the Hooke's law relation for isotropic materials:

$$G = \frac{E}{2(1+\nu)} \quad (3)$$

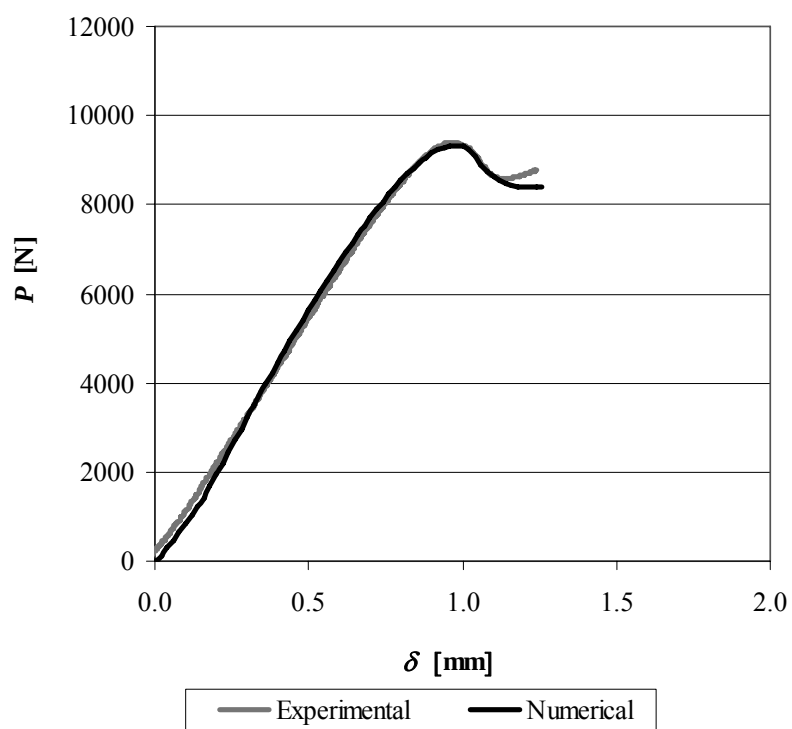
where ν is the Poisson's ratio.

The values introduced in ABAQUS[®] for the simulation of damage in the adhesive layer are listed in Table 3.

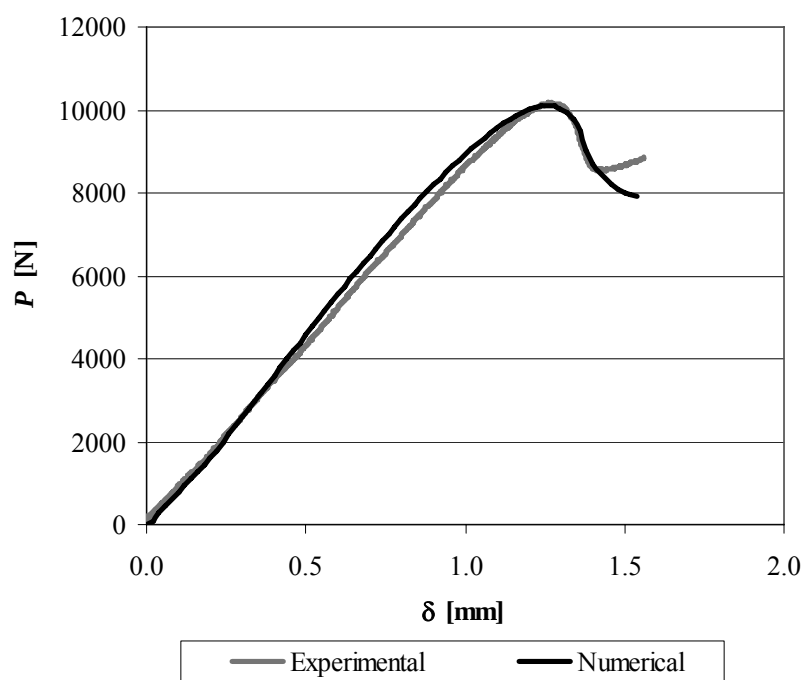
Table 3. The adhesive material properties used for simulations.

Temperature (°C)	G_s^c [N/mm]	t_s^0 [MPa]	E [MPa]	G [MPa]
RT	2.2	37	5872	2150
100	2.9	25	4173	1527
150	1.7	12	72	28
200	0.3	-	40	14.6

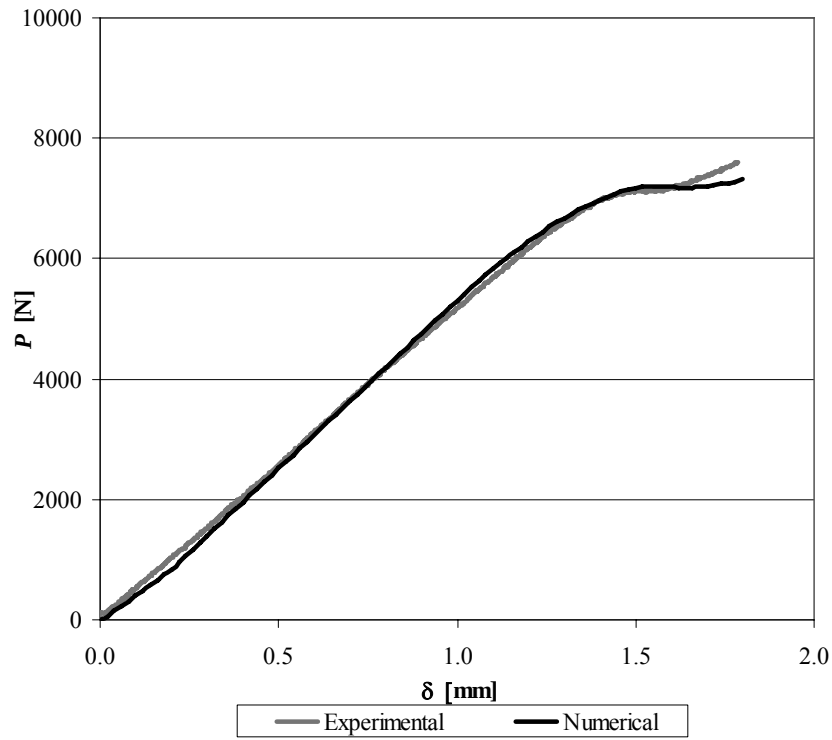
In Fig. 12, experimental and numerical P - δ curves of the ENF specimens at RT (a), 100°C (b), 150°C (c) and 200°C (d) can be seen. The simulation response at RT, 100°C and 150°C, matches the experimental results very well. However at 200°C, the model was not able to simulate the P - δ curve. This can be explained by the fact that at temperatures above T_g the behaviour of the adhesive is not completely understood and some uncertainties about the physical and the state of chemical degradation of the adhesive exists.



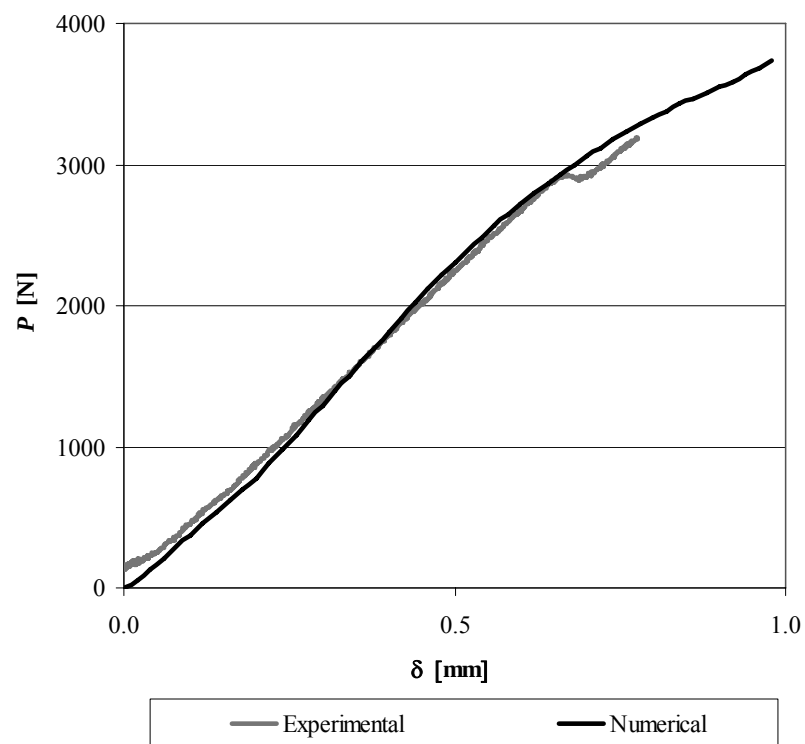
a)



b)



c)



d)

Fig. 12 – Experimental and numerical P - δ curves of the ENF specimens at RT (a), 100°C (b), 150°C (c) and 200°C (d).

5.2. Sensitivity of the cohesive parameters study

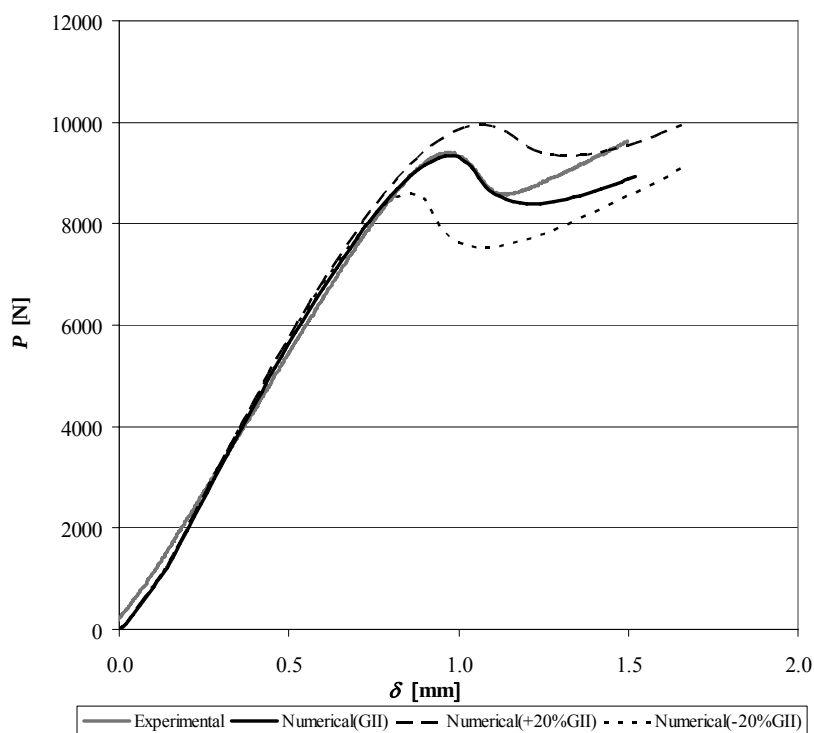
It is known that the numerical predictions for different geometries and material combinations can have different sensitivities to the cohesive parameters.

With the intent of understanding the sensitivity of the ENF output to the test parameters, it is recognized that the ENF data reduction method presented in Section 2.4. find that G_{IIc} is dependent on the square of the load (P^2). As a result, the FE sensitivity study presented here is motivated by the form of Eq. 1, and uses the maximum predicted load as the output. Thus, different values of toughness (approximately $\pm 20\%$ variation of the experimental value) and strength (in the range of $\pm 10\%$ and $\pm 50\%$ of the experimental value) were used for the bilinear traction–separation law shown in Fig. 13, and the resultant numerical load–displacement curves were compared to the experimental results.

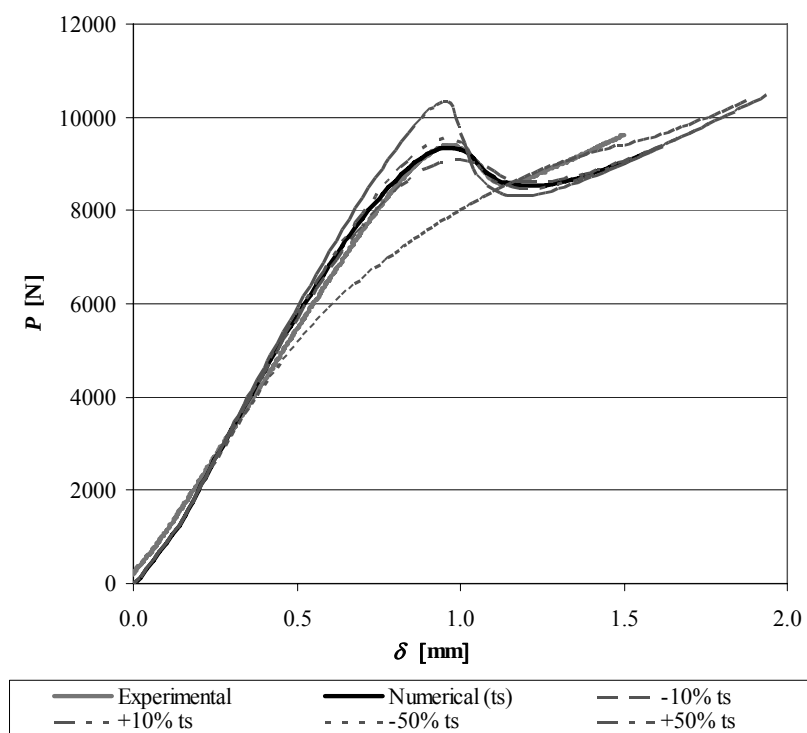
As can be seen from the Fig. 13a, the P – δ curve is sensitive to G_{IIc} . The peak load value is affected by variation of G_{IIc} . This is not surprising, as G_{IIc} is expected to be proportional to P^2 (Eq. 1).

Fig.13.b shows that the numerical results are also sensitive to the value chosen for the cohesive strength of the adhesive layer. The cohesive strength found is 37 MPa, which is different from the average value found from the TAST specimens tested at RT (32 MPa). This difference can be explained by the fact that different adhesive thicknesses were used in the tests, 0.2 mm in ENF test and 0.7 mm in TAST test.

In summary, in contrast to pure mode I loading where the agreement between the numerical results and the experimental results for the DCB geometry are essentially controlled by the value of toughness [12,36,37], for pure mode II all of the cohesive parameters influence the numerical P – δ curves profile. G_{IIc} , which is the inputted value in the numerical simulations, mainly influences the peak load value, while higher cohesive strengths increase the peak load, and the specimen stiffness up to the peak load, leading to a more abrupt post-peak load reduction.



a)



b)

Fig. 13 – Sensitivity of the cohesive parameters study: a) toughness; b) cohesive strength.

6. Conclusions

In this study, the cohesive parameters (mode II adhesive fracture toughness (G_{IIc}) and maximum shear stress) for subsequent use in building damage laws to model crack propagation of a thin layer of a structural epoxy adhesive in bonded structures at different temperatures (RT, 100°C, 150°C and 200°C) were determined.

Pure mode II ENF adhesive fracture toughness (G_{IIc}) tests were performed at room and high temperatures (100°C, 150°C and 200°C). The mode II fracture toughness was found to increase at 100°C and decrease at 150°C, as the temperature approaches T_g . Moreover, a drastic drop in fracture toughness was observed at 200°C, when the T_g of the adhesive was overpassed.

Cohesive zone models have been used to characterize the adhesive behaviour and to predict the adhesive P - δ curves as a function of temperature. The simulation response matched the experimental results very well at temperatures below T_g , while above this value of temperature it was not possible to mimic the experiments as a clear drop of load could not be related to crack propagation due to the excessive degradation of the adhesive. From the numerical simulations, the parameters that describe the bilinear CZM were determined. The agreement between the numerical results and the experimental results for the ENF geometry were essentially controlled by both values, of toughness and by the value chosen for the cohesive strength of the adhesive layer.

Acknowledgements

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Single Lap Bonded Joints (XN1244)

Effect of Temperature and Overlap Length on the Shear Strength of Aluminium Single-lap Bonded Joints for High Temperature Applications

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Abstract

An experimental and numerical investigation into the shear strength behaviour of adhesive single lap joints (SLJs) was carried out in order to understand the effect of temperature on the joint strength. The adherend material used for the experimental tests was an aluminium alloy in the form of thin sheets, and the adhesive used was a high strength high temperature epoxy. Tensile tests as a function of temperature were performed and numerical predictions based on the use of a bilinear cohesive damage model were obtained. It is shown that at temperatures below T_g , the lap-shear strength of SLJs increased, while at temperatures above T_g , a drastic drop in the lap-shear strength was observed. Comparisons between the experimental and numerical maximum loads representing the strength of the joints show a reasonably good agreement.

Keywords: High temperature adhesives, Single-lap Joints, Temperature tests, Cohesive zone model

1. Introduction

The influence of temperature on the strength of adhesive joints is an important factor to consider in the design of adhesive joints. The most significant factors that determine the strength of an adhesive joint when used over a wide temperature range are: the cure shrinkage [1], the coefficients of thermal expansion (CTE), (especially when compared to the CTE of the substrates) [2], and different adhesive mechanical properties with temperature [3-8]. However, due to the polymeric nature of adhesives, the variation of the mechanical properties of the adhesives with temperature is one of the most important factor to consider when designing a bonded joint.

Studies that present experimental results of adhesive joints with structural adhesives (especially epoxies) as a function of temperature generally show a decrease in strength with increasing and decreasing temperatures [9-15]. At high temperatures the cause is the lower adhesive strength while at low temperatures the high thermal stresses and the brittleness of the adhesive are the origin of such behaviour. Temperature also affects the fracture behaviour of adhesive joints [16-18]. However, the properties of adhesives over the range of service temperatures need to be studied for each type of application. The adhesives for aerospace applications need to withstand high temperatures, typically in excess of 150°C.

In this study, aluminium single lap joints (SLJs) were tested at room temperature (RT) and high temperatures (100°C, 125°C, 150°C, 175°C and 200°C). The behaviour of the joints was examined both experimentally and numerically. A bilinear cohesive damage model was used to obtain the numerical predictions as a function of temperature.

2. Experimental work

2.1. Adhesive selected

The adhesive investigated in this study was a one-component high temperature paste epoxy adhesive XN1244, supplied by Nagase ChemteX (Japan).

A key parameter in the testing of adhesive joints is the glass transition temperature (T_g) of the adhesive. When the adhesively bonded joints are tested below this temperature, the adhesive behaves like a low-strain rigid material while above this temperature the adhesive has a more

rubber-like behaviour. The T_g of XN1244 adhesive, obtained by dynamical mechanical thermal analysis (DMTA) method is approximately 155°C [19].

The characterization tests for the adhesive were carried out under tension (mode I loading) and shear (mode II loading) considering three specimens for each condition, which allowed the determination of the yield strengths and moduli in both loadings. The tensile properties (mode I loading) of XN1244 adhesive were determined using “dogbone” specimens, produced from bulk adhesive plates cured in a steel mould, using a silicone rubber frame, according to the French standard NF T 76-142 [20]. Curing of the bulk plates was carried out in a hot press (1h at 140°C). The XN1244 specimens were tested in tension using a universal testing machine Shimadzu[®] model AUTOGRAPH, under a constant crosshead rate of 1 mm/min. Strains were measured by a video extensometer Messphysik ME46, along a length of 50 mm between hand-painted marks. For the high temperatures, the environmental chamber of the machine was used to reach the desired temperature. Figure 1 illustrates representative XN1244 adhesive tensile stress-strain curves as a function of temperature obtained from tensile tests, which show a decrease in XN1244 adhesive strength with increasing temperature and an increase in the ductile response of the adhesive. The characterization of the mechanical properties of XN1244 adhesive has been described in more detail elsewhere [19].

The Thick Adherend Shear Test (TAST) tests for mode II loading followed the guidelines of the standard ISO 11003-2:1999 [21], using steel substrates of dimensions 110 x 25 x 12 mm³ (Figure 2). The joint surfaces were grit blasted and degreased with acetone prior to the application of the adhesive. The bondline thickness was nominally 0.7 mm and the length of the overlap test section was 5 mm. Two spacers (1.5 mm thick) were inserted in the gaps between the adherends after the application of the adhesive and prior to curing in order to provide the necessary spacing between the two adherends. These spacers were removed after the adhesive was cured. The joints were cured in a hot press following the manufacturer’s suggested curing conditions (1h at 140°C). TAST tests were performed at RT on a MTS servo-hydraulic machine, model 312.31, at a constant crosshead rate of 0.1 mm/min. The displacement was measured with two methods: a 25 mm length MTS extensometer and a non-contact method (video microscopy). As the extensometer is mounted in the metallic substrate, the extensometer measures not only the displacement of the adhesive, but also the displacement of the adherend. Therefore, it is necessary to apply a correction to the measured

displacements. At the same time, a video microscopy was used to record the displacements, which gives only the adhesive displacement. The strains were calculated using the spatial correlation method developed by Chousal [22]. A characteristic shear stress-strain curve of XN1244 adhesive measured by the two methods (MTS extensometer and video microscopy) at RT is shown in Figure 3.

Table 1 summarizes the collected data on this adhesive, which was subsequently used for the finite element simulations and strength predictions. The yield strength was calculated for a plastic deformation of 0.2%.

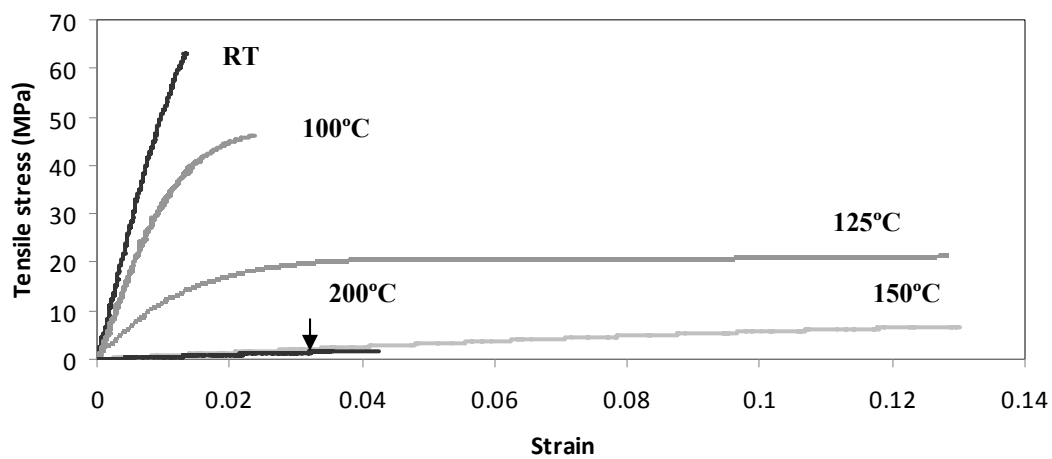


Figure 1. σ - ε curves of representative tensile bulk tests to XN1244 adhesive as a function of temperature.

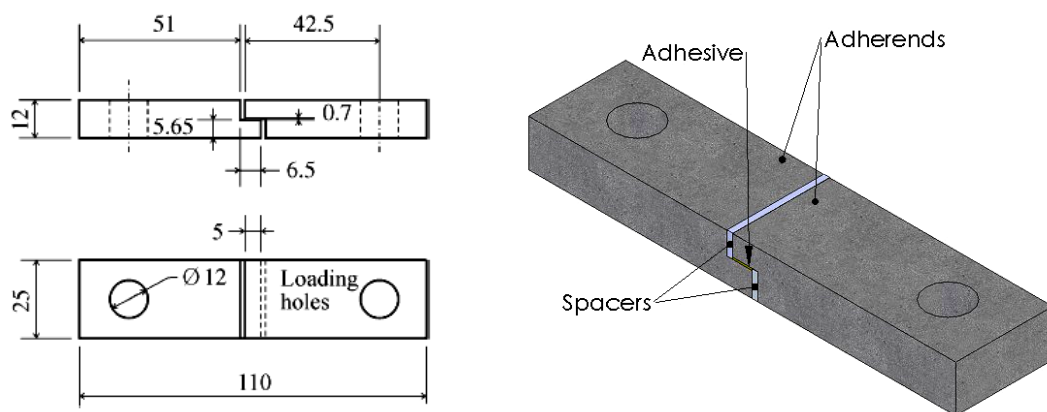


Figure 2. Standard TAST specimen (dimensions in mm).

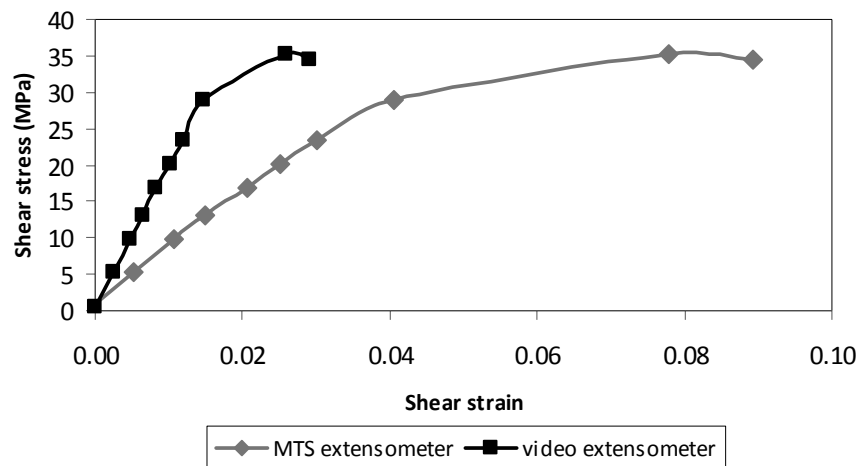


Figure 3. Typical XN1244 shear stress-strain curve measured by the two methods (MTS extensometer and video microscopy).

Table 1 – Properties of the adhesive XN1244.

Property	XN1244
Young's modulus, E [GPa]	5.87 ± 0.33
Poisson's ratio, ν^*	0.35
Tensile yield strength, σ_y [MPa]	46.90 ± 2.70
Tensile failure strength, σ_f [MPa]	68.23 ± 5.06
Tensile failure strain, ε_f [%]	1.46 ± 0.23
Shear modulus, G [GPa]	2.15 ± 0.10
Shear yield strength, τ_y [MPa]	26 ± 2.6
Shear failure strength, τ_f [MPa]	31.61 ± 2.83
Shear failure strain, γ_f [%]	8.05 ± 1.2

* Manufacturer's data

2.2. Specimens manufacture

Aluminium alloy 6082-T651 (Al Si1MgMn) substrates characterized by a high tensile strength with a thickness of 3 mm and 25 mm width were used in order to avoid plastic deformation of the adherends. The bulk stress-strain (σ - ε) response of the aluminium adherends, obtained according to ASTM-E8M-04 standard [23], is presented in Figure 4 for the three specimens tested [24]. The aluminium has a Young's modulus of 70.07 ± 0.83 GPa, a yield stress of 261.67 ± 7.65 MPa, a maximum strength of 324 ± 0.16 MPa and a failure strain of $21.70 \pm 4.24\%$. The bilinear approximation of Figure 4 was used as input in the simulations.

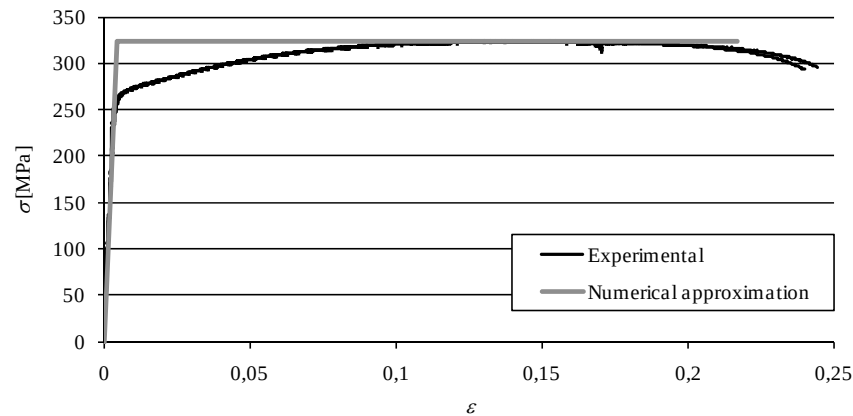


Figure 4. Experimental σ - ε curves of the aluminium AW6082 T651 and approximation for the FEM analysis.

The joint surfaces were grit blasted and degreased with acetone prior to the application of the adhesive. The substrates were bonded and then the specimens were cured in a hot press following the manufacturers' suggested curing conditions (1h at 140°C). A mould with spacers for correct alignment of the substrates was used (see Figure 5). The bondline thickness was controlled using packing shims. The bondline thickness was 0.2 mm and the length of the overlap was 25 mm. The geometry of the lap shear joint specimens used is shown in Figure 6. Aluminium tabs were glued at the specimens edges for a correct alignment in the testing machine.

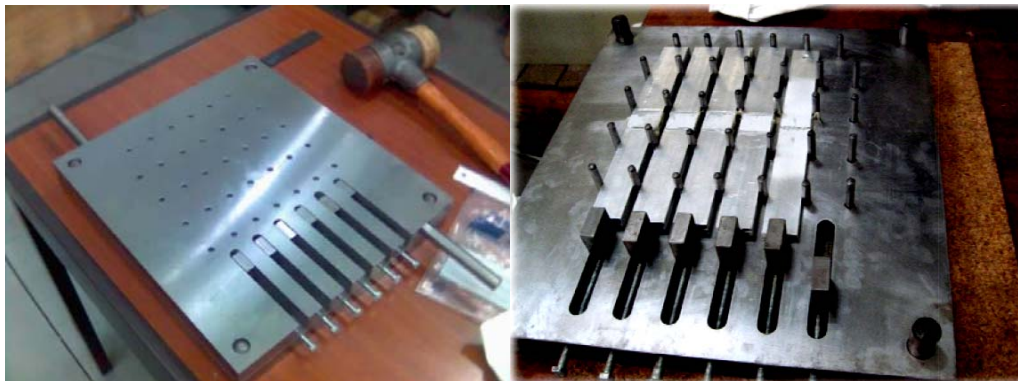


Figure 5. Mould for SLJ specimens fabrication.

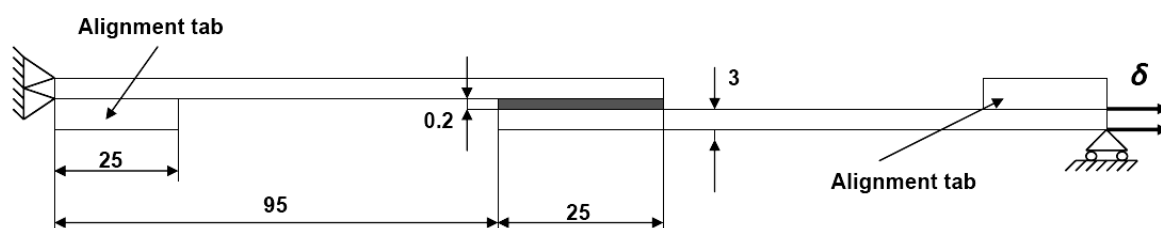


Figure 6. Single lap joint specimen geometry (dimensions in mm).

2.3. Test method

The SLJs were tested at RT and high temperatures (100°C, 125°C, 150°C, 175°C and 200°C) using a universal testing machine Instron[®] model 8801 (Instron Co., USA), under a constant crosshead rate of 1 mm/min. For the high temperature tests, the environmental chamber of the machine was used to reach the desired test temperatures.

At least three joints were tested to failure at each temperature. For each joint tested, load-displacement curves were produced. A thermocouple was applied to the specimen in order to assure that the air temperature inside the chamber was equal to the specimen's temperature. The tests were always performed after approximately 5 min. of achieving the test temperature in the specimens, to ensure a steady-state temperature throughout the specimen prior to testing.

3. Experimental results

3.1. Failure modes

After the tests, the failure modes of the specimens were evaluated visually. It is well known that for brittle adhesives the failure of the adhesive joints is dominated by stress concentrations [25]. Thus, at RT, failure of the adhesive occurs in regions of maximum stress or strain concentration. However, increasing the temperature, the adhesive becomes more ductile and there is a larger adhesive yielding which might result in a change in failure mode. At RT, cracks which run through the adhesive close to the adherend-adhesive interface meet at the middle of the overlap as can be seen in Figure 7a. For SLJs tested at 100°C (Figure 7b), the failure surface is different from that observed at RT in that main part of the adhesive inside the overlap remain on one of the adherends (there was a thin film left on the other adherend). For SLJs tested at 125°C the failure mode was a cohesive failure within the middle of the adhesive layer (Figure 7c). A similar failure mode occurred at the other temperatures (see Figure 7d, e and f). The failure surface at 175°C and 200°C was rougher which indicates a higher plastic deformation of the adhesive.

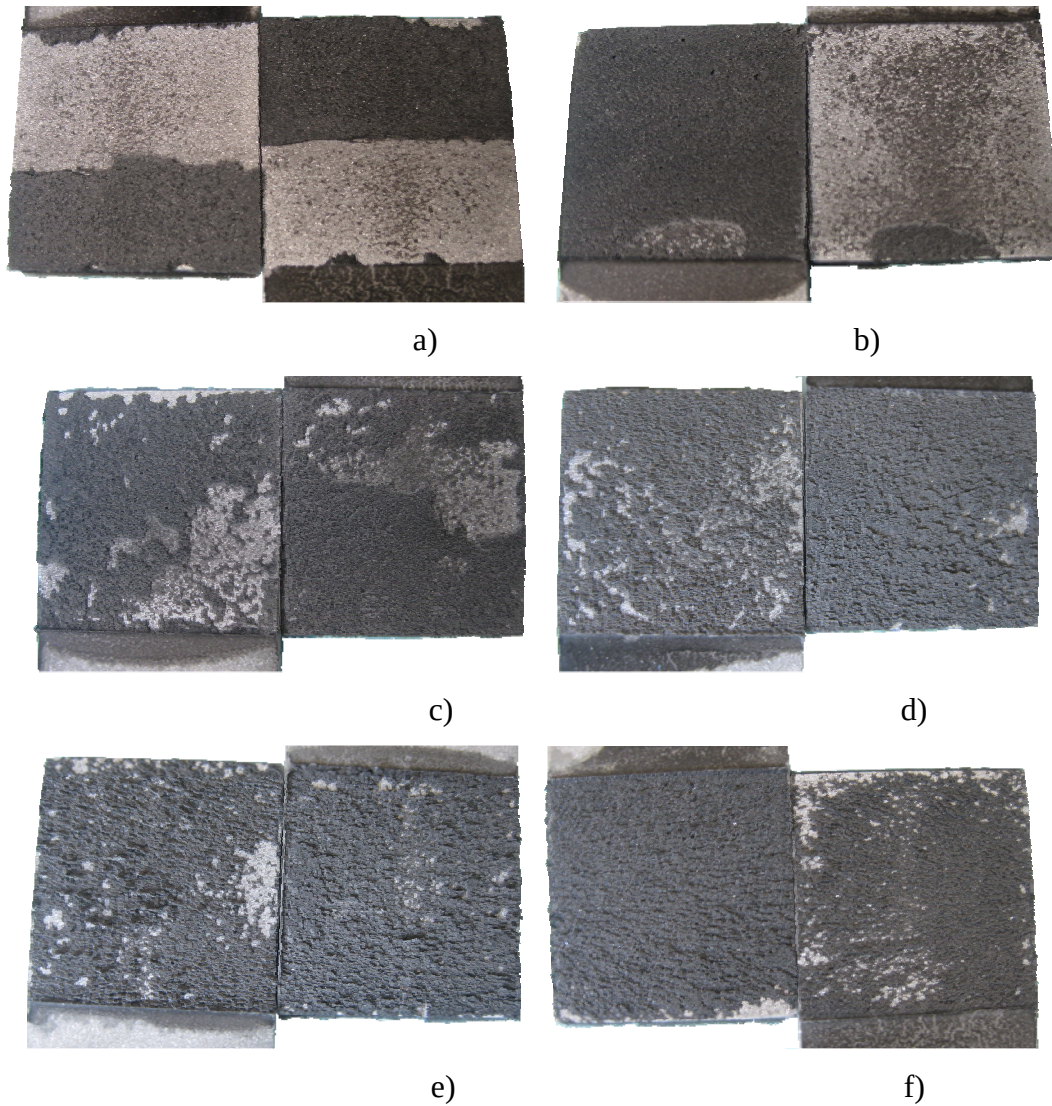


Figure 7. Failure mode of SLJs tested at a) RT, b) 100°C, c) 125°C, d) 150°C, e) 175°C and f) 200°C.

3.2. Effect of temperature

Representative load-displacement curves of XN1244 adhesive SLJs as a function of temperature are presented in Figure 8. It can be seen that the failure load increased with temperature up to 150°C, and decreased at 175°C and 200°C as the T_g of the adhesive is overpassed. The XN1244 SLJs stiffness does not substantially vary with temperature up to 175°C.

Average lap shear strengths and failure displacements of XN1244 SLJ as a function of temperature is presented in Figure 9. The average lap-shear strength (τ_{av}) is given by:

$$\tau_{av} = P/bL \quad (1)$$

where P is the maximum load, b is the joint width and L is the joint overlap length.

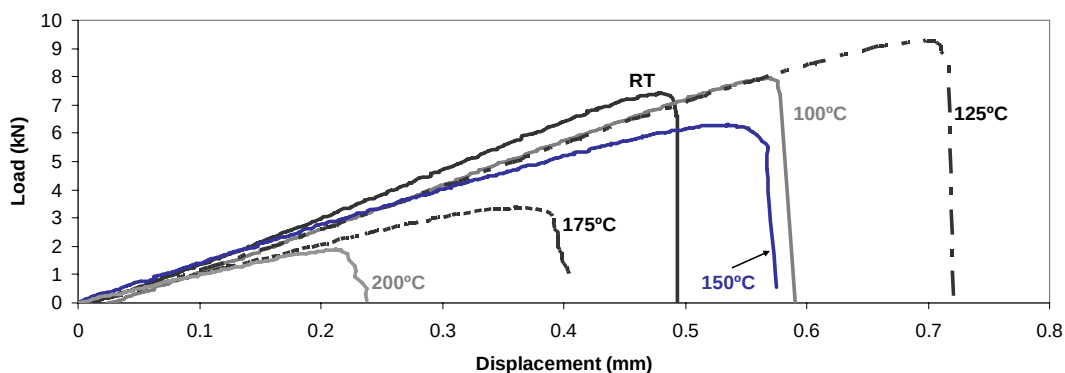


Figure 8. Representative load-displacement curves of XN1244 adhesive SLJs as a function of temperature.

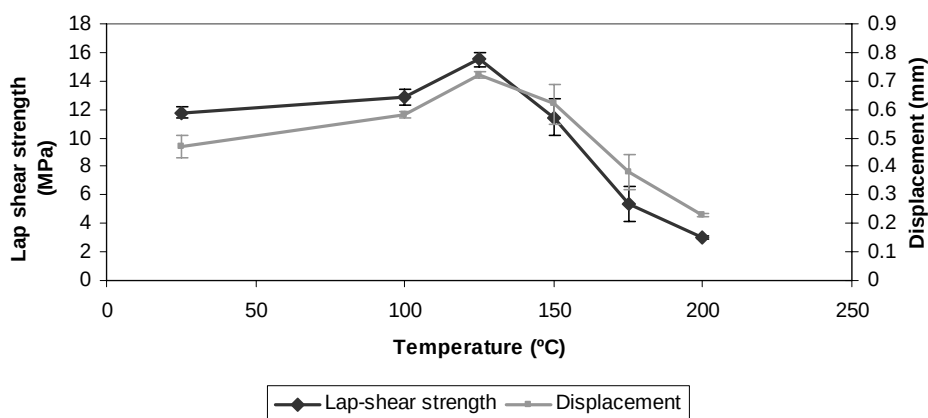


Figure 9. Average lap shear strength and displacement of XN1244 adhesive as a function of temperature.

With an increase of temperature, a slight increase of the lap shear strength occurs at 100°C. The lap shear strength of the adhesive joints tested at 100°C is approximately 9% higher than that of specimens tested at RT. Data obtained from tests at 125°C showed an increase of the lap shear strength of the adhesive by approximately 30%. This can be explained by the fact that at RT, the failure takes place at the end of the overlap and, once a crack has initiated, the adhesive has no ductility to absorb the fracture energy, while as the temperature increases and gets closer to the adhesive T_g (155°C), the adhesive becomes more ductile and the overlap contributes more to the strength as the adhesive yields. The yielding of the adhesive redistributes the stresses, making more use of the less strained parts of the overlap. In other words, joint strength depends not only on the peak values of the stresses, but also on the quantity of material that is exposed to these stresses. The decrease in stress peaks and increase in the amount of material subjected to stresses seemed to attain an optimum resulting in

maximum joint strength at around 125°C. This type of behaviour is similar to that observed by da Silva *et al.* in [26].

Generally, as the temperature is raised, the adhesive strength decreases and the ductility increases. There is a temperature when the adhesive starts to behave like a rubber (above T_g). At T_g , there is a drop in the elastic modulus and in the strength. Below T_g , it is difficult to say at which temperature the lap shear strength is the highest as there are two factors to consider: the ductility and the strength. Figure 10 shows schematically the lap shear strength behaviour as a function of the adhesive bulk properties. The lap shear strength increases with the adhesive ductility up to the best compromise between the ductility and the bulk strength. According to Figure 9 the temperature corresponding to the best combination strength/ductility for XN1244 adhesive is the 125°C.

At temperatures above T_g , the lap shear strength of the adhesive joints tested at 175°C was approximately 54% less than that of the specimens tested at RT, while data obtained from tests at 200°C showed a decrease of the lap shear strength of the adhesive by approximately 75%.

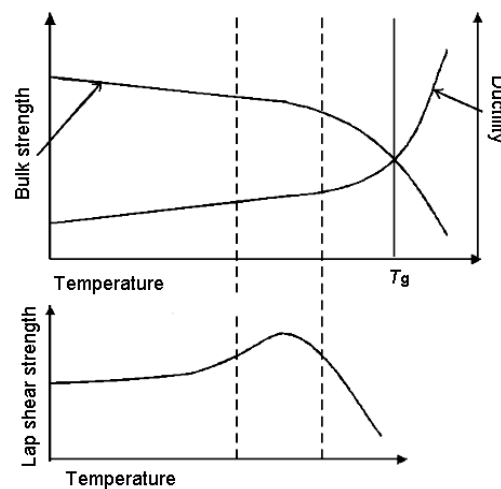


Figure 10. Lap-shear strength behaviour as a function of adhesive ductility and strength.

The variation in the test results (see standard deviation at 175°C and 200°C in Figure 9) increased at temperatures above T_g because the adhesive properties had a sharp decrease making the joint behaviour very sensitive to small temperature changes in this range.

The adhesive deformation to failure increased with increasing the temperature up to 150°C by approximately 23% at 100°C, 53% at 125 °C and 32% at 150°C respectively, while at temperatures above T_g (175°C and 200°C) the adhesive deformation to failure decreased by approximately 19% and 51%.

4. Numerical analysis

A non-linear geometrical numerical analysis was performed in the commercial FEM programme ABAQUS[®]. The mesh used for the single-lap joints can be seen in Figure 11. Restraining and loading conditions are visible in Figure 6, consisting on clamping the joint at one of its edges and applying a vertical restraint and tensile displacement at the opposite edge, to faithfully reproduce the testing conditions [27,28]. The joint was meshed by ABAQUS[®] CAE meshing algorithms from the user introduced seeding preferences (including bias effects). The mesh was particularly refined at the overlap edges to accurately capture spots of stress concentrations [29,30]. The joints were modelled as two-dimensional, with plane-strain solid elements (referenced as CPE8 from the ABAQUS[®] library).

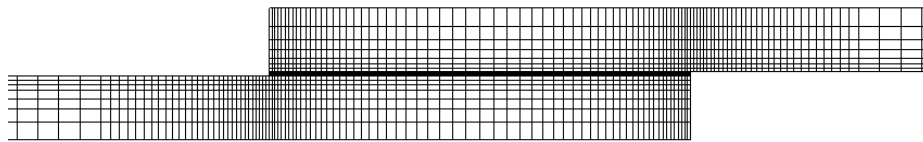


Figure 11. Finite Element mesh of a SLJ.

4.1 – Cohesive Zone Modelling

Cohesive zone models (CZM's) model the elastic loading, initiation of damage and further propagation due to local failure within a material. CZM's are based on a relationship between stresses and relative displacements connecting initially superimposed nodes of the cohesive elements (Figure 12), to simulate the elastic behaviour up to a peak load and subsequent softening, to model the gradual degradation of material properties up to complete failure. Generically speaking, the shape of the softening laws can be adjusted to conform to the behaviour of the material or interface they are simulating [31,32]. The areas under the traction-separation laws in each mode of loading (tension and shear) are equalled to the respective fracture energy. Under pure mode, damage propagation occurs at a specific integration point when the stresses are released in the respective traction-separation law.

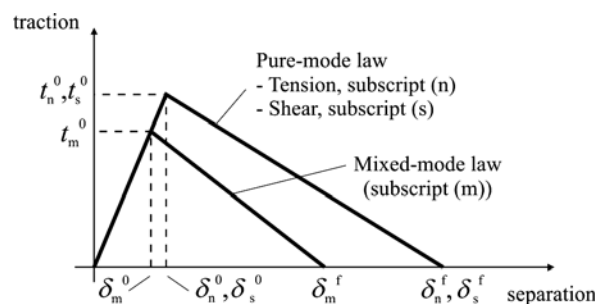


Figure 12. Traction-separation law with linear softening available in ABAQUS[®].

Under mixed mode, energetic criteria are often used to combine tension and shear [31], thus simulating the typical mixed mode behaviour inherent to bonded assemblies. In this work, a continuum-based approach was considered to model the finite thickness of the adhesive layer. The cohesive layer was assumed to be under one direct component of strain (through-thickness) and one transverse shear strain, which are computed directly from the element kinematics. The membrane strains were assumed as zero, which is appropriate for thin and compliant layers between stiff adherends. The traction-separation law assumes an initial linear elastic behaviour followed by linear evolution of damage. Elasticity is defined by an elastic constitutive matrix relating stresses and strains across the interface [33]

$$\mathbf{t} = \begin{Bmatrix} t_n \\ t_s \end{Bmatrix} = \begin{bmatrix} K_{nn} & K_{ns} \\ K_{ns} & K_{ss} \end{bmatrix} \cdot \begin{Bmatrix} \varepsilon_n \\ \varepsilon_s \end{Bmatrix} = \mathbf{K} \boldsymbol{\varepsilon}. \quad (2)$$

The matrix $\mathbf{K}$ contains the stiffness parameters of the adhesive layer, given by the quotient between the relevant elastic modulus and t_A . A suitable approximation for thin adhesive layers is provided with $K_{nn}=E$, $K_{ss}=G$, $K_{ns}=0$, where E and G are the longitudinal and transverse elastic moduli [31]. Damage initiation can be specified by different criteria. In this work, the quadratic nominal stress criterion was considered for the initiation of damage, already shown to give accurate results [31], expressed as [33]

$$\left\{ \frac{\langle t_n \rangle}{t_n^0} \right\}^2 + \left\{ \frac{t_s}{t_s^0} \right\}^2 = 1. \quad (3)$$

t_n^0 and t_s^0 represent the pure mode (normal or shear, respectively) peak values of the nominal stress. $\langle \rangle$ are the Macaulay brackets, emphasizing that a purely compressive stress state does not initiate damage. After the peak value in Figure 12 is attained, the material stiffness is degraded under different possible laws, depending on the material to be simulated. For brittle materials such as XN1244 adhesive, a linear softening law is sufficiently appropriate [34]. Complete separation is predicted by a linear power law form of the required energies for failure in the pure modes [33]

$$\frac{G_n}{G_n^c} + \frac{G_s}{G_s^c} = 1. \quad (4)$$

The quantities G_n and G_s relate to the work done by the traction and corresponding relative displacements in the normal and shear directions, whilst the relating critical fracture energies required for pure mode failure are given by G_n^c and G_s^c for normal and shear loadings, respectively.

5. Numerical results

In the simulations, by modelling the adhesive layer as a traction-separation law with CZM's and the adherends as elastic-perfectly plastic using the approximation of Figure 4, fracture occurred due to cohesive crack propagation in the adhesive bond, beginning at the overlap edges with fast propagation to the inner regions of the bond.

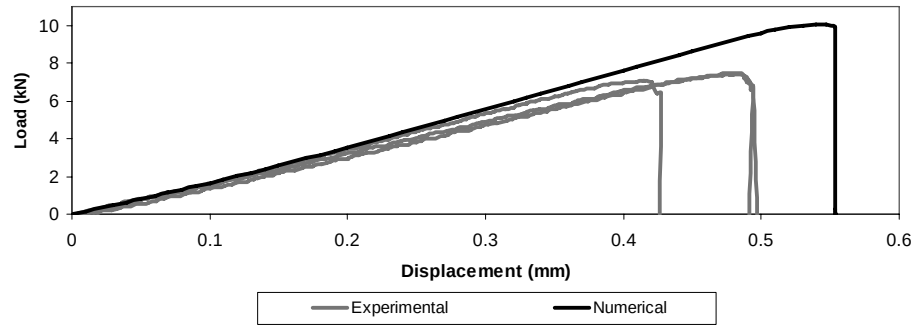
The parameters G_n^c and G_s^c (the fracture toughness in pure mode I and II) were defined by testing at each temperature, with the double cantilever beam (DCB) tests and the end notched flexure (ENF) tests, respectively in previous studies [18,19,35]. The t_n^0 (normal cohesive stress) was determined from tensile bulk tests as a function of temperature [19], while t_s^0 (shear cohesive stress) at RT, 100°C and 150°C was extracted by comparing numerical predictions from cohesive-zone simulations to the results of ENF experimental tests [35].

It is known that for ductile adhesives the average lap shear stress in the joints at failure in quasi-static loading is very similar to the measured shear strength with the TAST. Actually, in a previous work [5], the authors found for Sikaflex 552 adhesive that the TAST gave a $\tau_r = 2.39$ MPa, while from SLJs tests the results were very similar ($\tau_{av} = 2.27$ MPa). This indicated that the shear stresses in the joints with ductile adhesives are essentially uniformly distributed whether a TAST or SLJ specimens are used. In other words, the SLJ can be used to determine the shear strength of ductile adhesives, contrarily to stiff and rigid adhesives. Therefore, at 200°C, the t_s^0 was estimated from the SLJs experiments, as it was not possible to determine it with the inverse method [35]. The material properties used for the CZM simulations are listed in Table 2.

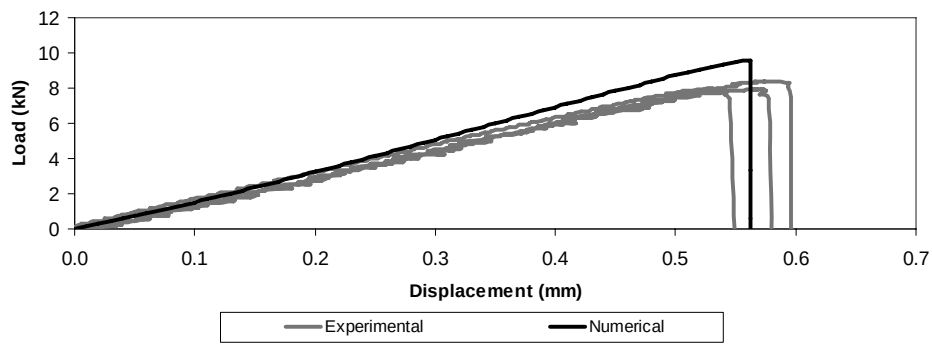
Table 2 - Properties of XN1244 adhesive for CZM modelling.

Temperature (°C)	G_n^c [N/mm]	G_s^c [N/mm]	t_n^0 [MPa]	t_s^0 [MPa]	E [MPa]	G [MPa]
RT	0.47	2.2	68	37	5872	2150
100	0.50	2.6	45	25	4173	1527
150	0.42	1.7	6.5	12	72	25
200	0.07	0.3	1.4	3	40	14.6

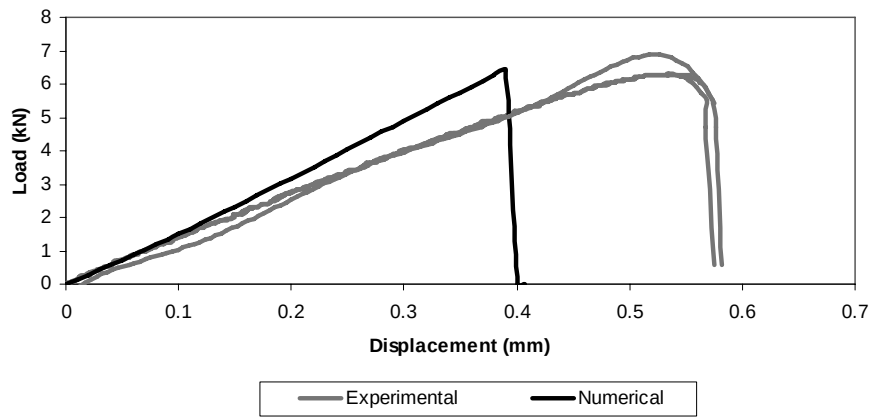
In Figure 13, representative experimental and numerical P – δ curves of the SLJ specimens at RT (a), 100°C (b), 150°C (c) and 200°C (d) can be seen.



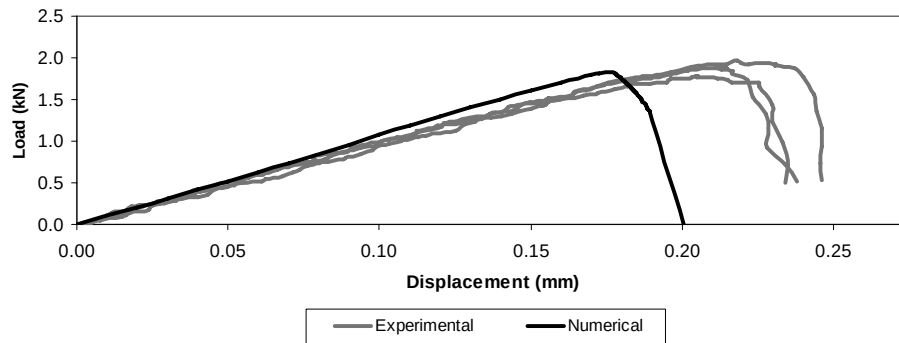
a)



b)



c)



d)

Figure 13. Representative experimental and numerical P - δ curves at a) RT, b) 100°C, c) 150°C and 200°C (d).

At RT, the failure load predicted by the CZM is around 25% higher than the experimental failure load. One factor that can explain the errors in predictions is the residual stresses. When adhesive joints are cured at high temperature and then cooled to room temperature, residual stresses will inevitably be present (the brittle adhesive XN1244 used in this study cures at high temperature (140°C)). These stresses can arise either from differential thermal shrinkage between the components of the specimen or from chemical shrinkage of the adhesive. When a crack grows in a specimen with residual stresses, the residual stresses can contribute to the total amount of energy released. On the other hand, it has been previously noted that the FE model predictions tend to be stiffer than the measured joint responses [36].

The simulation response matches the experimental results well at 100°C (approximately 11% error). At 150°C and 200°C (temperatures close and above T_g), the simulation response matches the experimental results reasonable well. However, the displacement at fracture is not well captured. The fracture in the model occurs well before the displacement where fracture occurred in the experiment. It seems that some mechanism of failure is not captured by the CZM at these temperatures.

As we have seen before, with the increase of temperature close to T_g , the ductility of the adhesive increases. Hence, even though several authors showed that the shape of the traction-separation law is relatively unimportant to the outcome of the models [37-39], the trapezoidal traction-separation law, to model the adhesive at 150°C, might be more appropriate to be used as it accounts for large plasticization up to failure occurring under these conditions. Nevertheless, while the bilinear traction-separation law is readily available in ABAQUS®, the trapezoidal traction-separation law has not yet been implemented; hence, the trapezoidal model needs special subroutines in ABAQUS® and is more difficult to use. On the other hand, at temperatures above T_g (at 200°C), some uncertainties about the state of chemical degradation of the adhesive exists. However, further investigations on the use of these laws at temperatures close and above to T_g are needed.

Conclusions

The main objective of this work was to evaluate the capabilities of using the current implementations of CZM available in ABAQUS[®] to simulate the behaviour and strength of adhesively-bonded joints as a function of temperature and validate experimentally the damage laws for pure mode I and pure mode II, determined in previous studies. With this purpose, single lap joints between aluminium adherends were fabricated and tested at RT and high temperatures. The following conclusions can be drawn:

- At temperatures below T_g , the lap-shear strength of SLJs increased (by approximately 9% at 100°C and by 30% at 125°C), while at temperatures above T_g , a drastic drop in the lap shear strength was observed (by 54% at 175°C and by approximately 75% at 200°C).
- The lap shear strength increased with the adhesive ductility up to the best compromise between the ductility and the bulk strength. The temperature corresponding to the best combination strength/ductility for the specific aluminium SLJ bonded with XN1244 adhesive was the 125°C.
- CZMs have been used to characterize the adhesive behaviour as a function of temperature. The simulation response for various temperatures matches reasonably well the experimental results.

Acknowledgements

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