

Master Thesis in Mechanical Engineering

**Fatigue resistance of bonded Arcan joints:
Interactions of loading angle, temperature, and
ageing**

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*"We can't solve problems by using the same kind
of thinking we used when we created them."*

- Albert Einstein

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To my family, all.

Abstract

Adhesive bonding technology is experiencing an exponential growth in engineering applications, replacing conventional joining methods. However, when adhesives are exposed to severe environmental conditions, they can experience a large drop of performance. Furthermore, in real life applications, most structural components can also experience fatigue load conditions and mixed mode loading conditions. Although significant research work has been carried out to assess the effect of environmental conditions over adhesive's performance, pure mode conditions were rarely considered, creating a barrier in the application of this techniques in design processes. Thus, it is now essential to understand the effect of those conditions in the fatigue behaviour. The development of an efficient procedure to predict the fatigue of those joints could be very helpful to increase the use of this technology in industrial applications.

The main aim of the present work is to understand the effect of different environmental conditions on the static and fatigue behaviour of adhesive joints, when subjected to different mode mixities and how it affects the property degradation of the adhesive; to do so, the effect of temperature and moisture were considered. Firstly, an experimental study was conducted. Strength and fracture tests were performed, using joints bonded by a two-part epoxy adhesive. All tests were done considering different mode mixities/loading angles. Arcan joints were also tested statically and under fatigue for different mode mixities.

For the samples subjected to water exposure for long periods of time, an additional experimental study was performed to understand the rate of water uptake through the adhesive layer. After that, and based on the experimental results, possible causes were searched for the different failure mechanism observed. To do this, a numerical analysis was performed to analyze the water content along the bondline. Also a fracture surface analysis of the Arcan joints was performed.

To evaluate the effect of temperature, the curing cycle was adjusted to ensure the service temperature was above glass transition temperature, T_g , by an experimental study. The same tests performed to study the effect of ageing were repeated for different temperatures. Based on the results from the fatigue tests, different master curves were obtained for different temperatures as a function of a proposed parameter. Afterwards, a relation between the curves is suggested, to create a predictive model on the effect of temperature on the fatigue life of the adhesive joints.

Resumo

O uso de juntas adesivas estruturais tem tido um crescimento exponencial nas últimas décadas, surgindo como alternativa aos métodos de ligação tradicionais. No entanto, quando expostos a condições ambientais severas, os adesivos apresentam muitas vezes uma quebra no seu desempenho. Por outro lado, em aplicações reais, os componentes ligados estão muitas vezes sujeitos a solicitações de natureza cíclica, o que complica a sua análise. Apesar de algum trabalho de investigação já ter sido desenvolvido nesta área, a forma como as condições ambientais afetam a resistência de juntas adesivas em modos puros de solicitação ainda não se encontra totalmente compreendida. Assim sendo, o desconhecimento das propriedades dos adesivos sujeitos a condições extremas é ainda uma grande barreira à implementação deste processo de ligação, uma vez que as atividades de projeto de junta requerem o conhecimento das propriedades dos adesivos pelo menos segundo as duas direções principais. De forma complementar, entender a forma como estes fatores afetam o comportamento de juntas adesivas sujeitas à fadiga é também essencial. O desenvolvimento de modelos de previsão da vida destas juntas apresenta-se por isso como muito importante para suportar o uso crescente desta tecnologia na indústria.

O principal objetivo deste trabalho é perceber de que modo o contacto do adesivo com água e a temperatura afeta as propriedades estáticas e de fadiga juntas adesivas estruturais, e em que medida o tipo de solicitação (tensão, corte) influencia o peso relativo destes fatores. Assim, um estudo experimental foi realizado: testes normalizados de tensão e fratura foram realizados para tensão/modo I e para corte/modo II, sendo que adesivo usado foi um epóxi. Juntas Arcan foram também testadas estaticamente e à fadiga segundo diferentes ângulos e diferentes condições ambientais.

Para os provetes expostos a água, um estudo paralelo foi realizado para perceber de que forma e quanto a água é absorvida pela camada adesiva. Seguidamente, e tendo em conta os resultados experimentais, possíveis causas para os diferentes mecanismos de falha verificados foram discutidas. Para tal, uma análise numérica foi realizada para avaliar a quantidade de água presente nas juntas Arcan. Também uma análise das superfícies de fratura foi feita.

Relativamente ao efeito da temperatura, o ciclo de cura do adesivo foi ajustado para garantir que os testes realizados estavam garantidamente abaixo da temperatura de transição vítrea, T_g . Os mesmos testes realizados para estudar o efeito da humidade, foram repetidos para diferentes temperaturas. Com base nos resultados dos testes de fadiga, diferentes curvas mestras foram obtidas com base num modelo proposto. Finalmente, uma relação entre as diferentes curvas é proposta.

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1 Introduction

1.1 Background and motivation

Adhesive bonding has been growing as an alternative to conventional and older joining methods such welding, riveting or bolting. [1] and today, adhesives are not only used in advanced industries such as those operating in the automotive and aerospace fields but can be found in all types of products, such as household appliances or even toys. In fact, it's hard to imagine a product that doesn't include adhesive to bond its components together.

The ubiquity of this joining process, especially in industrial sectors, is in large part supported by the extensive research work done in the past decades, which highlighted the unique advantages that this process has when compared with the other techniques mentioned before. The most significant of these include the uniform stress distribution along the bonded area, leading to higher resistance to fatigue loading conditions and better load transmission capacity. Another important advantage is its ability to join dissimilar materials from different metallic materials to composite structures. Finally, the design flexibility is also a significant advantage compared to other methods [1, 2].

Nevertheless there is still some skepticism on the use of the adhesive bonding since this is one of the most recent load bearing structural joining methods, especially when compared to classical joining methods such as welding, which have been used for multiple centuries. However, adhesive bonding has some important drawbacks as well, such the loss of performance under severe environmental conditions such humidity and high temperatures. Furthermore, the bonding process itself requires a careful preparation and a controlled and clean environment [1, 3].

This said, there is still a clear path to travel for the adhesive bonding technology in order to extract all the potential from this joining process, especially for structural adhesives in high performance applications, as in the case of the aerospace industry. For that purpose, the role of researchers is essential to ensure constant innovation and to increase the knowledge available for this technology.

1.2 Objectives

This thesis has three main objectives. The first one is to evaluate the effect of the environmental conditions on the strength and fracture properties of adhesives. The second one, is to experimentally investigate the fatigue behaviour of Arcan joints subjected to hygrothermal ageing and different temperatures under different mode mixities. Finally, the third one is to develop an efficient method for fatigue lifetime prediction, for adhesive joints subjected to different temperatures and loading angles.

1.3 Research methodology

The key objectives of this thesis are the following:

- Perform an extensive literature review, with the main focus on the state of the art on the effect of environmental conditions on adhesive's performance.
- Carry out experimental tests to determine key material properties and assess the joint behaviour under different conditions, namely bulk tensile tests, Thick Adherend Shear Tests (TAST), Double Cantilever Beam (DCB) tests, and End Notched Flexure (ENF) tests.
- Perform static and fatigue tests using an Arcan joint on different mode mixities and different environmental conditions.
- Taking into account the experimental results from Arcan under hygrothermal ageing, relate them with different failure mechanisms and find possible explanations for the results obtained.
- Propose a fatigue life prediction model for different temperatures, based on both the experimental results and on stress parameters obtained through finite element analysis (FEA).

1.4 Thesis outline

This thesis consists of a literature review, an explanation of the experimental procedures, a chapter on the numerical models and finally two papers were prepared out of this research.

This chapter has an introductory role, describing the background and motivation of this work, as well as a brief description of the problem in hand and its objectives.

Chapter 2 starts with an overview on the state of the art in adhesive joints, their different classifications and some important design considerations. Afterwards, some industrial applications of adhesive bonding are briefly reviewed, focusing on the effect of moisture and temperature on adhesives joints. Finally, the fatigue life prediction methods are discussed and reviewed , particularly using S-N approach.

In Chapter 3, the experimental procedures used throughout this work to manufacture and test the adhesive joints under study are explained. It's also mentioned how the data was processed in order to obtain the properties and curves of the material by the raw data from the testing machines. Finally, some considerations are done regarding the design of a thermal chamber that was custom developed during this thesis to perform high temperature tests.

In Chapter 4 the details of the numerical models developed for the finite elements analysis (FEA) are explained.

Paper 1 - F.V.B. de Castro Lopes, A. Akhvan-Safar, R.J.C. Carbas, E.A.S. Marques, R. Goyal, J. Jennings, L.F.M. da Silva. **The interaction of loading mode and humidity on the properties degradation of an epoxy adhesive subjected to strength, fracture, and fatigue tests**

Abstract of Paper 1: Hygrothermal ageing has a significant impact on the performance of bonded joints. Despite extensive investigations into the ageing effect of bonded structures, there is no comprehensive study that examines the rate of the ageing effect as a function of the loading type and the load angle.

Accordingly, in this study, an epoxy adhesive was tested under different loading types/angles including tensile and shear standard strength tests, mode I and mode II fracture tests, and fatigue and static Arcan tests. All these experiments were performed for both aged and unaged conditions. To understand the influence of loading angle on the failure response of aged bonded joints, Arcan samples were subjected to mixed mode loading conditions as well. A fractography analysis was conducted to provide better understanding of the failure mechanism of aged samples loaded at different modes. Based on these results, it was found that mode I (tensile loading conditions) is more sensitive to the aging conditions than mode II (shearing) for all loading types (strength, fracture, and fatigue tests). The influence of moisture on fracture toughness was however less relevant than on strength. The results also showed that for the same loading mode and for the same Arcan joints, the fatigue strength is more degraded by aging conditions than the static strength.

Paper 2 - F.V.B. de Castro Lopes, A. Akhvan-Safar, R.J.C. Carbas, E.A.S. Marques, R. Goyal, J. Jennings, L.F.M. da Silva. **The interaction of loading mode and temperature on the property degradation of an epoxy adhesive subjected to strength, fracture, and fatigue tests**

Abstract of Paper 2: The type of loading and high temperatures have a significant impact on the mechanical behavior of adhesive joints. In real applications, joints are often under mixed stress states and usually the service temperature is different from room temperature. For these reasons, understanding how the mechanical properties of adhesives are affected by these factors is crucial in joint design. Although multiple studies have approached the effects of temperature on the behaviour of bonded joints, the role of loading mode and test type on the rate of degradation of adhesive/joint properties at high temperature has not been properly investigated.

Accordingly, the aim of the current study is to determine if adhesives experience the same rate of degradation at high temperature when they are subjected to different mode mixities and different types of tests. To do so, an epoxy adhesive was subjected to different loading conditions (strength tests, fracture tests, and fatigue tests, all for modes I and II) at different temperatures. The results show that there is a significant difference between the rate of degradation in mode I and mode II

loading conditions and also that, for the same loading mode, the rate of degradation is mostly a function of the test type. For the fatigue experiments, the S-N response of the tested adhesive was obtained as a function of loading mode using Arcan joint testing. Based on the results, an S-N model was also developed to create a master S-N curve as a function of temperature.

2 Literature review

2.1 Adhesive joints

Throughout the years, many authors have tried to find an accurate definition for adhesives. Kinloch [4] defined an adhesive as a material that has the ability to resist loads and resist separation when applied to surfaces of materials. Before the joining process itself, the materials which are going to be bonded are called substrates, and afterwards they are referred as adherends. The interphase is defined as the physical region between the adhesive and the adherend with different properties from the materials to bond, while the interface can be defined as the geometrical plane of contact between the surface and the two materials [1]. Figure 1 shows a schematic representation of an adhesive joint. The attraction forces between the adherend and the adhesive are a consequence of intermolecular interactions existing between them, and can be designated as an adhesion phenomenon, which is different from the cohesion phenomenon where the intermolecular forces are only between one substance itself [1]. The differences between these two phenomena can be observed in Figure 2.

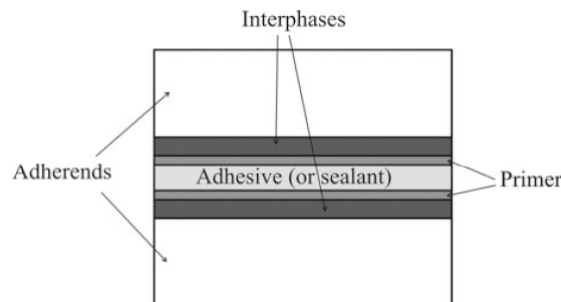


Figure 1: Adhesive Joint [1].

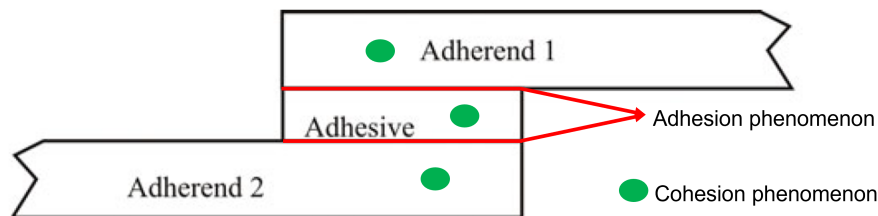


Figure 2: Adhesion vs cohesion phenomena - adapted from [1].

In the last 80 years, adhesives based on synthetic polymers, such as polyurethanes and epoxy resins, have been developed and explored as alternatives to typical joining methods. Structural adhesives were created for mechanical and aerospace engineering applications, providing strength and stiffness to the structures they join. [1, 5].

When compared to conventional joining processes, adhesive bonding brings many important advantages [1, 3]:

- Uniform stress distributions - Figure 3;
- Excellent fatigue strength;
- Damps vibration and absorbs shock;
- Prevents galvanic corrosion due by avoiding the direct contact between the 2 substrates.
- Work well with a variety of combination of similar or dissimilar materials;
- Large design flexibility, joining all shapes and thicknesses ;
- Allows smooth surface finishes;
- Heat, if required in the process, is to low to affect the metal parts involved.
- Attractive strength-weight ratio, specially useful for transportation industries.

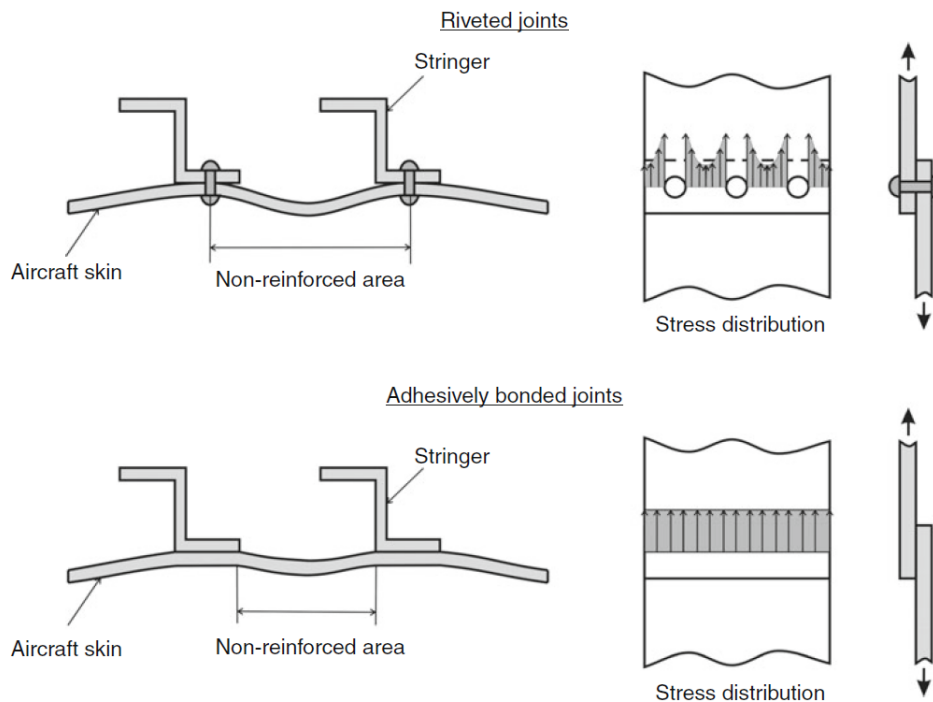


Figure 3: Stiffness and stress distribution - adhesive bonded Vs. riveted joints [1].

However, this method is not perfect, and that's why different joining methods coexist. Thus, adhesive bonding also presents some disadvantages [1, 3]:

- The need for careful surface preparation;

- Sometimes, long cure times are required (can go from few minutes up to several weeks);
- They are limited in the operating temperature, due to the existence of a glass transition temperature which is under 180 °C in the majority of the adhesives.
- Limited resistance to extreme humidity conditions, due to their polymeric nature;
- Jigs and fixtures can be needed;
- Hard to inspection the joint quality, without invasive methods.
- Poor performance under peeling and cleavage stresses;
- Difficulties in performing quality control and also requires special training sometimes.

2.1.1 Adhesive classification

Adhesives can be classified according to different parameters as we can see in Table 1.

When we refer to the polymeric base of the adhesive, natural adhesives include vegetable or animal based adhesives as well as natural gums such as natural rubber or bee wax. Such types of adhesive have been used for thousand of years. As for synthetic base adhesives, we are coming to the recent days; those include epoxies, polyurethanes, silicones, among many others [1, 6].

Adhesives can also be classified by the functionality of the polymer “backbone”. Thermoset adhesives are polymers whose cure process is irreversible due to chemical reactions at room or high temperature that make it permanent. They can be supplied as one-part, with dormant reactive agents, or multiple-part systems. The one-part adhesives usually need high cure temperatures and since their shelf life is reduced it is common to store them at low temperatures. Multiple-part systems often cure at room temperature and have a longer shelf life. Thermoplastic adhesives are usually cured from a molten state and can be remelted several times without significant change in their properties. As for elastomers, they are typically capable of high elongation, good toughness and peel strength. Hybrid adhesives result from a combination of the previous ones [1, 7].

Physical form is divided into liquid, paste, film or powder, and generally can be a sign of how it must be applied. Liquid adhesives are applied using rolls, sprays or brushes and have good wettability, filling any existent gap with ease. Paste adhesives can be comfortably applied on vertical surfaces, due to their high viscosity. Film adhesives are perfect to restrict bondline thickness as constant and are easy to

handle. Powder adhesives typically have to be heated or dissolved into a solvent to be transformed into a liquid form, prior to application [1, 7].

According to the chemical family, they can be characterised as epoxies, polyurethanes, acrylics and many more. Epoxies are the most important group of the structural adhesives, and generally are strong and brittle, and can work under a big range of temperatures. They cure usually at room temperature, being suitable to bond a significant variety of substrates. Polyurethanes, which are part of the elastomers group, are usually tough and flexible, being able, in some cases, to reach over 200 % elongation. Those are highly sensitive to severe environmental conditions, especially high temperatures. [1].

Adhesives are usually designed to have different properties to fit different functions. They can be mainly separated as structural or non-structural adhesives. Structural adhesives are characterized by having high strength and stiffness; they are used to transfer load in their service conditions and usually are composed by a cross-link of thermosetting polymers. [1].

Table 1: Adhesive Classification [1]

Criteria	
Polymer base	- Natural - Synthetic
Functionality in the polymer “backbone,”	- Thermoplastic - Thermoset - Elastomeric - Hybrid
Physical form	- One-component - Multi-components - Films - ...
Chemical families	- Epoxy - Polyurethane - Silicon - ...
Function	- Strucutural - Hot-melt - Pressure-sensitive - Water-based - ...

2.1.2 Failure modes

When evaluating the failure of an adhesive joint, where the substrates are made from a metallic material, we can have 3 possible failure modes: adhesive failure, cohesive failure, or a mixed failure of the two mentioned before. Figure 4 shows in a schematic way those 3 possible failure modes [1].

Cohesive failure is usually the most desirable type of failure because is the easiest to predict and, consequently, to control. It is defined as the tear of the material itself. When cohesive failure of the adhesive occurs it's easy to identify because both adherends stay covered with adhesive as it's observed in Figure 4. On industrial applications, when this type of failure occurs, the typical causes are inadequate overlap length, or even thermal stresses not taken into account when the joint was designed. However, this type of failure can also occur on the adherend, especially when bonding non-metallic materials [1, 6]

Adhesive failure (see Figure 4) occurs when the joint fails at the interface region. The strength of this region depends on multiple factors such as the substrate materials, its affinity with the adhesive, and also the production conditions such as surface cleaning and preparation. As a result, some causes for this type of failure include poor surface preparation involving contaminants. Another common cause for this type of failure is the continuous contact of the adhesive joint with water; in fact, moisture is one of the biggest weakness of adhesive bonding. [1, 6]

However, fully adhesive failure is not really common in real life applications, and usually companies deal with a mix of both adhesive and cohesive failure as we can see in the bottom part of Figure 4.

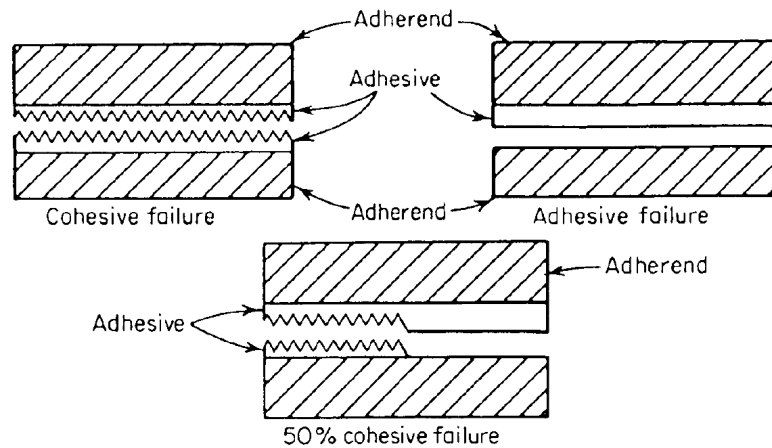


Figure 4: Different failure modes of an adhesive joint [3]

2.1.3 Joint design

Adhesive joints must be carefully designed taking into account the loads that the joint will need to withstand. The type of stress felt by the adhesive joint depends

on the way the load is applied. In Figure 5 are shown the most common types of stresses acting on adhesive joints.

However, when designing adhesive joints, other variables should be taken into account as environmental conditions or the prediction of possible defects. In fact, each real life application is different and that's why there are many different geometries (see Figure 6). The single lap joint is by far the most common and also the target of most studies. da Silva et al. [8] studied the effect of many variables as different materials and environments on the shear performance of single-lap joints. Grant et al. [9] tested single-lap joints by many load conditions to investigate their failure process; for that they used an epoxy structural adhesive and mild steel adherends. [1]

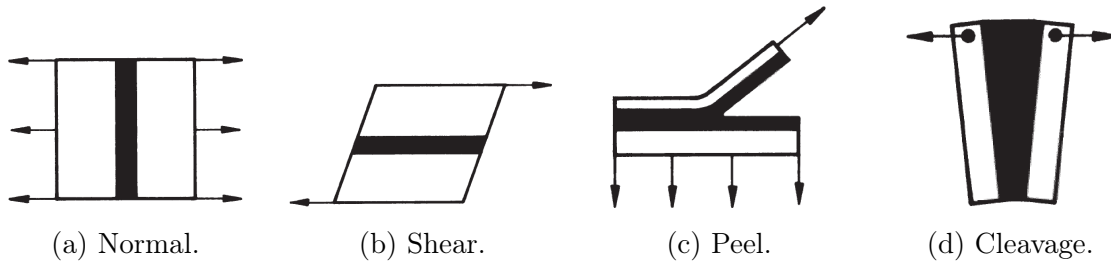


Figure 5: Types of stresses on adhesive joints. Adapted from [4].

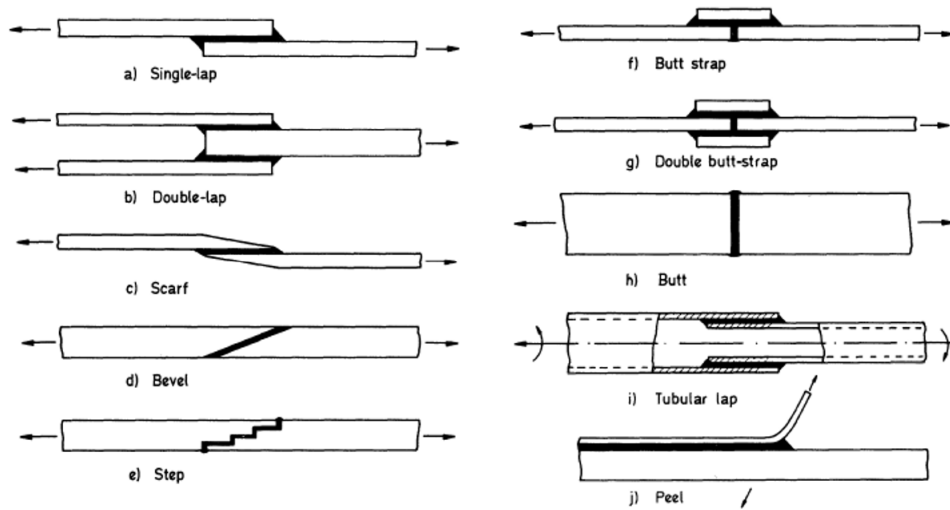


Figure 6: Different joint designs - adapted from [5]

Finally, another important variable in the joint design process is the adhesive layer thickness: it's not only important to determine the optimum value for the adhesive thickness, but it is also important to ensure its uniformity along the bonded area. In fact, a non-uniform adhesive layer can conduct to considerable stress concentrations which can lead to premature failure. Some methods to control this phenomenon include [3]:

- Play with the adhesive’s viscosity;
- Application of pressure during cure;
- Using fixturing designed for the application.

2.2 Fatigue life in adhesives joints

Fatigue, in engineering applications, relates to the behaviour of structure under cycling loading conditions. We can find some type of fatigue loading in most applications, from airplanes to buildings or bridges. This phenomenon can be very dangerous because it can lead to a sudden failure of the structure after decades working. To characterize a fatigue load, some parameters are needed. A fatigue cycle is defined with a sinusoidal waveform. For example, with the frequency, amplitude, and average stress, it becomes possible to characterize a simple fatigue load as Figure 7 illustrates. [1]

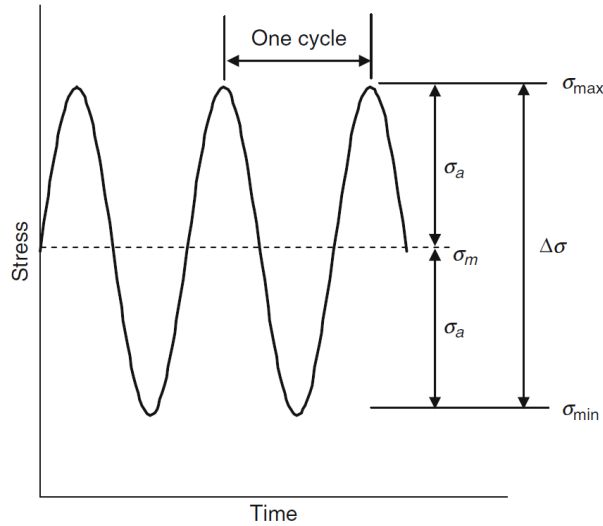


Figure 7: Fatigue load following sinusoidal waveform [1].

The multiple advantages of adhesive bonding have already been mentioned before. One of the most notable ones is its higher fatigue resistance when comparing with traditional techniques. Fatigue is, incontestably, a very important type of loading for many structural components. When subjected to fatigue conditions, a joint can fail at a small percentage of its static strength and thus fatigue predictions methods are required for components designing. [10]

The fatigue life of component, is usually divided in two phases, crack initiation and crack propagation:

- Crack initiation: in adhesives, this phase is very complex and yet not fully understood. For this reason, many authors have ignored this phase and focus

their analysis only on crack propagation. This is related with difficulties on modelling the initiation of a crack and to detect this phase. Some techniques to do this include video microscopy and back-face strain measurements. In real life applications the crack initiation is registered visually, by the detection of flaws or by observation of crack onset. [1, 10]

- **Crack propagation:** Fatigue crack propagation is one of the most studied topics related to fatigue life in adhesive joints. Usually, to do it, fracture mechanics tests are performed with the objective of identifying a relationship between a fracture parameter and the crack growth rate. Figure 8 shows a typical fatigue curve for adhesive bonding, where the crack growth rate (da/dN) is plotted as a function of the maximum strain energy release rate ($G_{\max}$) over time.

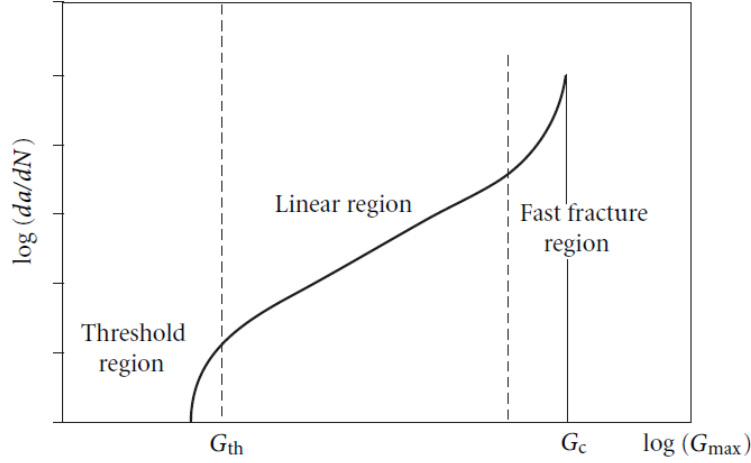


Figure 8: Fatigue crack propagation [10]

2.2.1 Fatigue prediction methods

Predicting the failure of a structure, requires the ability to determine its life as a function of the loading conditions. By this reason, predicting the fatigue behaviour of an adhesive joint can bring many advantages; first of all, safety is increased because if a structure fails unexpectedly, it can result in deaths. Furthermore, by having an estimation on the fatigue life of a component, the ones who design the joints can make them more efficient and more cheap, which is very important in a capital world where the humans live. The different methods to predict the fatigue life of an adhesive joint can be subdivided in four groups: total-life methods, phenomenological methods, fracture mechanics methods. The study will more deeply analyse the first of these approaches, since this is the one that will be used in this project.

1. Total-Life Methods

With this approach, a plot of the number of cycles until failure (N) as a

function of a load-relatable variable, as the max load itself, or the stress is made. Usually, and when the deformation suffered by the material is mainly plastic, the stress is the most common variable to use. As a result, the curve is named "*S-N*" curve or Wohler plot. Since the number of cycles usually varies a lot with a small variation on the load, a logarithmic plot of the number of cycles is recommended. For the load-related variable, it depends from case to case. Some caution should be exercised when using this method: first of all, S-N curves do not usually provide any info on the progression of the damage on the joint. Furthermore, the fitted curve depends on many factors, such as material, geometry or environmental conditions. Although this is a very useful methodology, S-N curves are limited due to these particularities.

2. **Phenomenological Methods** In these approaches, the fatigue damage is characterized as a function of a measurable parameter.
3. **Fracture Mechanics Methods** In these methods, the focus stays on the crack propagation region.
4. **Damage Mechanics Methods** These methods try to solve a problem by employing fracture mechanics methods, which is not taking into account the first phase of the failure, the crack initiation. This problem try to be solved by allowing progressive failure to be modeled. This requires a new variable, *D*, to evaluate the severity of the damage.

2.3 A review on the effect of environmental conditions on adhesive bonding

The sensitivity of adhesives to extreme environmental conditions is one of their main disadvantages. In fact, it is known that humidity and temperature are the two main sources of property degradation in adhesives. As a result, several authors have done a review on the effect of external conditions on adhesives' performance [11]. Furthermore, and since fatigue loading conditions are the also really common in real life applications, authors also have done specific reviews over the effect of the environmental conditions on the fatigue behaviour of the adhesives [12, 13].

2.3.1 Humidity

The effect of the humidity level on adhesive joints is currently one of the most studied subjects on the field. To analyse this effect, it's essential to fully understand the hygrothermal mechanism to replicate it in laboratory conditions.

Authors have shown that this degradation due to the ageing mechanism cannot be avoided when joins operate under these conditions for long periods. Wei et al. [14]

carried out multiple studies on the hygrothermal effect in many shaped adhesive joints on aluminium substrates. Two main conclusions were drawn: the failure mode changed with the passing of ageing time and that the joint degradation can be accelerated by exposing the joints to water at higher temperatures. Figure 9 shows the effect of ageing on the failure load and displacement for different geometries.

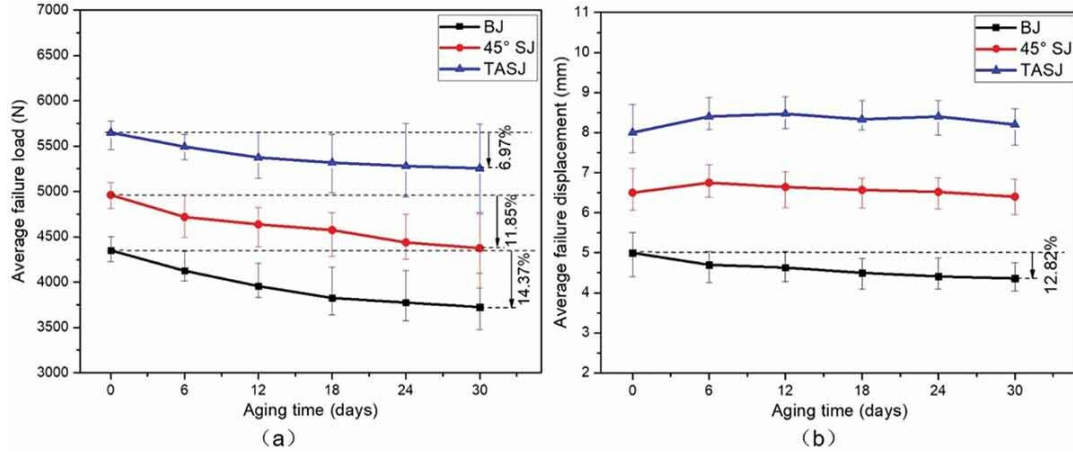


Figure 9: (a) Average failure load and (b) average failure displacement of the BJs, 45°SJs, and TASJs after high-temperature ageing [14].

Zhang et al. [15] studied the degradation of the shear properties degradation under hygrothermal conditions. Hirulkar et al. [16] studied the effect of hygrothermal ageing on the failure load of single lap joints for different adhesives. The authors concluded that, independently from the adhesive studied, the exposure time and the temperature are the most important parameters (see Figure 10). Loh et al. [17] concluded that the interfacial fracture energy decreases by increasing the moisture levels. This was achieved by measuring the interfacial strength on a epoxy-steel interface. Furthermore, Borges et al. [18] concluded that the water absorption can occur not only through the interface adhesive-substrate, but also by the cracks and voids of the adhesive.

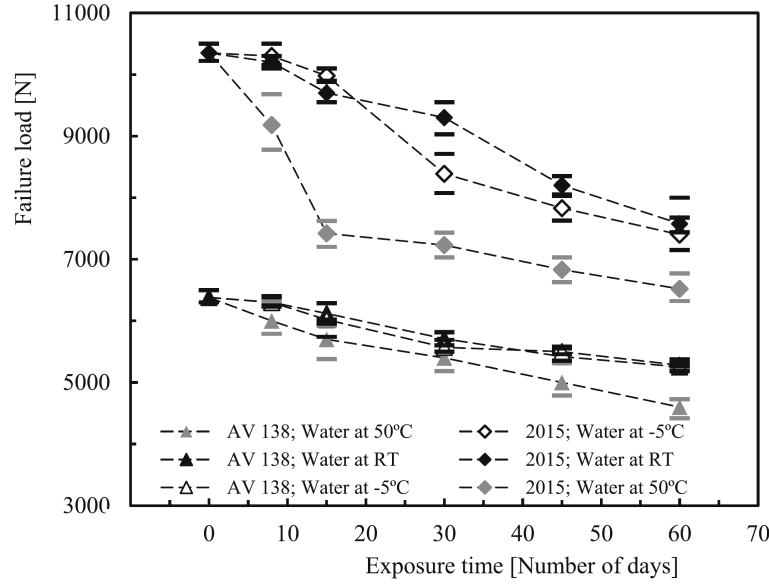


Figure 10: Hygrothermal effect on the ultimate extension [16].

Some authors have also studied the effects of cyclic ageing on the strength and fracture performance of adhesive joints. Moazzami et al. [19] subjected an epoxy-based adhesive to cyclic ageing conditions. The results showed a decrease on both elastic modulus and the tensile strength with the aging cycles. Results, presented in Figure 11, also showed the first absorption cycle is the most important. Also da Costa et al. [20] studied the impact of single aging and cyclic ageing on the properties of an epoxy-based adhesive. They concluded that the first ageing cycle is the one with the largest impact on the decrease of both the Young's modulus and tensile strength, with the subsequent cycles having a lower impact on the adhesive degradation.

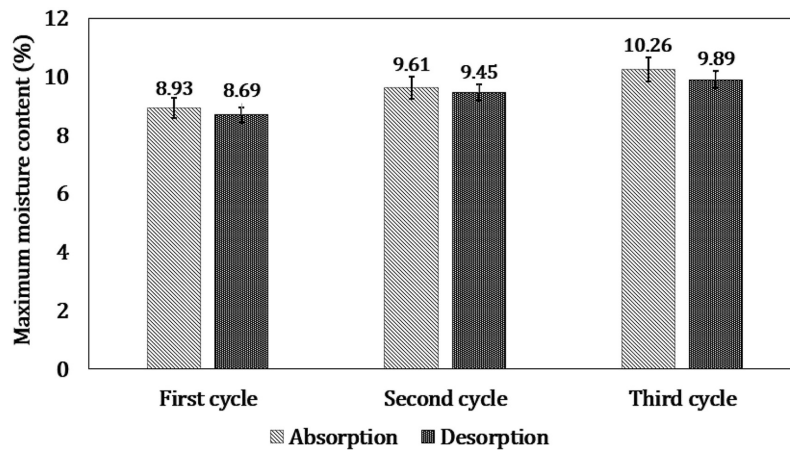


Figure 11: Variation of maximum moisture content in the absorption and desorption processes for different cycles [19]

When fatigue is combined with those conditions, this usually results in a premature failure of the joint. The effect of water on the fatigue behaviour on aluminium

substrates bonded with two different adhesives was studied by Costa et al. [21] by using DCB substrates. Their results showed the fatigue life decreases with the increase of water level at the joint. Machado et al. [22] studied the hygrothermal ageing over fatigue behaviour, using hybrid joints with aluminium and carbon fiber reinforced polymer (CFRP). They concluded the water lead to a loss in the interfacial strength affecting the fatigue performance, being this more evident for higher overlap lengths (see Figure 12).

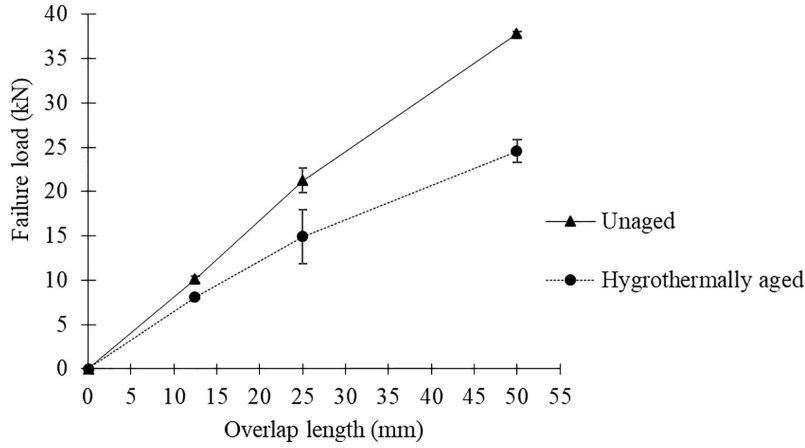


Figure 12: Comparison between the failure load of unaged and hygrothermally aged SLJs as a function of the overlap length [22].

As those studies show, the effect of the ageing over the adhesive joint performance is vastly study. However, the effect of interfacial ageing on different pure mode conditions has been rarely considered. Some investigation was conducted by da Costa et al. [23] on the effect of ageing on the S-N response of the adhesives on pure mode I using Arcan joints.

2.3.2 Temperature

In real life applications, adhesives work under severe service conditions, which often include high temperatures. Temperature itself, is one of the main factors affecting an adhesive's performance; for different temperature ranges we observe large differences on adhesive's strength, elasticity, and stiffness. A big reason behind sudden changes in the behavior of the adhesive is the glass transition temperature, T_g . For temperatures above T_g , the adhesive will experience more elongation, with lower strength and stiffness. Adams et al. [24] analyzed the effects of thermal stresses by testing single lap joints at different temperatures. Banea et al. [25] tested dumb-bell adhesive specimens at room temperature, 80 and -40, and concluded for higher temperatures the adhesive increased his toughness, while for lower temperatures is behavior tend to be more brittle. Some studies show that the T_g of an adhesive can vary with the curing cycle he is subjected; Zhang et al. [26] found out a significant relationship between the T_g of epoxy adhesives and the curing temperature, with

higher curing temperatures leading to higher T_g also (see Figure 13). They also concluded that a post-cure after a low temperature cure also elevates the T_g of the adhesive. They finally also concluded that for very high curing temperatures, T_g decreases, as Figure 14 shows; the authors believed this is due to thermal degradation of the adhesive itself. Carbas et al. [27] also studied this phenomenon reaching the same conclusions.

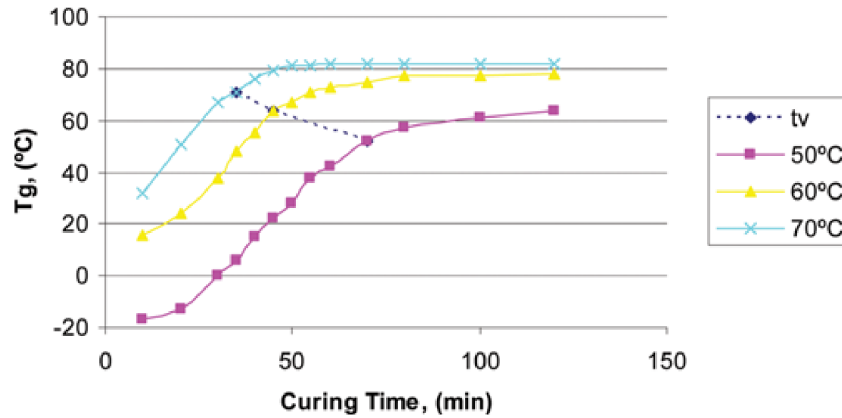


Figure 13: T_g vs. curing time for 2011 at different temperatures, tv is the time for vitrification [26].

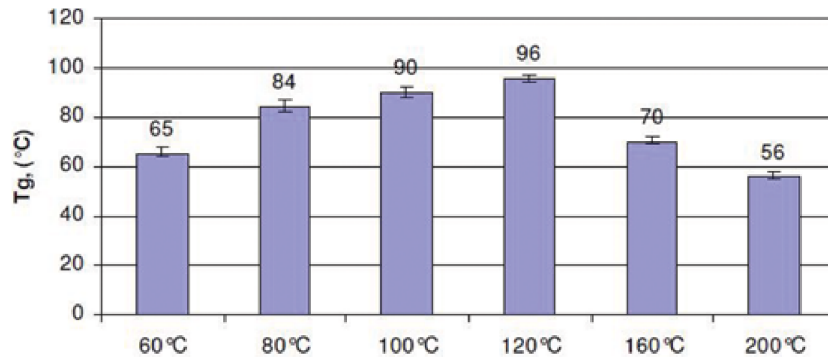


Figure 14: T_g of post-cured at different temperatures for 10 min after an initial cure of 12 h at room temperature [26].

Subsequent studies have corroborated the property degradation due to the exposure to different temperatures. Na et al. [28] subjected dissimilar adhesive joints to different temperatures concluding the increase of temperature also brings a decrease into the joint's strength, but a significant increase in its strain, as Figure 15 shows. Deb et al. [29] used double lap joints with a rubber-toughened epoxy-based adhesive to study the effect of different extension rates and temperatures; they concluded that for higher temperatures the adhesive becomes softer and his strength decreases; on the other hand for temperatures under room conditions, they found

out an increase on the strength of the joint without a similar value in terms of elongation. However, solutions have been search from some authors to try to solve this disadvantage of adhesive joints. The potential of using dual adhesives to increase temperature working range of adhesives was successfully demonstrated by da Silva and Adams [30].

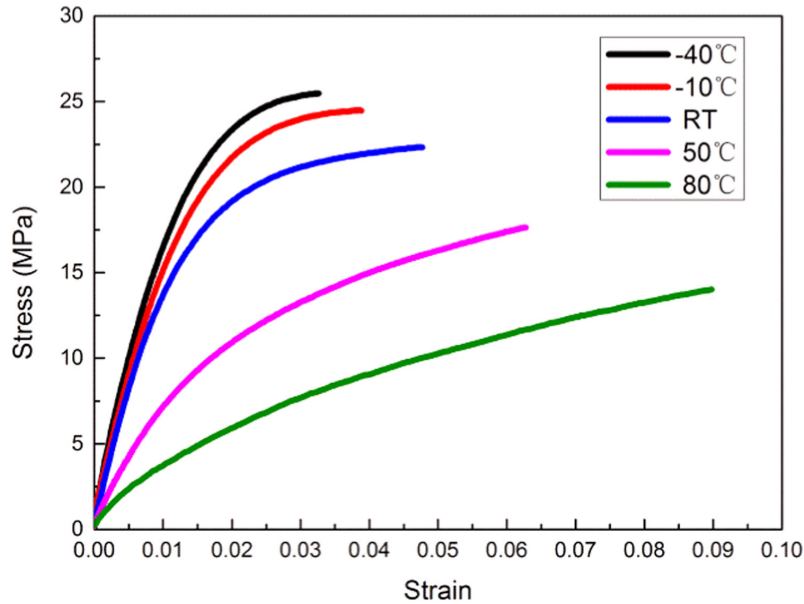


Figure 15: Representative tensile stress–strain curves at different temperatures [28].

When fatigue loads are combined with different temperatures, this can lead to a premature and unexpected failure of the adhesive joint. There is still limited research published on the influence of different temperatures on the fatigue behavior and performance of adhesives. Schneider et al. [31] studied the fatigue damage behavior of an epoxy adhesive using scarf joints under 3 different temperatures: 10, 23 and 50 . They experienced a reduction of fatigue strength with increasing temperature for constant load conditions, as Figure 16 shows. Wei Tan et al. [32] studied the fatigue behavior of a polyurethane adhesive under different service temperatures, using TASJ joints. He subjected them to five different temperatures from -40 to 80 . They concluded the fatigue performance of the adhesive gradually decreases with the increase of temperature. They also concluded the failure mechanism may change with the increase of the temperature. Beber et al [33] studied the influence of temperature on the fatigue behavior of an epoxy, suing many joint geometries. From the experimental results, they concluded with the increasing temperature, the fatigue lifetime decreased.

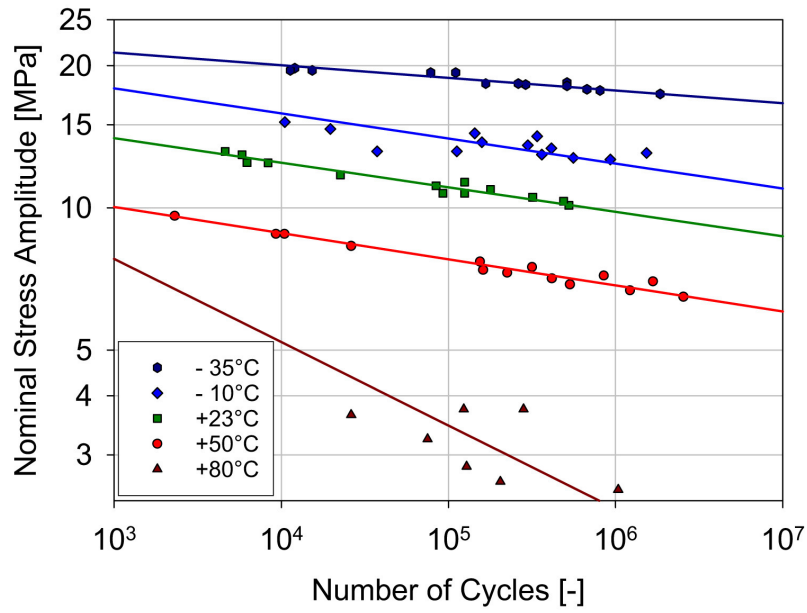


Figure 16: S-N curves under constant amplitude fatigue [31].

Studies show, the effect of temperature over the glass transition temperature, T_g , and on adhesive's property is vastly study. However, rarely pure mode conditions has been considered. In fact, no studies were found on the effect of temperature on different pure mode load conditions.

3 Experimental procedures

The knowledge of the properties of a material is very important for any design process. On a joint, targeting a real-life application, this is even more essential since there is a combination of different materials, loading modes and service conditions which complicates the design process.

3.1 Materials

The adhesive used within these experiments is a toughened and high impact-resistant two-component epoxy structural adhesive. This adhesive is used for manufacture of vehicles and other general structural applications. It can also be combined with spot-welding, clinching or riveting for repair operations. The most relevant properties of the adhesive, measured under room conditions, are presented in Table 2.

Table 2: Most relevant properties of the adhesive used for the study.

Property	Adhesive	
	Part A	Part B
Chemical Base	Epoxy	Amine
Density/g·cm ⁻³	1.08	1.06
Viscosity/Pa·s	430	100
Tensile strength/MPa	33	
Shear strength/MPa	25	
Elongation at break/%	6	
Young's modulus/MPa	1700	
Glass Transition Temperature/°C	57.5	

Since some of the tests carried out in this work were performed after ageing processes, involving contact of water with the substrates, stainless steel substrates were used to avoid the formation of rust. To guarantee the consistency of the results, stainless steel substrates were always used, even for non-aged joints. This said, two different steels were used throughout all the experiments: AISI 304 and AISI 420. The main mechanical properties of each steel are presented in the Table 3.

Table 3: Properties of the substrates used for the adhesive characterisation.

Property	AISI 304	AISI 420
Phase	Austenitic	Martensitic
Carbon/%	< 0.07	0.16 – 0.25
Chromium/%	18 – 20	12 – 14
Nickel/%	8 – 10.5	no reference
Young's modulus/ MPa	210000	
Poisson's ratio	0.28	

3.2 Geometry and manufacture procedure

In this section, the manufacturing process of the joints is described in detail. In order to define the curing cycle a preliminary study was performed. It is important to mention that all stainless steel substrates were sandblasted and cleaned with acetone in order to ensure a good level of adhesion.

T_g specimens

The effect of the curing cycle on the T_g proved to be significant. As a result, and in order to define the curing cycle to use to all other tests, small bulk specimens were produced.

Diffusion tests

In order to obtain the diffusion coefficient and determine the saturation point of the adhesive, diffusion tests were performed. For that purpose, 60 x 60 x 1 [mm] adhesive plates were made. They were placed in silica for 2 days to remove all the free water and their mass was measured. Finally the specimens were placed in a closed container filled with distilled water at 50 °C, and the mass of the plates was measured regularly to evaluate the evolution of the amount of water being absorbed by the adhesive.

"Dogbone" bulk tensile test

To evaluate the tensile properties of the adhesive and the influence of that the ageing mechanism and the temperature exerts over these properties, "dogbone" bulk tensile tests were performed, according to the British standard BS2782 [34]. To manufacture the specimens, a 2 mm adhesive plate was produced using a specific

mould (see Figure 17). Afterwards, the plate was cut in the desired shape for testing using a CNC machine, the dimensions of which are shown in Figure 18.

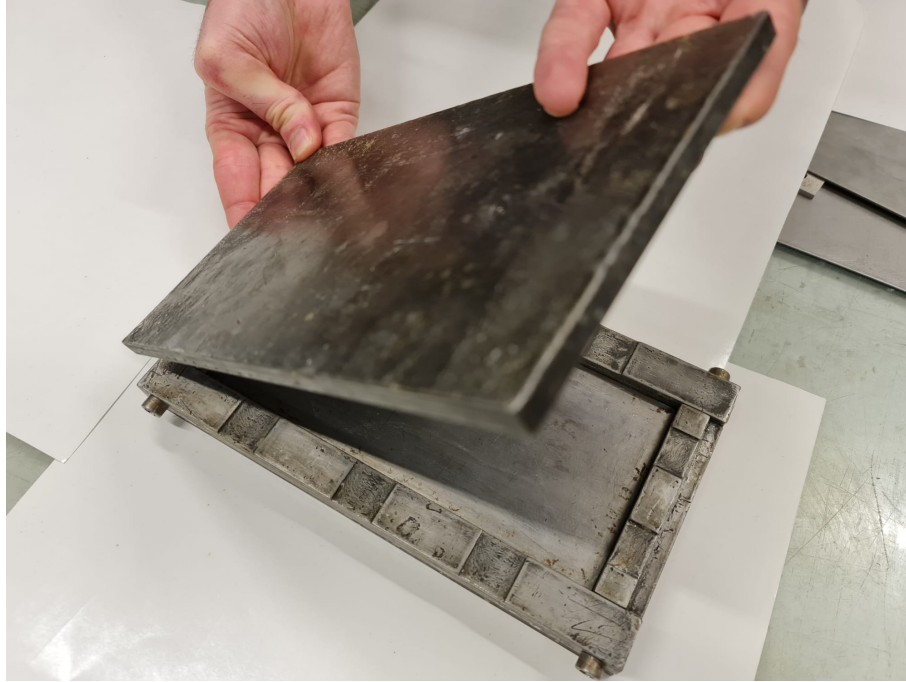


Figure 17: Mould used to produce adhesive bulk plates.

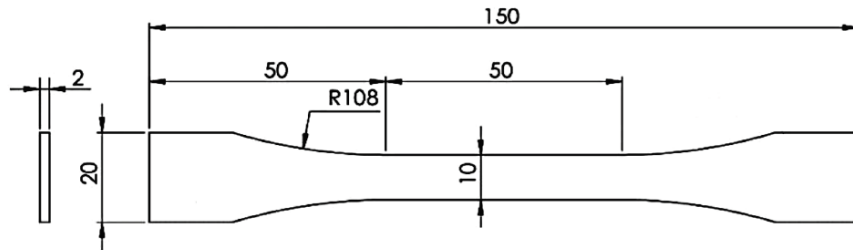


Figure 18: Bulk tensile specimen's geometry, dimensions in millimetres.

Thick adherend shear test (TAST)

TAST specimens were produced to evaluate the behaviour under pure shear loading conditions, following the ISO 11003 2:2019 [35] standard. To produce the joints and to ensure consistent substrate alignment and adhesive thickness values, a custom designed mould was used, as it can be observed in Figure 19. The dimensions of the specimen can be seen in Figure 20. To measure the strain, a non-contact method was employed, using a digital image correlation (DIC) technique. A speckled pattern was painted in the specimen, as shown in Figure 21, to support this process.

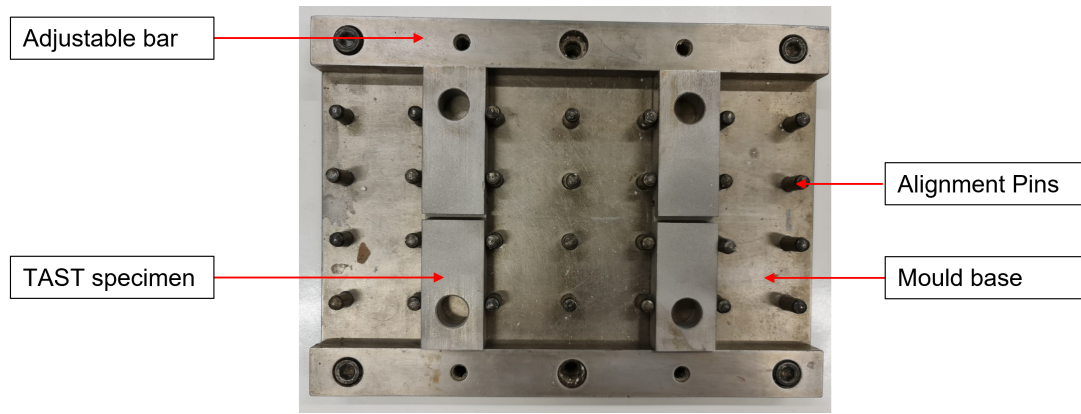


Figure 19: Mould used to produce TAST joints.

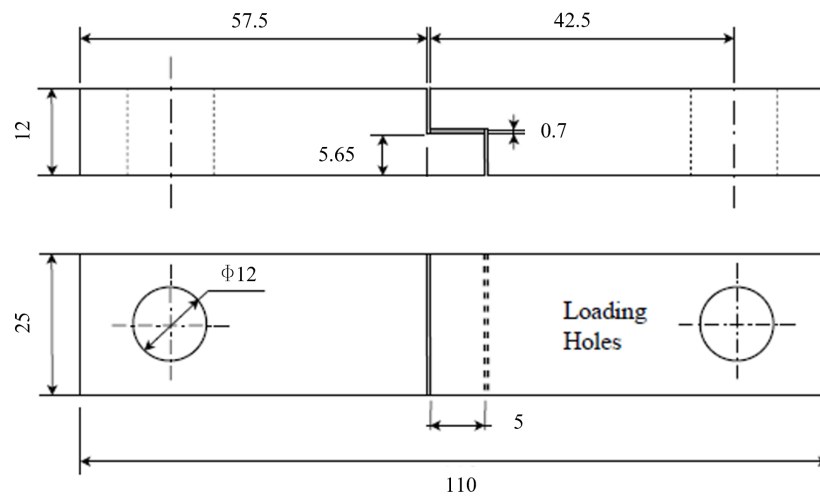


Figure 20: TAST joint drawing, dimensions in mm.

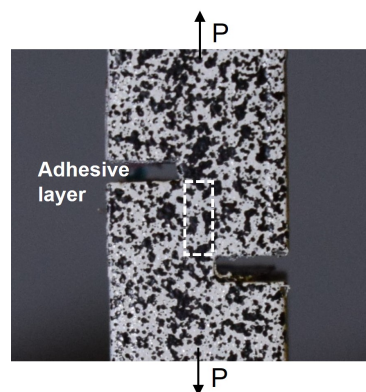


Figure 21: Speckle used in TAST joints.

Double cantilever beam (DCB) and end notched flexure (ENF) tests

A complete understanding of the fracture behaviour of an adhesive joint is of crucial importance for joint design. Accordingly, specimens were produced to deter-

mine fracture toughness of the adhesive. The same specimen is used for the DCB and ENF procedures (see Figure 22). To produce the specimens, a specific mould was used and the adhesive thickness was controlled via the use of a spacer in both sides of the specimen. The crack length, a_0 , was set to 45 [mm], generated using a blade embedded in the adhesive layer.

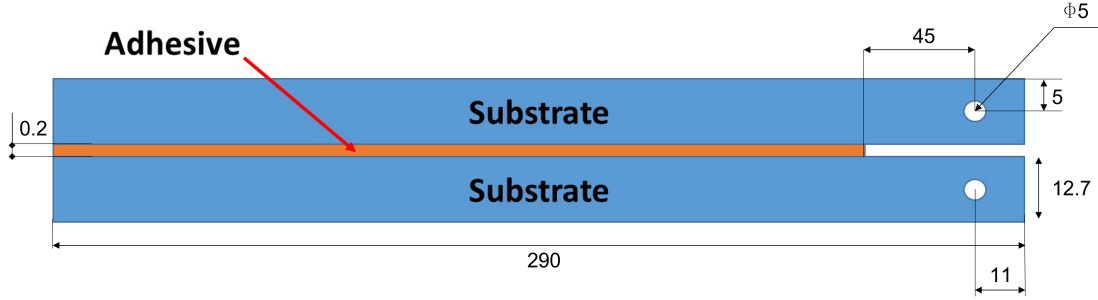


Figure 22: DCB drawing, dimensions in mm (not to scale).

Arcan

Arcan joints were used to study the effect of the ageing mechanism and high temperature service on the fatigue life of the adhesive. The geometry of the Arcan substrates, made from stainless-steel AISI 316, can be seen in Figure 23. Arcan joints were manufactured using a specific mould, presented in Figure 24. The adhesive layer thickness was set to 0.2 [mm] using a stainless steel spacer. During the curing process the joints were kept under pressure to avoid any movement or misalignment.

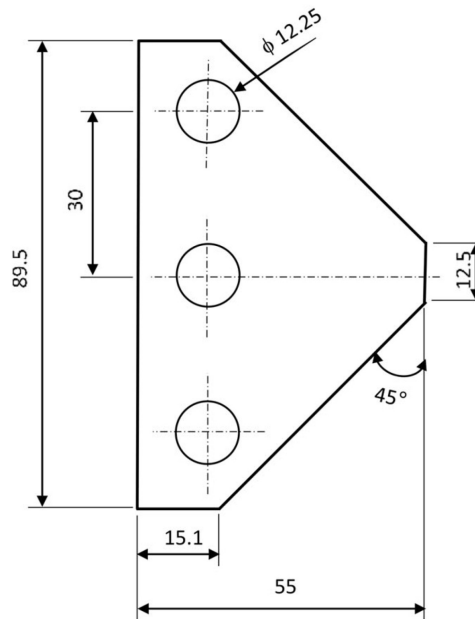


Figure 23: Arcan drawing, dimensions in mm.

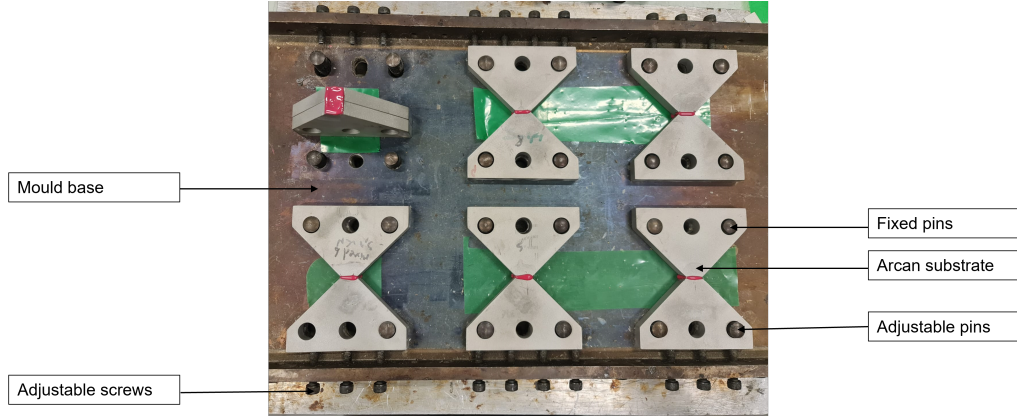


Figure 24: Arcan mould.

3.3 Curing cycle

Figure 25 shows the curing cycle used, which features a stage of 24 hours at room-temperature followed by a post-curing stage of 8 hours at 80 °C. The main objective of the post-curing process is to increase the adhesive's glass transition temperature, T_g .

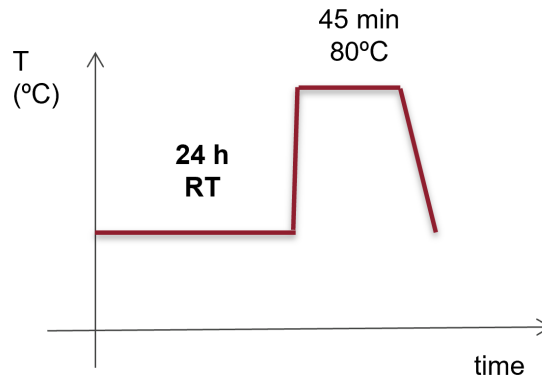


Figure 25: Curing cycle used for the adhesive through the tests.

3.4 Ageing procedure

To ensure the homogeneity of the water level in the tested samples before the ageing process takes place, all the samples were kept in silica for two days to remove the free water absorbed by the adhesive layer during the manufacturing process and the curing cycle. Following this step, all joints were kept in an oven at 50 °C in closed containers filled with distilled water, for a total of 15 days. This temperature is used to accelerate the ageing mechanism.

3.5 Testing conditions and methods

In this section, the parameters used in each test and the different methods to treat the data will be explained. The Table 4 and 5 shows all the tests performed and the respective conditions.

Table 4: Testing conditions for strength and fracture tests.

Strength	Tensile, bulk test	Unaged tested at room temperature 15 days ageing, tested at room temperature Unaged, tested at 50 °C
	Shear, TAST	Unaged tested at room temperature 15 days ageing, tested at room temperature Unaged, tested at 50 °C
Fracture	Mode I, DCB	Unaged tested at room temperature 15 days ageing, tested at room temperature Unaged, tested at 50 °C
	Mode II, ENF	Unaged tested at room temperature 15 days ageing, tested at room temperature Unaged, tested at 50 °C

Table 5: Testing conditions for Arcan tests.

Static	Mode I	Unaged tested at room temperature 15 days ageing, tested at room temperature Unaged, tested at 50 °C
	Mode II	Unaged tested at room temperature 15 days ageing, tested at room temperature Unaged, tested at 50 °C
	Mixed Mode (45°)	Unaged tested at room temperature 15 days ageing, tested at room temperature
Fatigue	Mode I	Unaged tested at room temperature 15 days ageing, tested at room temperature Unaged, tested at 50 °C
	Mode II	Unaged tested at room temperature 15 days ageing, tested at room temperature Unaged, tested at 50 °C
	Mixed Mode (45°)	Unaged tested at room temperature 15 days ageing, tested at room temperature

For bulk tensile tests, an universal testing machine (INSTRON® 3367) was used to apply load until the failure of the specimen. The displacement rate was set to 1 [mm/min]. To measure the strain, an extensometer was used. For the high

temperature tests, an environmental chamber was used. With the generated stress-strain curve, it becomes possible to extract the maximum stress, the strain at failure and the Young's modulus. Figure 26 shows a test at high temperature.



Figure 26: Tensile test setup for high temperature [50 °C].

TAST tests generate pure shear stresses to the adhesive layer, via the application of a tensile load to the specimen. In this case, a digital image correlation (DIC) technique was also used to determine the strain field, extracted with the GOM[®] Correlate software. With the values from the load cell and the data from the images, stress-strain curves were obtained. This data was used to calculate the maximum shear stress, the shear strain at failure and the shear modulus (G).

For fracture characterization, DCB and ENF tests were performed, seeking to determine the fracture toughness in mode I and mode II, respectively. To perform the different tests, only the loading mode is changed, as Figure 27 shows. The tests were carried out in the same machine used for the tensile strength tests, but the displacement rate was set to 0.2 [mm/min]. To process the data from the testing machine and to calculate the energy release rate, the compliance-based beam method (CBBM) was used; more details on this method are in Chapter 3.6. Figure 28 shows the setup from both fracture tests.

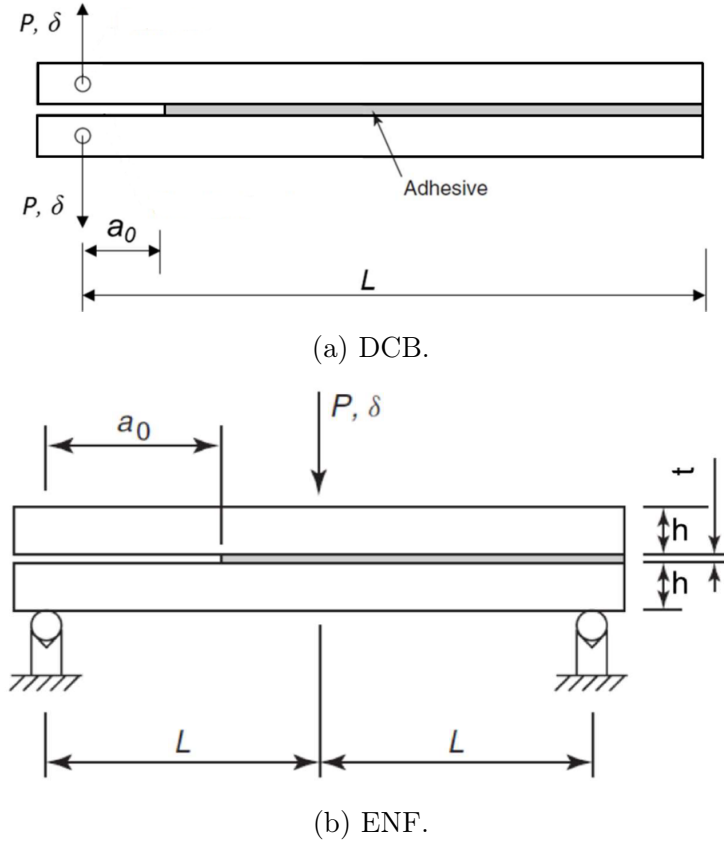


Figure 27: Schematic representation of fracture tests.



Figure 28: Fracture tests set up.

Finally, for the Arcan tests, a servo-hydraulic machine was used (INSTRON[®] 8801). The specimens were tested in both unaged and after 15 days ageing conditions, and also for high temperature [50°C]. The different mode mixities considered were pure mode I, pure mode II, and 45°C, which theoretically is a balance were both pure modes have the same weight; Figure 29 shows the Arcan apparatus used for testing. Also in this case, for the high temperature tests, a simple chamber also

developed during this project was used (see Chapter 3.5.1). For the fatigue testes, the frequency was set for 10 [Hz] and the Load Ratio, R , to 0.1, where:

$$R = \frac{L_{\min}}{L_{\max}} \quad (1)$$

being L the Load.

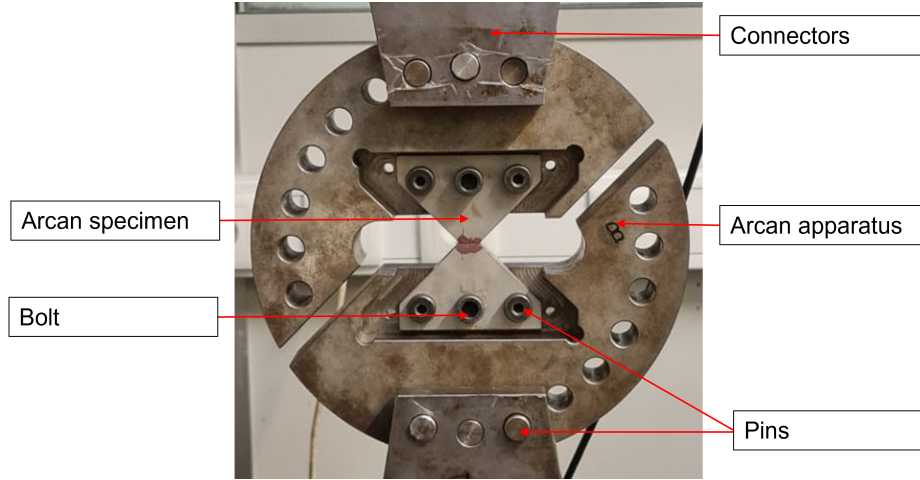


Figure 29: Arcan apparatus.

3.5.1 Thermal chamber development

The thermal chamber used for the strength tests was determined to be too small to fit the fracture specimens and also the Arcan apparatus and, for this reason, it found to be necessary to develop a new thermal chamber to support these procedures.

In this design process, it was first necessary to define the minimum dimensions necessary to fit all the specimens and associated fixtures and, at the same time, to ensure that the chamber is suited for connection to the servo-hydraulic machine. Accordingly, all the equipment that had to fit inside the chamber was measured to help define the minimum dimensions of the internal size of the chamber. Concurrently, the maximum dimension were limited by the clearance available in the servo-hydraulic testing machine (see Figure 30).

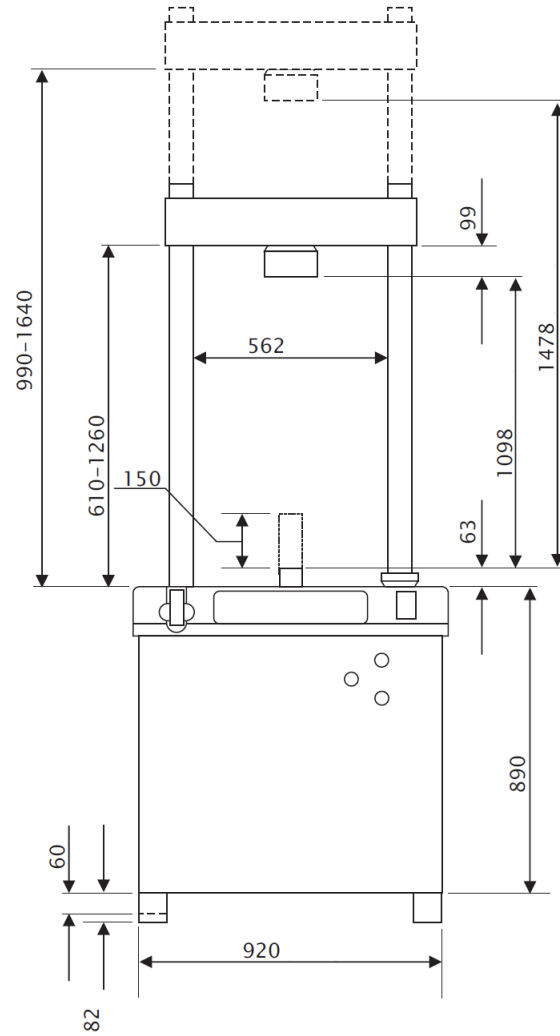


Figure 30: Servohydraulic machine sketch, dimensions in mm. Adapted from [36]

To simplify assembly, the chamber was composted of two separate halves, using a double wall construction, with an insulation material filling the gap. Riveting was used to assemble these halves. Figure 31 show the final design of a chamber halve.

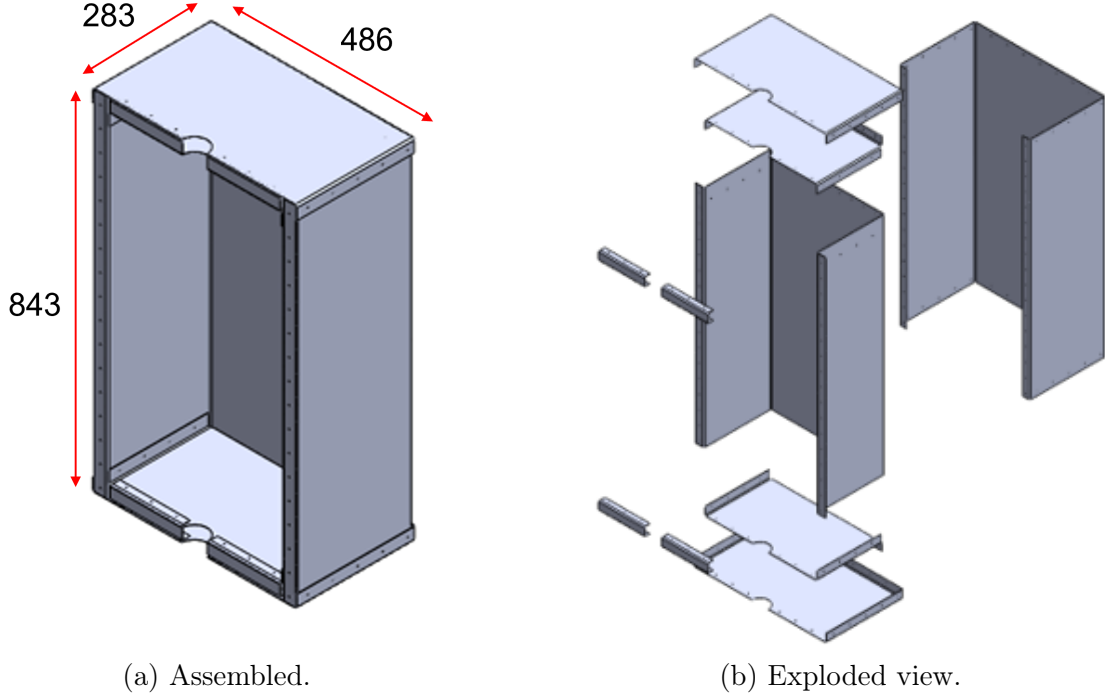


Figure 31: Half chamber design, dimensions in mm.

The final thickness of the chamber walls was the result of material selected for the insulation purposes. Based on the availability, stone wool was selected as the insulation material. Using a the electrical-thermal analogy, and taking into account the thermal conductivity of the insulation, a simple calculation was performed to determine the ideal ($\eta = 100\%$) heat losses:

$$\dot{Q} = \frac{\Delta T}{\sum R} \quad (2)$$

where $\dot{Q}$ is the heat flux, ΔT the difference between the temperature in the chamber and outside, and R the equivalent resistance calculated by the expression:

$$R_{cond} = \frac{L}{K \cdot A} \quad (3)$$

being L the thickness of the material, A the contact area and k the thermal conductivity.

Considering a $\delta T = 60^\circ C$, and for a $30mm$ thickness of stone wool and $1.5mm \times 2$ of steel, the $\dot{Q} = 109[W]$. If a worst case scenario is considered, where $\eta = 10\%$, $\dot{Q}_{real} = 1090[W]$. This chamber is designed to be heated with a commercial heat gun, which has over $2000W$ of power. Based on these calculations, the insulation should be sufficient to meet the design requirements.

Stainless steel was selected as the main construction material of the chamber, ensuring corrosion resistance and a relatively low thermal conductivity. The selected stainless steel was AISI 304 was chosen for the chamber construction since the

cost/benefit relation was the best between the options available. The steel sheet thickness was based on commercial chamber's models available in the laboratory, such as the one used for the strength tests. Figure 32 shows the chamber during its assembly process, where the use of the stone wool as insulation material can be seen.



Figure 32: Thermal chamber during the assembly process.

Finally, to design the riveted joints, a calculation was done based exclusively on the steel sheet thickness, since the only load the chamber has to support is its own weight. Riveted joints were designed according to ISO 14588:2000 [37]:

$$d_{rivet} \geq 1.5 \cdot S \quad (4)$$

where S is thickness of the smallest from the two sheets to join.

According to this equation, the minimum normalized diameter for the rivet would be $d = 2.4[mm]$; however, due to availability from the suppliers, $d = 3.2[mm]$ was the selected size. The rivets chosen were also made out of stainless steel to avoid corrosion (see Figure 33).



Figure 33: Rivet detail in the thermal chamber.

Once the chamber was assembled, it was mounted on the testing machine and a simple temperature stability test was performed. The chamber was instrumented with a type K thermocouple and a thermal camera was used to control the temperature in the external surface of chamber. The test started by heating the chamber until a temperature of 50 °C was reached, upon which the heat source was turned off (see Figure 34). It was then determined 11 minutes were needed for temperature to cool down 1 °C (until 49 °C). The exterior wall of the chamber was found to no surpass 32 °C.

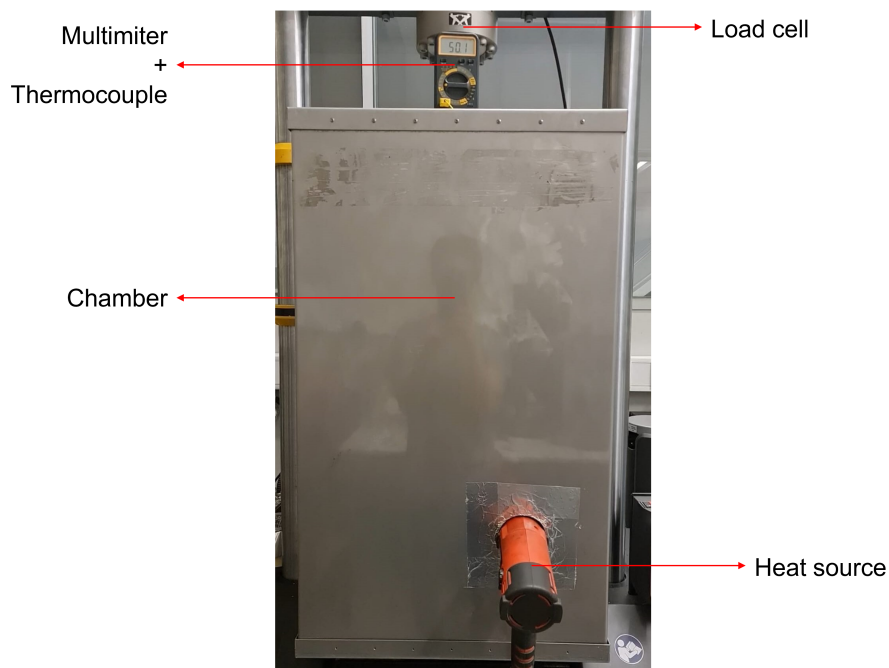


Figure 34: Thermal chamber setup.

3.6 CBBM - Data reduction scheme

In order to obtain the energy release rate in both mode I and mode II, a data reduction scheme must be applied [38]. Two main classical methods have been developed based on the Irwin-Kies equation. The first is the Compliance Calibration Method (CCM) [39,40], :

$$G_I = \frac{P^2}{2b} \frac{dC}{da} \quad (5)$$

where cubic polynomials are used to fit the compliance ($C = C_3 \cdot a^3 + C_2 \cdot a^2 + C_1 \cdot a + C_0$) as a function of the crack length (a). The second is the Corrected Beam Theory (CBT) [40–44], also derived from the previous equation:

$$G_I = \frac{3P\delta}{2b(a + |\Delta|)} \quad (6)$$

but with a correction factor Δ , proposed in 1992 by Wang and Williams [41], to take into account the crack tip rotation and deflection:

$$\Delta = h \sqrt{\frac{1}{13k} \left(\frac{E_x}{G_{xy}} \right) \left(3 - 2 \left(\frac{\Gamma}{1 + \Gamma} \right)^2 \right)} \quad , \quad \text{with} \quad \Gamma = \frac{\sqrt{E_x E_y}}{k G_{xy}} \quad (7)$$

where h is the thickness of the substrate, E_x and G_{xy} its longitudinal normal and shear modulus, E_y the modulus in the thickness direction, and k the shear stress distribution constant for correcting the deflection caused by the shear forces.

To determine the necessary crack length correction, a linear regression can be obtained experimentally after loading the specimens under three different load levels, which can be graphically represented in Figure 35.

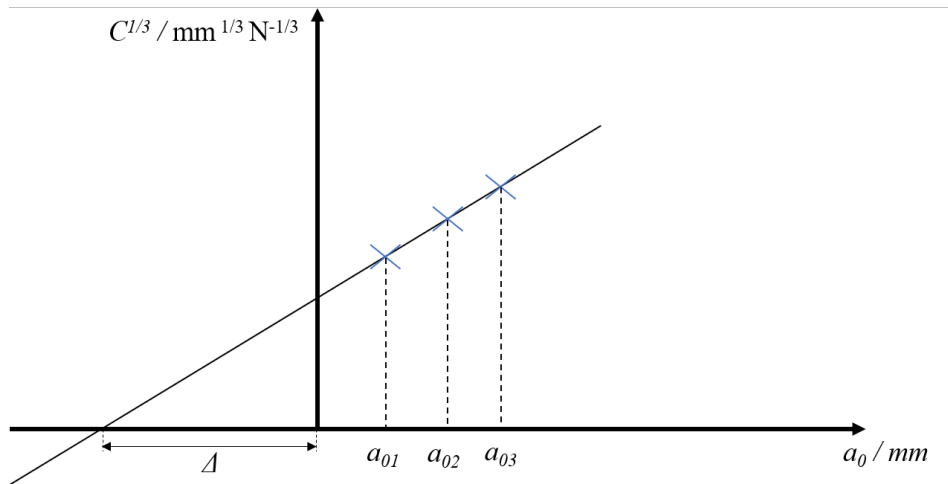


Figure 35: Graphical representation of the method used to determine the crack length correction, Δ . Adapted from [45].

However, both these methods are dependent on the assessment of the exact crack length, increasing the difficulty of their implementation, especially when sudden crack propagation occurs and/or when analysing mode II fracture [39, 40, 46].

Considering that, the crack tip does not fully behave accordingly to linear elastic fracture mechanics, there is the need to consider a fracture process zone (FPZ), seen in Figure 36, that develops ahead of the major crack tip caused by plasticization and nucleation of several micro-cracks. This phenomenon is more severe in ductile materials since the energy dissipated in the FPZ increases, necessitating its inclusion in the data reduction scheme [38].

Accordingly, the compliance-based beam method (CBBM) was the one chosen to process the fracture toughness data, since it is based on the concept of equivalent crack length, which accounts for the previously described phenomena [38, 47], and only requires data on the compliance of the specimen during the test.

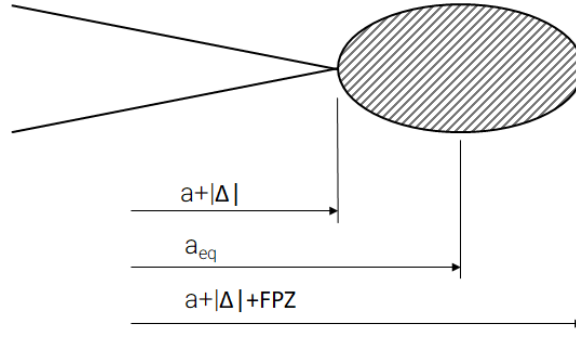


Figure 36: Schematic representation of the equivalent crack length and fracture process zone concepts. Adapted from [38].

Using this method, the energy release rate in mode I is defined as [38]:

$$G_I = \frac{6P^2}{b^2h} \left(\frac{2a_{eq}^2}{h^2E_f} + \frac{1}{5G_{13}} \right) \quad (8)$$

being P the applied or measured load depending on the type of loading condition, b the width of each specimen, h is the specimen height, G_{13} is the respective shear modulus of the substrates, a_{eq} is the equivalent crack length, whose value is given by the solution of an equation derived from Timoshenko's beam theory [38]:

$$C = \frac{\delta}{P} = \frac{8a_{eq}^3}{bh^3E_f} + \frac{12a_{eq}}{5G_{13}bh} \quad (9)$$

and E_f is the corrected flexural modulus, the purpose of which is to correct the fact that this method does not take into consideration the stress concentration and substrates' rotation close to the crack tip [38]:

$$E_f = \left(C_0 - \frac{12(a_0 + |\Delta|)}{5bhG_{13}} \right)^{-1} \frac{8(a_0 + |\Delta|)^3}{bh^3} \quad (10)$$

being 0 the index set for initial values - where a_0 is the initial crack length previously defined for each method and C_0 the initial linear compliance - and Δ a correction factor for the crack length derived from the one used in the CBT method given by the Equations 7, considering an isotropic material and 0.85 for the constant k [48].

Mode II fracture energy can be determined by [49]:

$$G_{II} = \frac{9P^2 a_{eq}^2}{16b^2 E h^3} \quad (11)$$

where

$$a_{eq} = \left(\frac{a_0^3 C_c}{C_{0c}} + \left[\frac{C_c}{C_{0c}} - 1 \right] \frac{2L^3}{3} \right)^{1/3} \quad (12)$$

with

$$C_c = C - \frac{3L}{10bhG_{13}} \quad (13)$$

and

$$E = \frac{3a_0^3 + 2L^3}{8bh^3 C_{0c}} \quad (14)$$

being L half of the ENF specimen total length and index c indicates the corrected value of a variable.

4 Numerical model

To investigate the water content present in the adhesive layer in Arcan joints, a numerical analysis using FEM (finite element method) was conducted with the commercial software Abaqus. The water diffusion into the adhesive layer is calculated using an analogy between mass and heat transfer, where the water uptake level in each point is related to the nodal temperature and the diffusion coefficient, D , obtained by the analytical application of Fick's Law [50] over the experimental results using MATLAB.

The model used was simplified to enable a finer mesh. For that, only $\frac{1}{2}$ of the total joint was modelled, following the use of symmetry conditions. Figure 37 shows the model used.

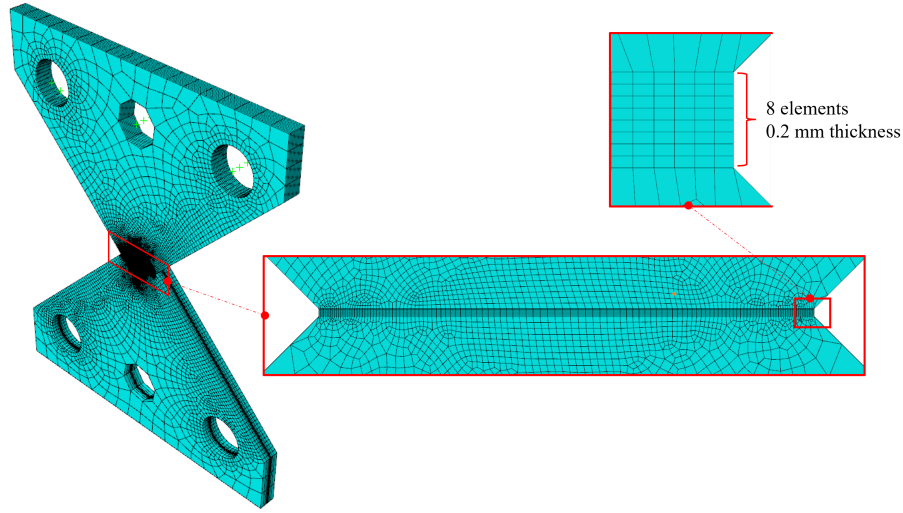


Figure 37: 3D Arcan model.

Stress on the Arcan joints subjected to high temperatures was evaluated using a 2D model (see Figure 38) since the properties along the bondline are constant. The elastic properties used were obtained by performing strength at the same temperature. For this case, a corrected effective stress adapted from other study [51] was calculated, based on the values from von Mises and hydrostatic stresses. That parameter takes into account the effect of the temperature and this data was later used relate S-N master-curves at different temperatures (see Appendix B).

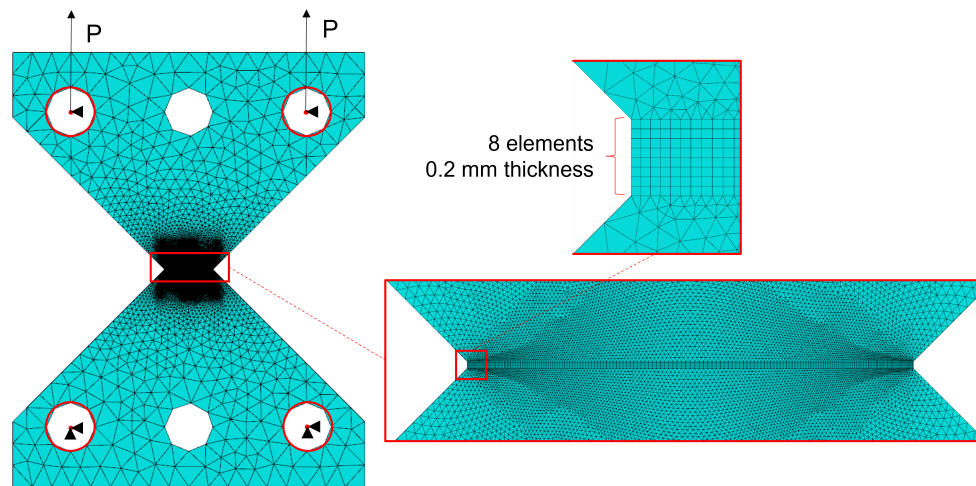


Figure 38: 2D Arcan model.

5 Results and discussions

Overall, the results from this study showed that the environmental conditions play a significant role in the adhesive properties and in the joint's behaviour. As a result, it is crucial to understand what are the effects that those parameters will have on the joint's performance.

5.1 Effect of ageing

When in contact with humid environments, adhesive are known to absorb water. The results showed that the epoxy adhesive under analysis experienced a two stage water absorption, which can be explained by the relaxation phenomenon on polymers, a result of its viscoelastic behaviour, as Figure 39 shows.

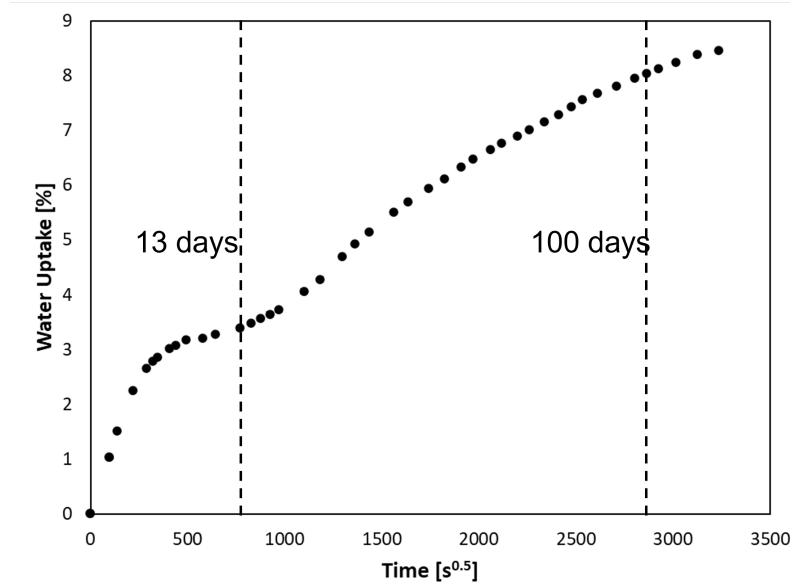


Figure 39: Moisture uptake curve for H₂O in the adhesive.

Figure 40 shows a summary of the static tests results. The tests showed that the adhesive experiences significant degradation when in contact with water, with this degradation being more severe for tensile/mode I load conditions than for shear/-mode II. TAST and Arcan tests showed similar results for shear conditions, with around 10% of property degradation; for tensile load conditions there was a small difference in the results between the strength tests and the Arcan tests, which can be explained by the fact that strength test for mode I is carried out in bulk material while the Arcan test is in a joint condition. Fracture tests showed a significant difference between mode I and mode II: for mode I, the fracture toughness decreased by 10% with ageing. On the other hand, in mode II, a fracture toughness improvement of 11% was verified. Fatigue tests at 50°C were compared with the result of another study which has tested the same adhesive under the same mode mixities but at room

temperature (although a different curing cycle was used). Results from Figure 41 show that moisture significantly affects life under fatigue loads, indicating that the ageing mechanisms not only weakens the joint, but also increases its sensitivity to cyclic loading conditions.

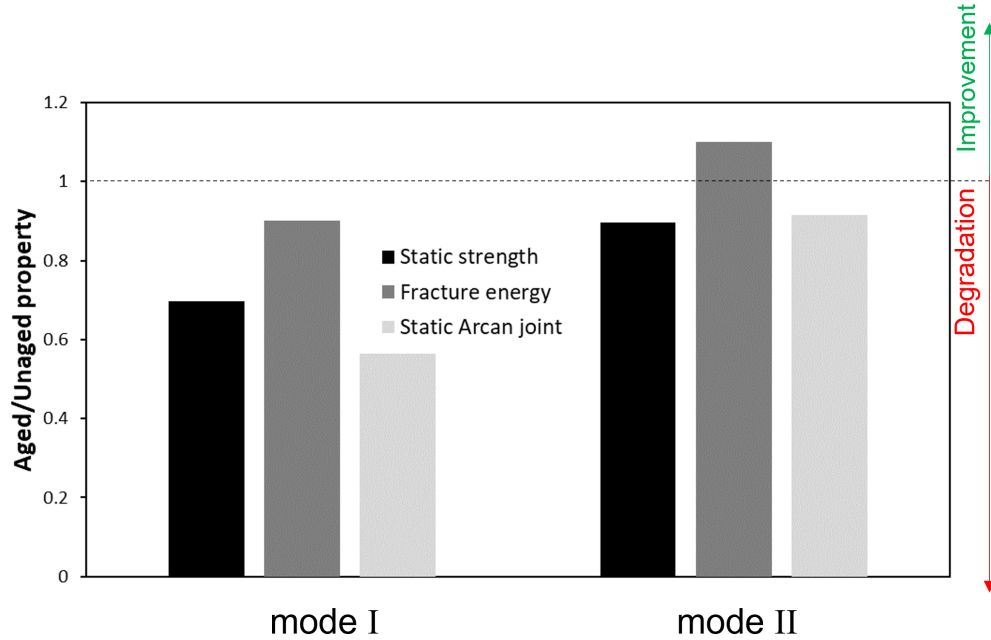
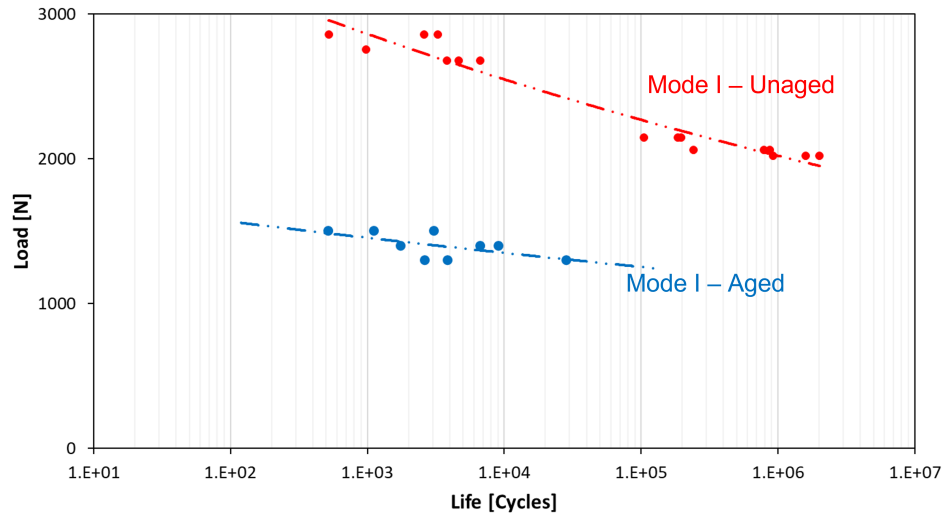


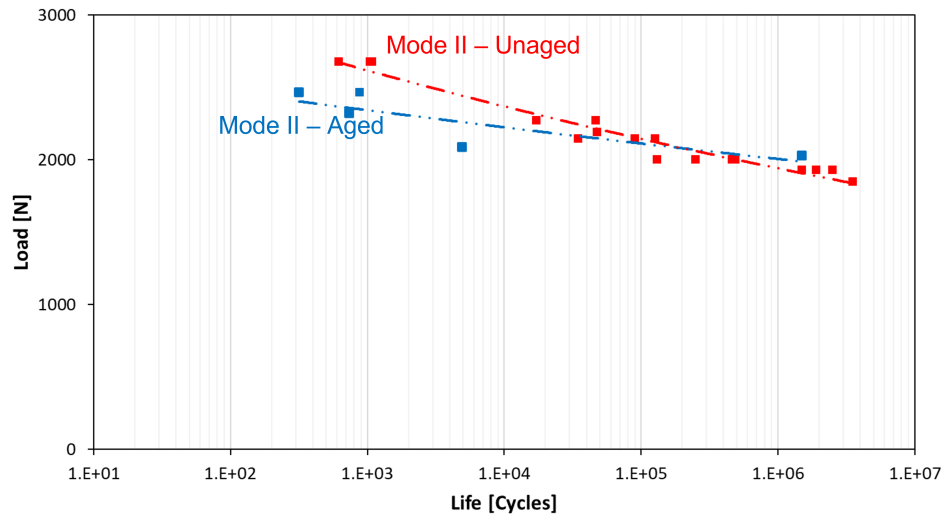
Figure 40: Influence of ageing on adhesive properties for different tests.

By analyzing the fracture surfaces of the Arcan joints, it was found that the ageing mechanism leads to a partial adhesive failure, more significantly at the edges where the ageing level is higher; mode II and mixed mode experienced higher adhesive failure area compared with mode I. A crack path analysis showed in Figure 42 for both modes I and II, the crack initiates at the interface. However, for mode I, it then propagates more through the adhesive layer while in mode II conditions it more often travels along the interface and only then moves towards the adhesive layer.

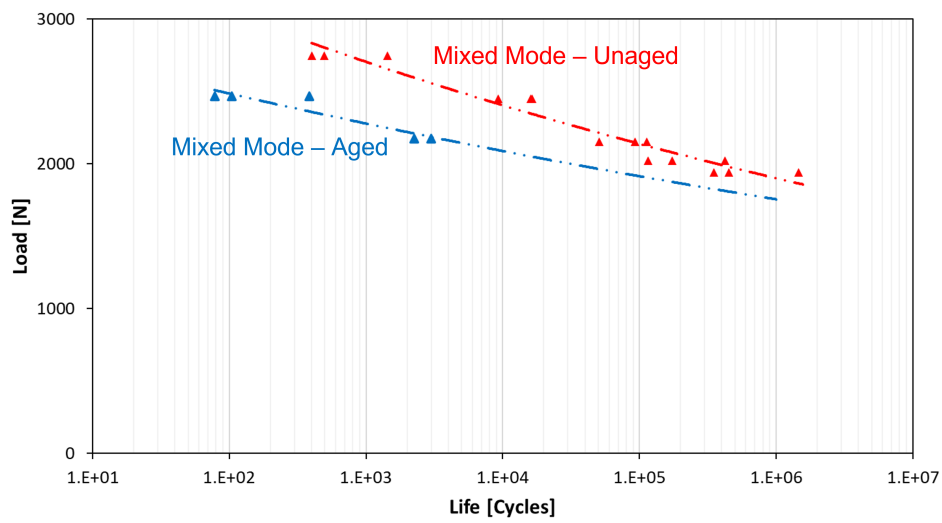
Mode discussion on the effect of ageing on the property degradation of an epoxy adhesive can be found in Appendix A.



(a) Mode I.



(b) Mode II.



(c) Mixed mode (45°)

Figure 41: S-N curves for unaged [52] Vs. 15 days ageing at 50 °C.

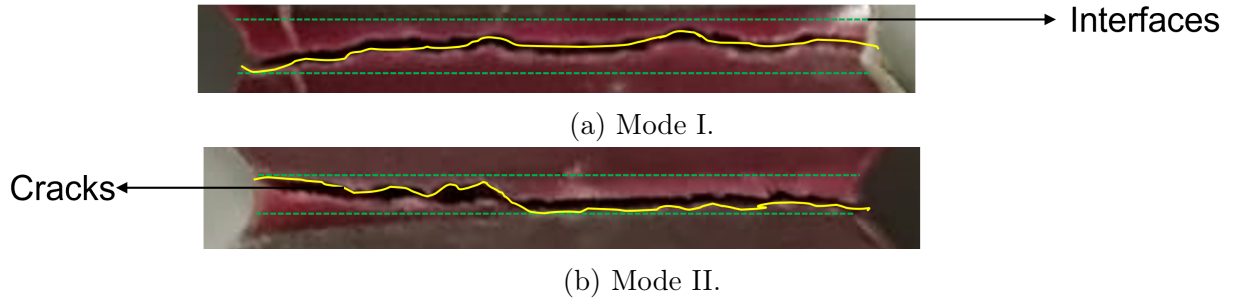


Figure 42: Crack path for aged Arcan joints under fatigue load conditions.

5.2 Effect of temperature

The behaviour of the adhesive at high temperature was noticeably different from that determined for room temperature. Firstly, the results showed the curing cycle plays a significant role on the glass transition temperature, T_g , as Figure 43 shows; however its effect on the mechanical properties is not so significant. This is discussed in more detail in Appendix B.

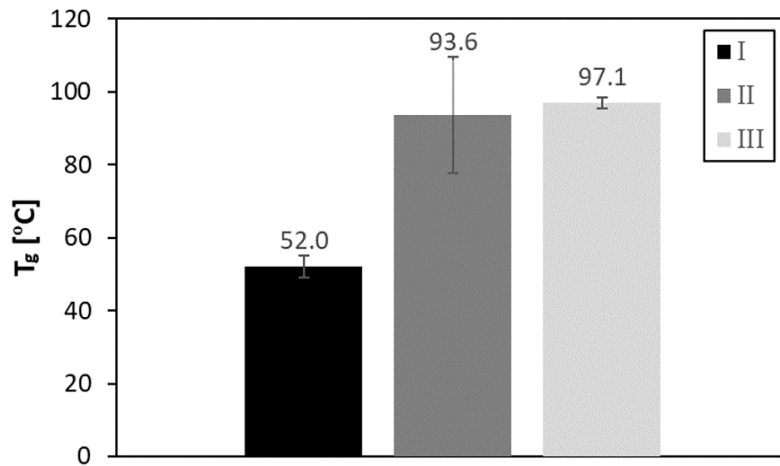


Figure 43: Effect of curing cycle on T_g .

The results from the static and fracture (see Figure 44) tests resulted in some important conclusions: while for mode I the increase of temperature was found to reduce the adhesive properties, for mode II there was in fact a slight improvement. While this was already expected for fracture tests, due to the increase of the ductility of the adhesive with the temperature, this was unexpected for strength testing. Fatigue tests also showed that the increased sensitivity of the adhesive to fatigue load conditions was similar for both loading modes; on the other hand, the sensitivity to cyclic loads was more significant at high temperatures.

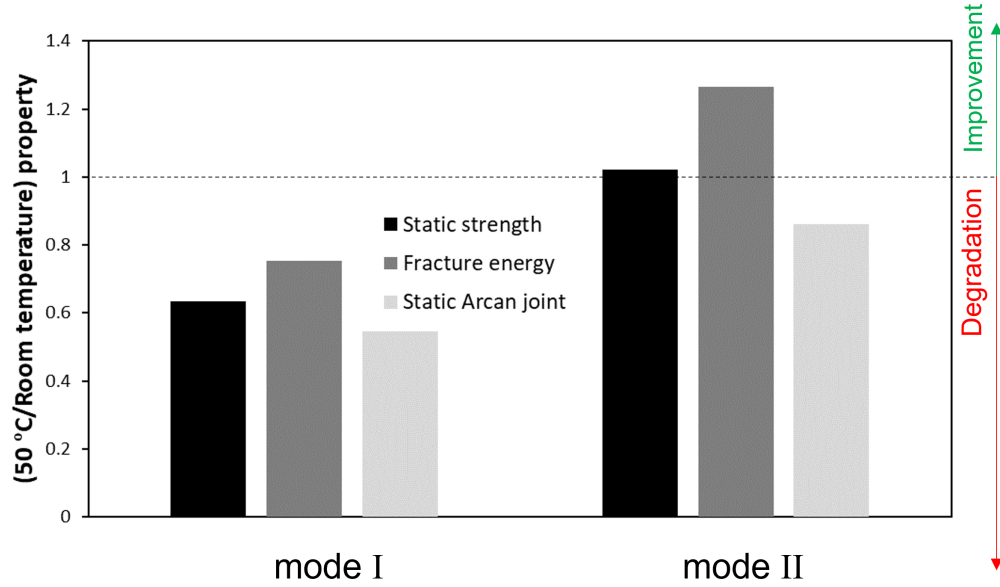
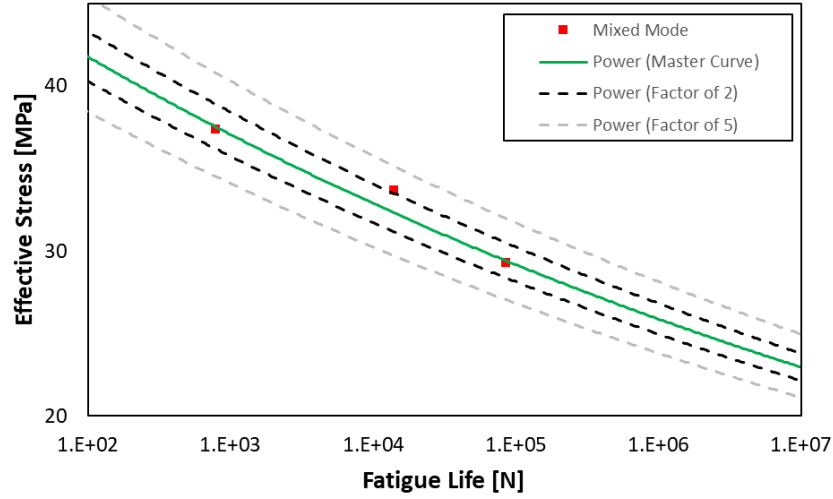


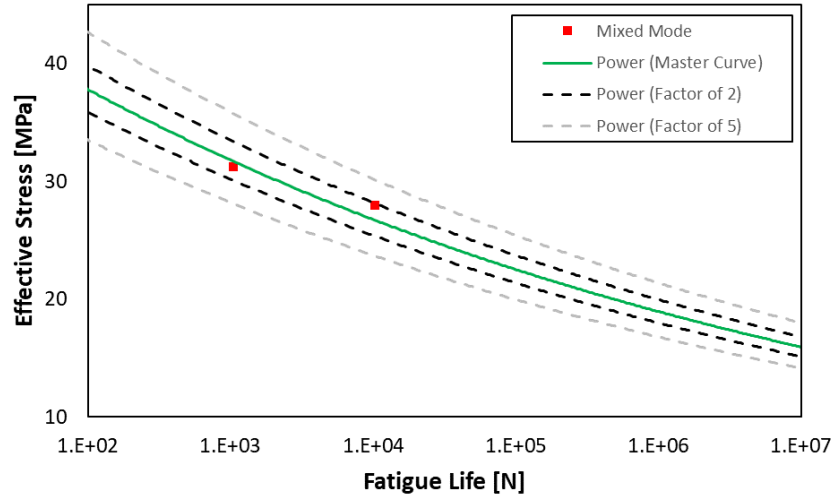
Figure 44: Influence of temperature on adhesive properties for different tests.

Finally, a fatigue life prediction model is proposed as a function of a "corrected effective stress", adapted from a model from Sousa et al. [53], which seeks to take into account the effect of temperature. Results, presented in Figure 45 show that the experimental results from mixed mode (45°) fitted the master curves obtained based on mode I and mode II results.

More discussion on the effect of temperature on the rate of property degradation of the adhesive and about the predictive model proposed can be found in Appendix B.



(a) Room temperature.



(b) 50 °C.

Figure 45: Comparing the predicted and the experimental experimental fatigue lives for mixed mode (45°).

6 Conclusions and future works

6.1 Conclusions

The effects of hygrothermal aging and temperature on different mode mixities on the property degradation of an epoxy adhesive was studied. To do so, multiple joint geometries and loading conditions were used. A fatigue life prediction model was proposed, which combines different mode mixities in one single curve. The model was tested at different temperatures.

The presence of water on the adhesive was found to decrease its mechanical properties. Results showed that 15 days of ageing time are enough to significantly reduce the durability of the joints, even though the water level on the adhesive remains only in around 3%.

Degradation was more severe in pure mode I conditions; in fact, results showed that when shear forces are applied to the adhesive, its sensibility to humidity is much lower for both static (strength and fracture) and fatigue loads. When analyzing the different tests performed, fracture properties were found to be less affected by the ageing than the adhesive's strength. Such discoveries could help to better design joints intended for operation under extreme environmental conditions. It was also verified that the Poisson's ratio of the epoxy underwent an increase in the presence of moisture. Fatigue results showed that moisture affects the rate of change in life, with fatigue load being significantly lower. This indicates that the ageing mechanisms not only weaken the joint, but also increase its sensitivity to cyclic loading conditions.

Regarding the effect of temperature, firstly, it was concluded that the curing temperature is the most significant parameter in the glass transition temperature of the adhesive. On the other hand, the presence of a post-cure at high temperature leads to a decrease in the maximum strain of the adhesive.

When tested at high temperatures, the adhesive experienced some property changes. Results showed that under tensile load conditions, the effect of temperature is significant, while in pure shear load the adhesive is almost insensitive to temperature. The Arcan tests have demonstrated that when subjected to pure mode I conditions, like in the strength tests, the joint experiences a significant drop of performance when compared to mode II conditions. However, both mode mixities appear to have similar sensibility to fatigue load conditions. By comparing those results with those of the DCB and ENF tests, results showed that the fracture toughness sensibility to temperature is much lower than that of the strength properties; in fact, G_{IIc} even improved by increasing the temperature.

Fatigue Arcan tests again corroborated the fact that the adhesive performs better under pure mode II conditions at high temperature. According to the S-N results, fatigue strength as a function of the static strength is expected to be higher for

mode II than for mode I.

Finally, the proposed fatigue model was able to predict the fatigue life of Arcan joints subjected to different mode mixities. The same model was tested for two different temperatures and both results were satisfactory.

6.2 Future works

There are many factors that can affect the performance of adhesive joints. From those factors, environmental conditions are still perhaps the most poorly understood. As a result, would be interesting to expose different adhesive systems to hygrothermal aging, to understand if the nature of the adhesive system (epoxy, polyurethane, silicon, etc.) plays a significant role on ageing resistance. Another aspect that can significantly influence the quality of adhesive joints is joint geometry: it is known that different geometries can lead to different stress distribution and different stress concentrations.

Additionally, it could be interesting to extend the study of the effect of the temperature on property degradation of adhesive joints to temperatures closer to T_g , or even above, to understand if a similar trend was followed. Finally, it would also be interesting to test the proposed model for fatigue life prediction for wider range of temperatures, to prove its validity and also to seek some sort of relation that governs this behavior.

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Appendices

A Paper A

The interaction of loading mode and humidity on the properties degradation of an epoxy adhesive subjected to strength, fracture, and fatigue tests

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ABSTRACT

Hygrothermal ageing has a significant impact on the performance of bonded joints. Despite extensive investigations into the ageing effect of bonded structures, there is no comprehensive study that examines the rate of the ageing effect as a function of the loading type and the load angle.

Accordingly, in this study, an epoxy adhesive was tested under different loading types/angles including tensile and shear standard strength tests, mode I and mode II fracture tests, and fatigue and static Arcan tests. All these experiments were performed for both aged and unaged conditions. To understand the influence of loading angle on the failure response of aged bonded joints, Arcan samples were subjected to mixed mode loading conditions as well. A fractography analysis was conducted to provide better understanding of the failure mechanism of aged samples loaded at different modes. Based on these results, it was found that mode I (tensile loading conditions) is more sensitive to the aging conditions than mode II (shearing) for all loading types (strength, fracture, and fatigue tests). The influence of moisture on fracture toughness was however less relevant than on strength. The results also showed that for the same loading mode and for the same Arcan joints, the fatigue strength is more degraded by aging conditions than the static strength.

A.1 Introduction

The use of adhesive bonding processes is growing for various industrial sectors, ranging from the automotive industry to the aerospace field. Compared with conventional joining techniques, such as bolting or riveting, adhesive bonding provides many important advantages. Key among these are the more uniform stress distributions, weight saving potential, the ability to join dissimilar materials and increased fatigue performance [1]. However, despite the multiple advantages of adhesive joints, the considerable reduction in mechanical performance they exhibit in humid environments still poses hindrances to its use in many critical applications. At the same time, under service conditions joints are often subjected to cyclic loads, leading to a mechanical behaviour and a failure process which is mostly governed by fatigue failure mechanisms. Accordingly, for a safe joint design, the influence of ageing conditions on both static and fatigue behaviour should be thoroughly investigated.

The effect of the humidity level on the performance of adhesive joints is currently one of the most active research subjects in the field of bonded joints. To analyse this effect, it is first essential to fully understand the hygrothermal ageing mechanism and learn how to precisely replicate it in laboratory conditions. Multiple authors have treaded this path and have shown that degradation due to the ageing mechanisms cannot be avoided when adhesive is used in humid conditions for long periods [2–5]. Wei et al. [6] studied the hygrothermal ageing effect in a few types of adhesive joints with aluminium substrates. They found that the failure mode is a function of ageing time. Zhang et al. [7] also studied the shear properties degradation under hygrothermal ageing conditions. Hirulkar N.S. et al. [8] studied the effect of hygrothermal ageing on the failure load of single lap joints for different adhesives. The authors concluded that the exposure time and the temperature are the main parameters responsible for property degradation for all adhesives. Loh et al. [9] analyzed the interfacial properties and found that the interfacial fracture energy decreases by increasing the moisture levels. This research team measured the interfacial strength on a epoxy-steel interfaces. Also, Borges et al. [10] concluded that the water absorption can occur by the interface adhesive-substrate, but also by the cracks and voids present in the adhesive layer.

The fracture toughness is an important material property that must be taken into account when studying an adhesive's performance [11,12]. The effect of ageing on this property has also been investigated in multiple research works. Sugiman et al. [13] studied the effect of ageing on the on mode II fracture toughness, concluding it increases on a first stage until the water content on the adhesive reaches 1.8% and, after that, it starts decreasing. Fernandes et al. [14] concluded the type of water has a significant influence on the property degradation on the adhesive's G_{Ic} ; for distilled water, they observed a property degradation, while for salt water the fracture toughness of the adhesive increased.

Although most of this research is mainly focused on continuous ageing conditions, in service joints can experience a cyclic ageing environment. Accordingly, some authors have studied the effects of cyclic humidity on the strength and fracture of adhesive joints [15,16]. Moazzami et al. [15] subjected an epoxy-based adhesive to cyclic ageing conditions. The results showed a decrease on both elastic modulus and tensile strength with the aging cycles. da Costa et al. [17] studied the impact of single aging and cyclic ageing on the properties of an epoxy-based adhesive, concluding that the first ageing cycle is the one with more impact on the decrease of both the Young's modulus and tensile strength, with the cycles afterwards having a lower impact on the adhesive degradation process.

Cyclic loading conditions are also known to significantly degrade the mechanical properties of adhesive materials [18]. When a fatigue loading is combined with ageing conditions, this usually results in a premature failure of the joint. The effect of moisture on the fatigue behaviour on aluminium substrates bonded with two different adhesives was studied by Costa et al. [19] using double cantilever beam (DCB) specimens. Their results showed the fatigue life decreases with the increase of water level at the joint. Some authors [20,21] studied the hygrothermal ageing on the fatigue behaviour using hybrid joints with aluminium and carbon fiber reinforced polymer (CFRP) as substrates, concluding the presence of water in the adhesive layer leads to a loss of the interfacial strength, affecting the fatigue performance of the joints. The authors also concluded that this effect is more obvious in the early ageing stages.

Some authors have studied the effect of loading angle on the static and fatigue response of adhesive joints [22–24]. Sousa et al. [24] studied the influence of different mode mixities on the fatigue life of an epoxy-based adhesive. From the experimental results they concluded that the fatigue life of the joint was reduced with the introduction of a shear-loading component. da Costa et al. [25] conducted some studies on the effect of adhesive and interfacial ageing on the S-N response in pure mode I loading conditions in Arcan joints, finding that a short ageing time drastically reduced the fatigue life of bonded joints in case of a local or a complete interfacial failure due to the ageing phenomenon. In order to surpass this weakness, Chen et al. [26] studied how a sealing treatment can improve the resistance of lap joints to ageing; the results were satisfactory, specially for long ageing times.

Although, as shown above, a large number of studies has been carried out to assess the behaviour of bonded joints subjected to humidity and different loading conditions, determining the rate of property degradation for aged samples as a function of loading type/angle has not yet been carried out. Accordingly, the aim of the current study is to better understand how the loading angle affects the properties of an adhesive under hygrothermal ageing. For this purpose, an epoxy adhesive was subjected to different loading angles, including standard strength tests, fracture

tests, static Arcan tests and S-N experiments using Arcan joints. An analysis of the fracture surface of Arcan joints for both fatigue and static loading conditions was also conducted, seeking to understand the effect of ageing on the failure mechanisms as a function of loading mode.

A.2 Experimental details

In this research work, an epoxy-based adhesive was tested under static and fatigue loading conditions at different mode mixities, using Arcan joints. Furthermore, strength and fracture testing at different loading modes was also carried out to assess the role of loading angle on the rate of property degradation in aged bonded assemblies.

A.2.1 Materials

A two-component epoxy based structural adhesive was used to study the effect of ageing and of the different mode mixities on the fatigue response of Arcan joints. The studied adhesive is suitable to join both metallic and composite substrates. The key mechanical properties of the adhesive are shown in Table 1. The Arcan substrates were cut from a stainless-steel AISI 316 bar in order to avoid oxidation [27].

Table 1: Mechanical properties of the adhesive

Properties	Adhesive
Maximum tensile strength, MPa	33.4+0.5
Maximum tensile strain, %	6.1+0.7
Maximum shear strength, MPa	22.9+1.1
Maximum shear strain, %	31.0+10.6
Young's modulus, E	1706+42
Shear modulus, G	649+9
Poisson's ratio, ν	0.32
Density, $g \cdot cm^{-3}$	1.07

A.2.2 Geometry and manufacturing procedure

The geometry of the Arcan specimens is shown in Figure 1. The Arcan joints were manufactured using a specific mould, shown in Figure 2 and the adhesive layer thickness was set at 0.2 mm, controlled using spacers. To ensure optimal adhesion, the surfaces were first sandblasted and then cleaned using acetone. All spacers were

also cleaned before contact with the substrates to avoid any contamination. During the curing process, weights were used to restrict the movement of the specimens.

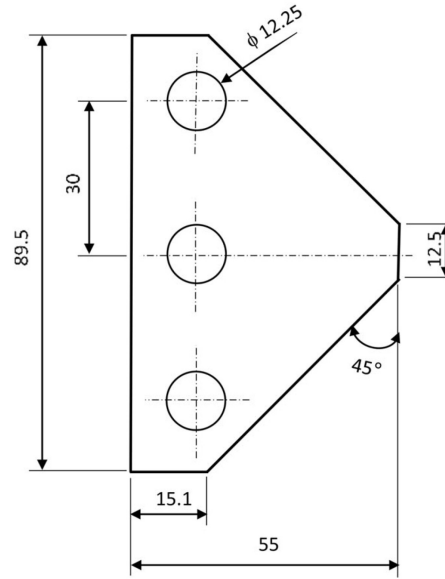


Figure 1: Arcan drawing, dimensions in mm.

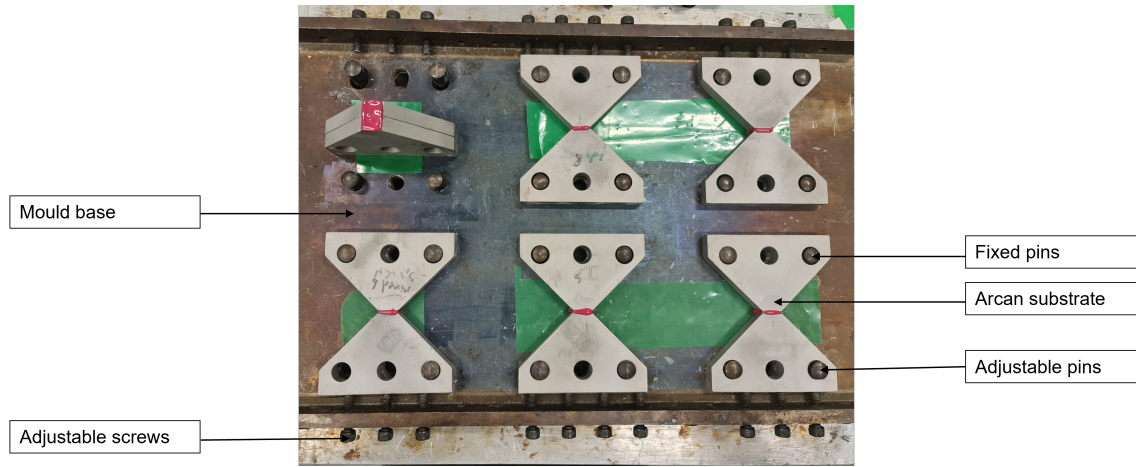


Figure 2: Mould used for Arcan joints production.

Bulk tensile tests were also performed to evaluate the adhesive's ultimate tensile strength and to measure the Young's modulus of the adhesive before and after ageing. These tests were carried out according to the British standard (BS2782) [28]. To manufacture these specimens, a 2 mm adhesive plate was produced using a specific mould. Figure 3 shows the dimensions of the tested bulk samples. To evaluate the behaviour of the adhesive under shear loading conditions, thick adherend shear test (TAST) specimens were manufactured and tested following the international standard ISO 11003 2:2019 [29]. To control the substrate alignment and also the adhesive thickness during the curing process, a specific mould shown in Figure 4a was used. Figure 4b shows the specimen dimensions. DIC (digital image correlation)

technique was used to precisely measure the shear strain. Accordingly, a speckled pattern was painted on the specimens, as shown in Figure 4c.

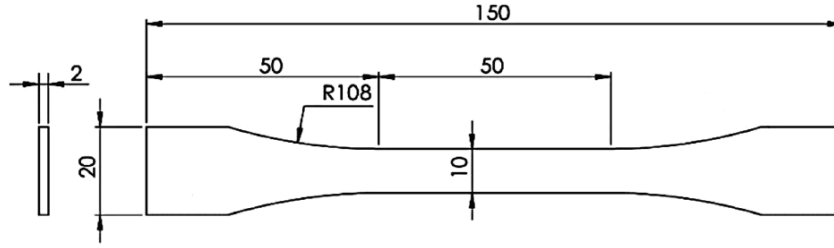
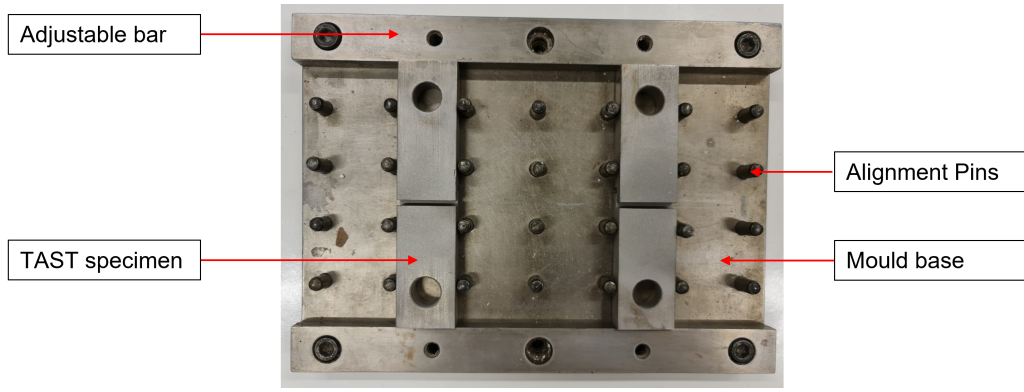
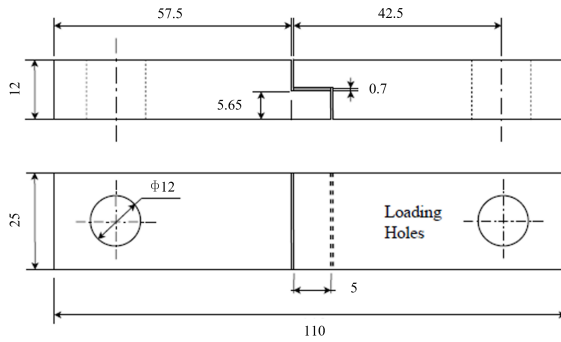


Figure 3: "Dogbone" tensile bulk specimen, dimensions in mm.

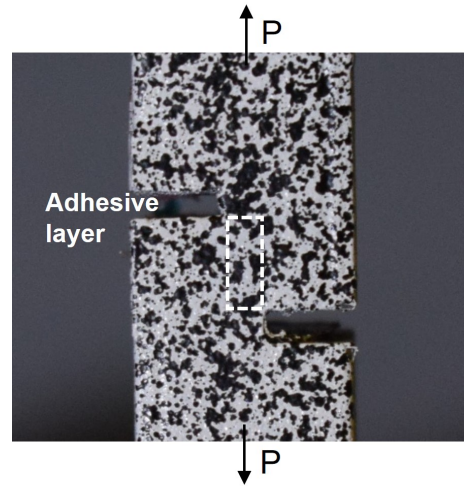
To evaluate the effect of moisture on the fracture behaviour of the adhesive, DCB and end notched flexure (ENF) tests were performed to calculate modes I and II fracture toughness, respectively. The specimens used for both tests are the same, where the only difference is in the loading conditions as the scheme from Figure 5 shows. The joints were produced in a specific mould. To obtain the diffusion coefficient and the saturation level of the adhesive, both to be further used in numerical models, a parallel study was made, using the same conditions described previously. In this case 60 x 60 x 1 [mm] adhesive bulk plates' immersed in water for multiple months and their mass was measured regularly to determine the water ingress rate.



(a) mould used.



(b) joint scheme, dimensions in mm.



(c) speckle used for DIC.

Figure 4: TAST joint.

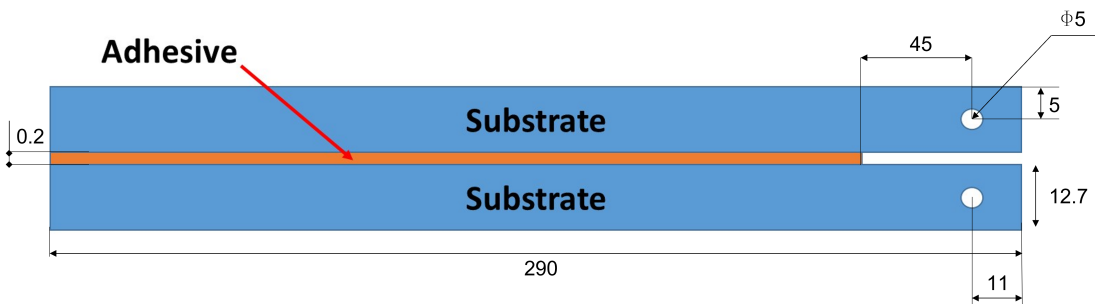


Figure 5: DCB and ENF specimen scheme, dimensions in mm (not to scale).

A.2.3 Curing cycle

The effect of the curing cycle on the adhesive's properties has already been extensively studied by several authors [30, 31], leading to the conclusion that this parameter plays a relevant role not only in the mechanical properties of the adhesive,

but also on its glass transition temperature. For this study, all the joints had the same curing cycle: 24 hours at room temperature, followed by a post-curing process of 8 hours at 80°C, as shown in Figure 6.

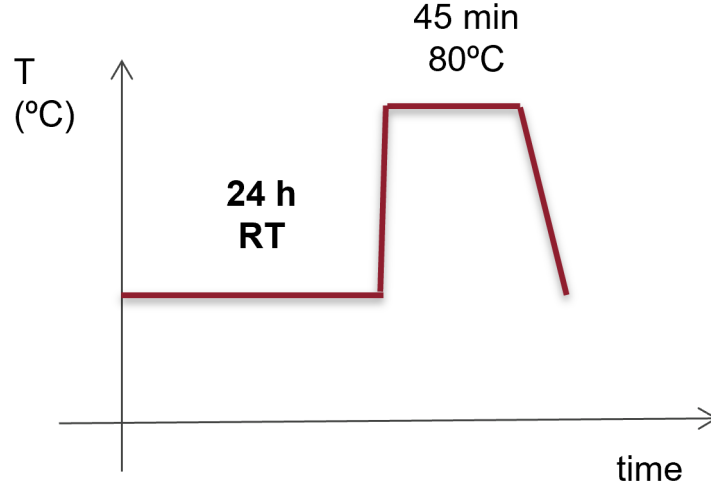


Figure 6: Curing cycle used.

A.2.4 Ageing process

Before the aging process took place all the samples were placed in a container filled with silica particles at room temperature for two days. This was carried out to remove any possible water from the adhesive layer and ensures that all joints start the ageing process at a consistent moisture level, allowing for a better analysis of the water uptake level after the ageing process. After that, the actual hygrothermal ageing process was started, by placing the joints in a container filled with distilled water at 50°C.

A.2.5 Water uptake measurements

To evaluate the diffusion of water through the adhesive layer, adhesive bulk samples were used. The initial mass of the specimens was measured to estimate the mass of water in adhesive layer after manufacturing. After that, the specimens were kept in a container filled with silica particles for 2 days at 50 °C to remove all the free water. After this drying process was completed, the weight of the sample was measured again, and the corresponding weight was considered M_0 . Finally the bulk specimens were placed in the same conditions as the ageing procedure, and its mass was measured regularly. The sample dimensions were 60x60x1 [mm³].

A.2.6 Testing conditions

Bulk tensile test, thick adherend shear test (TAST), and Arcan tests (different mode mixities), were all performed under quasi static loading conditions with a

displacement rate of 1 mm/min. For the fracture tests, the displacement rate was set to 0.2 mm/min. These specimens were tested in both aged and unaged conditions. The different mode mixities considered in Arcan joints include mode I, mode II and mixed mode (45°). For the fatigue tests, loading frequency and R ratio were set to 10 Hz and 0.1, respectively. All the tests were performed at room temperature using a servo-hydraulic machine (INSTRON[®] 8801). Tables 2, 3, and 4 summarize the test conditions.

Table 2: Strength test conditions

Test	Ageing Conditios
Tensile bulk test	Unaged
	15 days ageing
TAST	Unaged
	15 days ageing

Table 3: Strength test conditions

Test	Ageing Conditios
DCB	Unaged
	15 days ageing
ENF	Unaged
	15 days ageing

Table 4: Static and fatigue Arcan test conditions

Static		Fatigue	
Mode mixity	Ageing conditions	Mode mixity	Ageing conditions
Mode I	Unaged	Mode I	15 days ageing
	15 days ageing		
Mode II	Unaged	Mode II	15 days ageing
	15 days ageing		
Mixed Mode (45°)	Unaged	Mixed Mode (45°)	15 days ageing
	15 days ageing		

For the TAST tests, the commercial software “GOM Correlate”, digital image correlation (DIC) technique was employed to measure the strain.

As shown in Figure 7, load transfers through several components to finally reaching the adhesive layer. Since each component has its own stiffness and elongation,

the displacement measured by the machine is not the same as what the adhesive layer experiences. Accordingly, strain values are not reported for the Arcan tests.

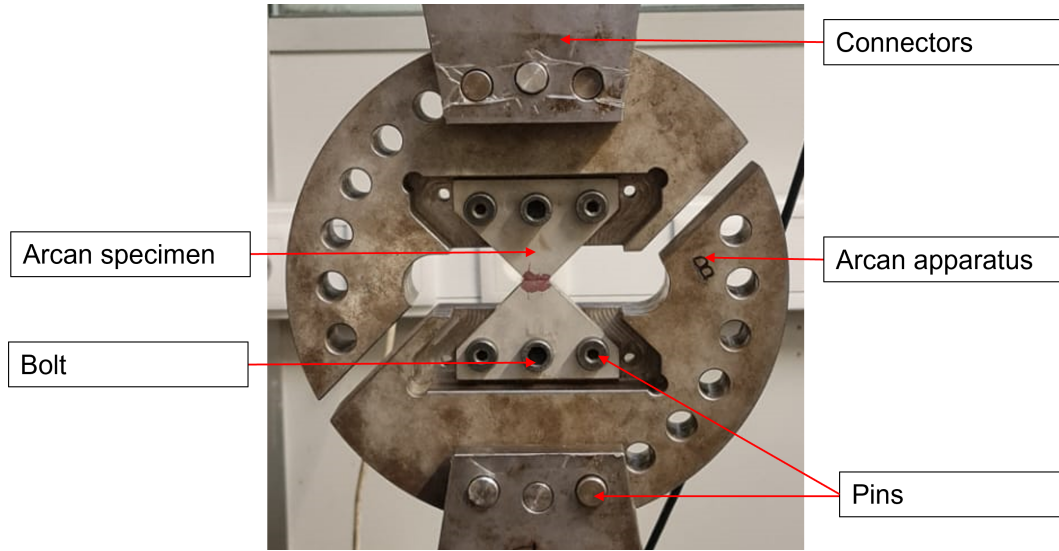


Figure 7: Arcan apparatus in pure mode I loading conditions.

A.3 Numerical model

Numerical analysis using FEM (finite element method) was conducted to investigate the distribution of water through the adhesive layer in bonded joints, as a function of the ageing time. Water diffusion into the adhesive layer is calculated using an analogy between mass and heat transfer, where the water uptake level in each point is related to the nodal temperature, and the diffusion coefficient, D , that is obtained by Fick's Law [32] using the experimental results. To implement this calculation, a two-step analysis was defined in Abaqus. In the first step the water distribution is analyzed and the second step corresponds to the stress distribution through the aged adhesive layer due to the applied external load. C3D8R, a reduced integration 8-node linear brick element was used for the substrates. For the adhesive, and since a multi-field analysis was needed (temperature + displacement), C3D8T elements were used. This an 8-node thermally coupled brick, trilinear displacement element. Since the presence of water degrades the properties of the adhesive, the material property should be defined as a function of the ageing level at each material point. Accordingly, and based on the results from the strength tests for unaged and aged conditions, the Young's modulus and Poisson's ratio were determined for both conditions in tabular format using a linear regression. The model used was simplified, to enable a finer mesh, by including only $\frac{1}{2}$ of the joint. This was done by defining a symmetry condition along a plane perpendicular to the joint's face. Figure 8 shows the Arcan model used in the numerical analysis.

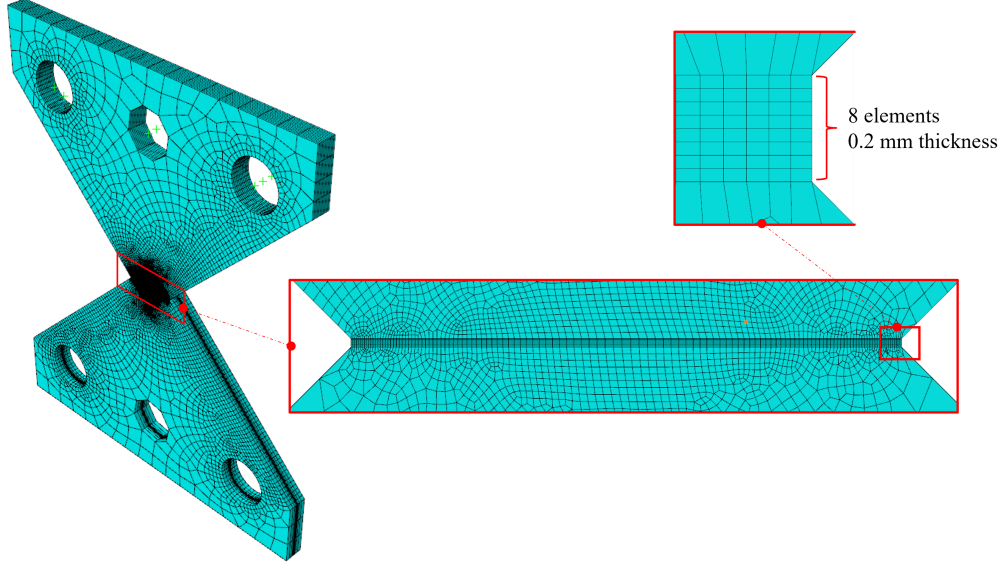


Figure 8: 3D Arcan model - Abaqus.

A.4 Results and Discussion

A.4.1 Water uptake diffusion

Throughout the hygrothermal ageing process, the new mass measured was divided by the initial mass, calculating the increment in weight percentage by the following relation:

$$wt_t[\%] = \frac{M_t - M_0}{M_0} \quad (1)$$

where wt is weight percentage of water. M is the mass of the specimen and t the time.

The water diffusion test results, presented in Figure 9, show a 2-stage water uptake: a 1st stage after approximately 12 days after the ageing procedure start, at 3.15%; a second stage was initiated with 8.5% of water after 115 days. Although not common, similar behaviour has already been reported in other studies [33, 34], where the authors found that other formulations, such as Langmuir model, could better fit this behaviour; the authors believe the reason for this behaviour is mostly related with relaxation phenomena in the polymeric chain of the adhesives [35, 36].

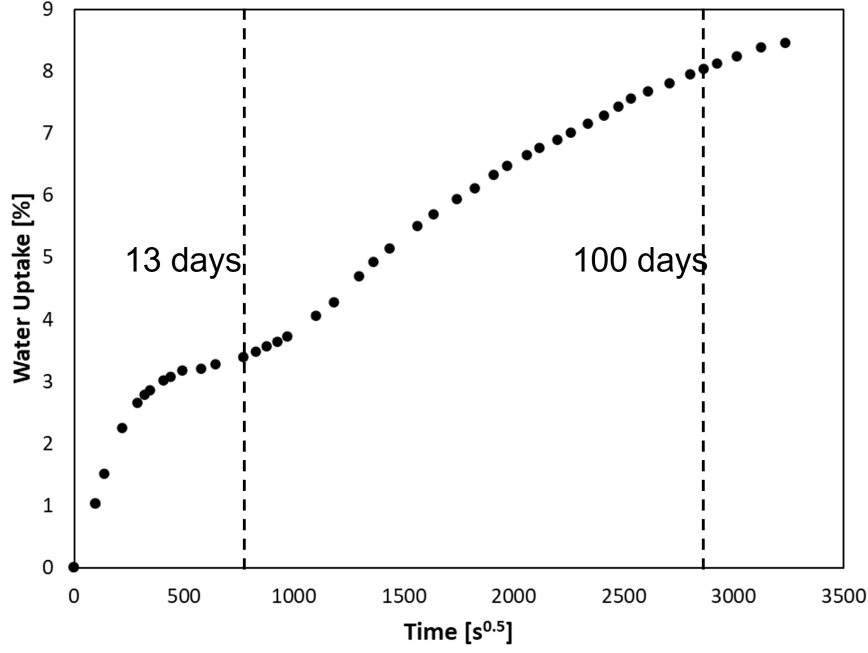


Figure 9: Moisture uptake curve for H_2O in the adhesive.

The water uptake behaviour for the 1st stage results was reproduced by the Fick's law relation [32] (see Equation 2) using an iterative process with Matlab[®]. This equation defines a substance absorption by the material using two parameters: M_∞ , corresponding to the *plateau* representing the first saturating point, and the diffusion coefficient, D , indicating how fast the water is absorbed by the adhesive. Figure 10 shows the comparison between the Fick's law curve and the experimental data. Based on these results, a good agreement between the two curves was obtained. The diffusion coefficient, D , obtained here will be used in the numerical simulation to analyze the distribution of water along the adhesive layer in tested joints.

$$\frac{M_t}{M_\infty} = 1 - \sum_{n=0}^{\infty} \frac{8}{(2n+1)^2 \pi^2} e^{\left[\frac{-D(2n+1)^2 \pi^2 t}{4l^2} \right]} \quad (2)$$

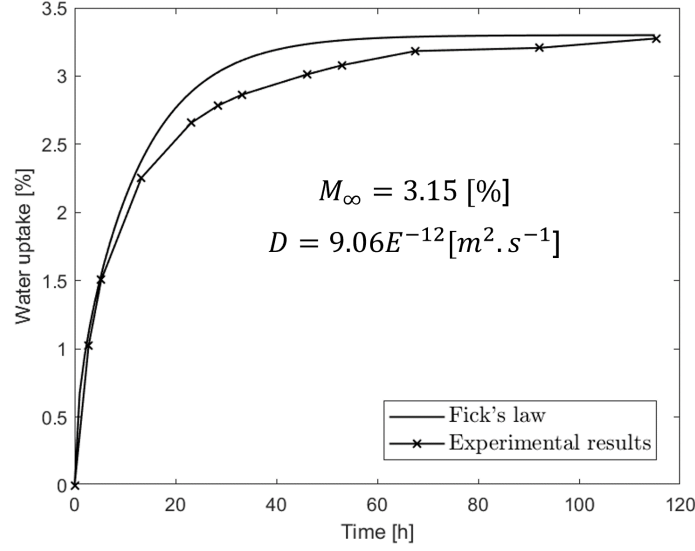


Figure 10: Fick's law fit to experimental results.

A.4.2 Strength results

Stress in the tensile “dogbone” specimens was simply obtained by dividing the load by the initial cross section of the specimen at the gauge length area. The strain was directly obtained from the extensometer data. The results from the stress-strain curves for both unaged and 15 days ageing are plotted in Figure 11. By analysing the results, the degradation of the adhesive due to moisture is noticeable where a significant reduction in tensile strength (30%) and in Young's modulus (22%) was observed after ageing; on the other hand, the strain at failure increased 21%. Figure 12 shows the values for maximum stress, strain at failure and Young's modulus for both aged and unaged testing conditions. By looking at the specimen's surface after failure, no signs of voids or defects were found.

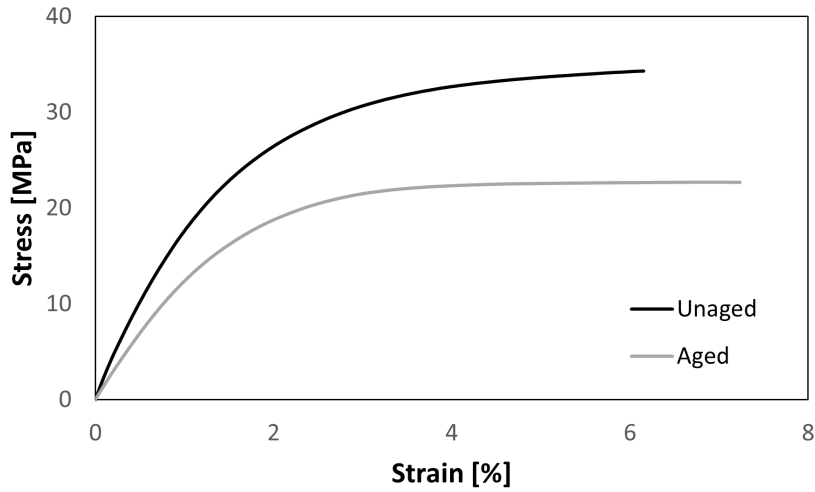
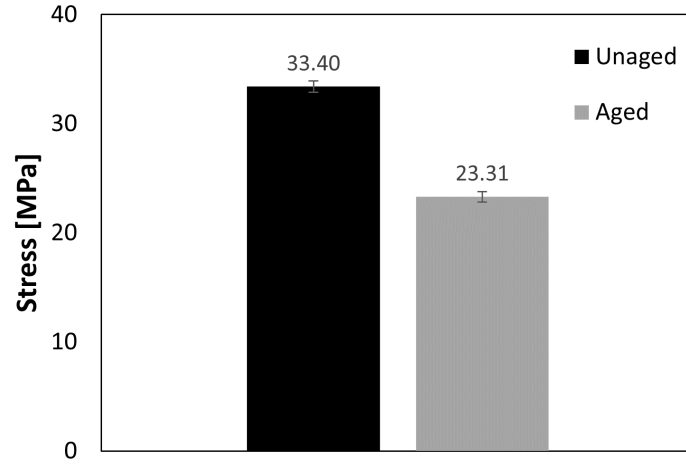
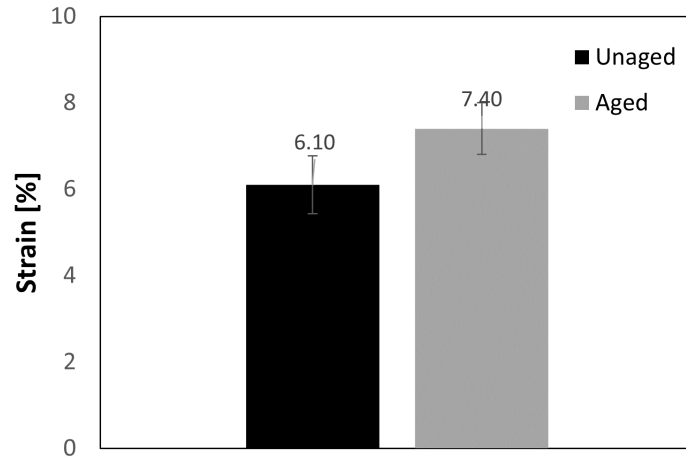


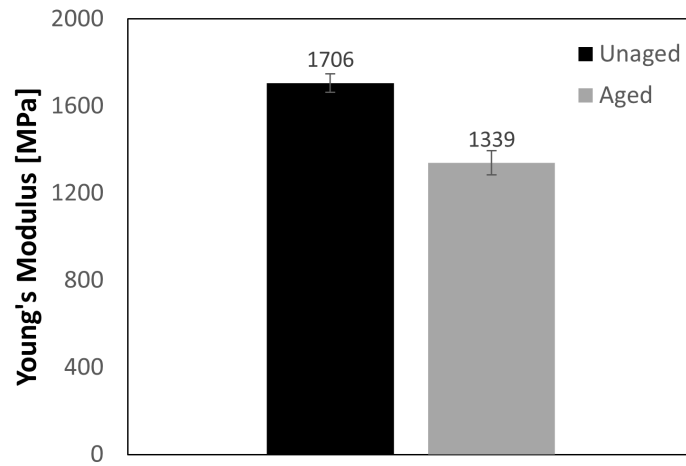
Figure 11: Stress-Strain curves for bulk tensile test for unaged and 15 days ageing.



(a) Yield Strength.



(b) Strain at failure.



(c) Young's modulus, E .

Figure 12: Properties from bulk tensile tests for unaged and aged conditions.

Using TAST specimens, the adhesive's shear strength and modulus can be obtained to subsequently calculate the Poisson's ratio. The average shear stress was calculated by dividing the applied load by the bonded area. Figure 13 shows a

typical shear stress-strain curve for unaged and aged conditions. By analysing the results, a degradation of the adhesive property with ageing is noticeable, whereas both the strength and shear strain at failure experience a significant reduction. The stiffness of the adhesive under pure shear loads, measured by the shear modulus, G , decreased by 26% with the ageing mechanism. Figure 12 shows the maximum shear stress, shear strain at failure, and G for both conditions.

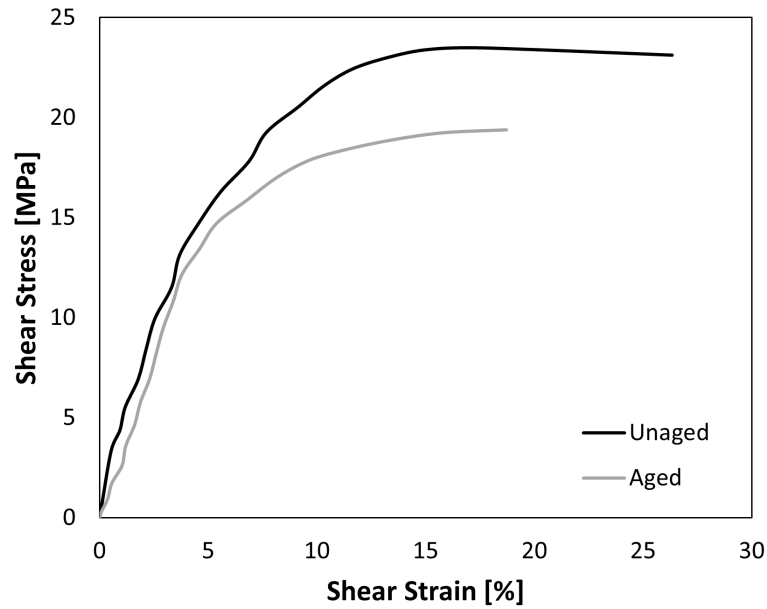
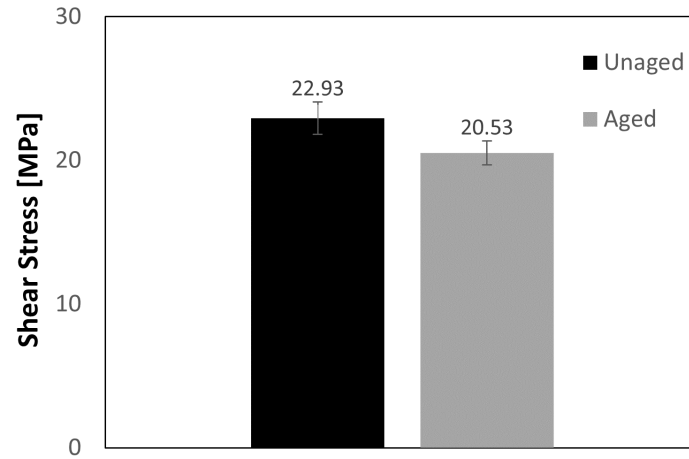
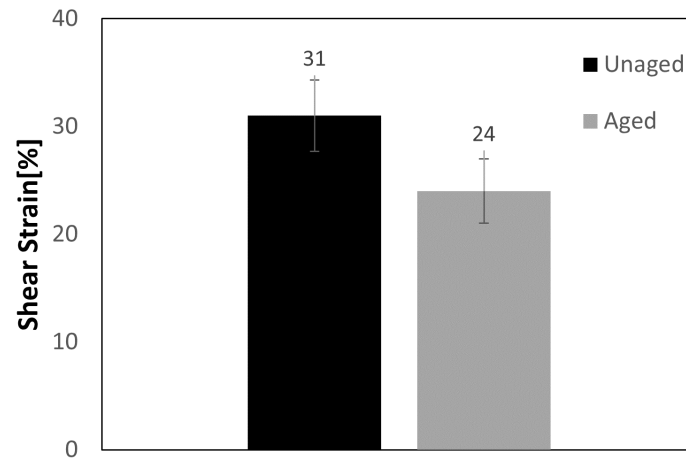


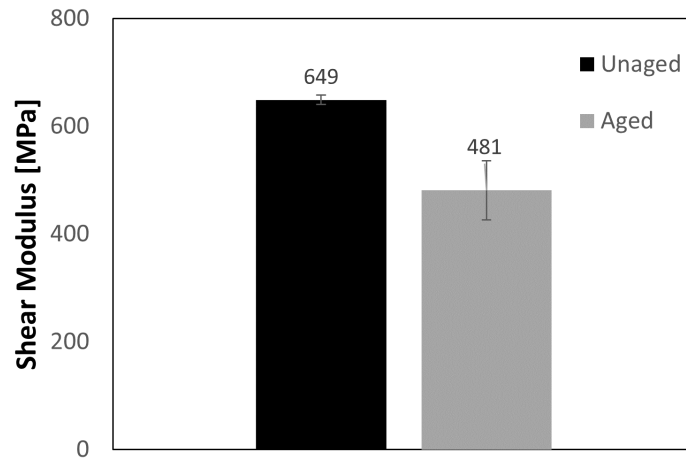
Figure 13: Stress-Strain curves for TAST for unaged and 15 days ageing.



(a) Shear Strength.



(b) Shear strain at failure.



(c) Shear modulus, G .

Figure 14: Properties from TAST for unaged and aged conditions.

By comparing the tensile and shear tests results, it becomes possible to conclude that, for this adhesive, the strength degradation due to humidity tends to be higher for tensile loading conditions, with around 30% of reduction, compared with only

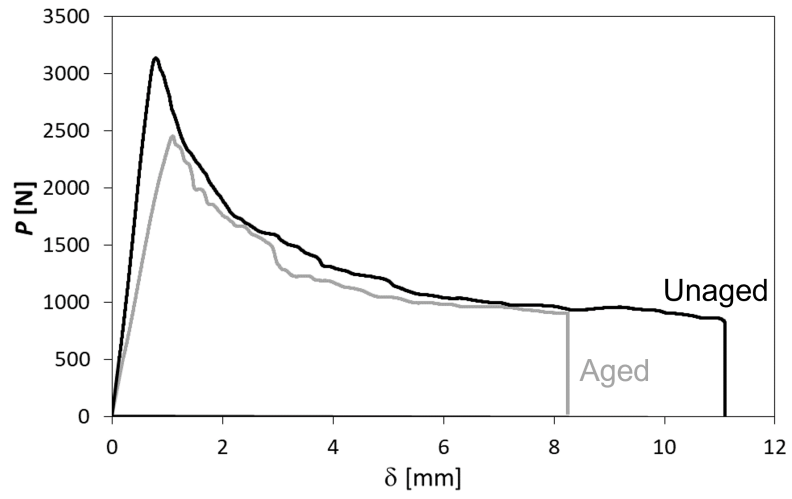
11% of reduction for the shear mode. However, it should be noted that the tensile and shear properties cannot be directly compared since they do not experience the same ageing mechanism. In shear tests using TAST joints, not only the adhesive is exposed to water but the same is true for the interface between the adhesive and the substrate. Thus, ageing will occur both through the adhesive and the interface. On the other hand, the possible presence of swelling stresses can influence the results in TAST specimens, as this source of stress is less important in bulk dogbone specimens than in the joint configuration found in TAST. Based on these results, it is also interesting to analyse the influence of the ageing process on the Poisson's ratio of the adhesive. With the Equation 3, and with the values obtained for Young's (E) and shear (G) moduli, it's possible to calculate.

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

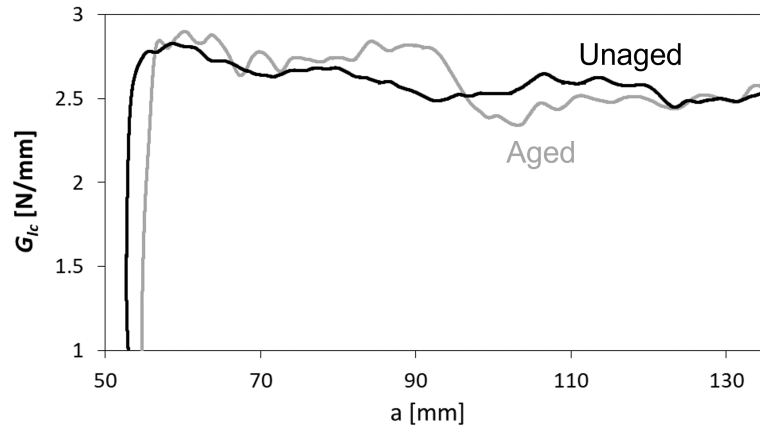
For unaged conditions, $\nu = 0.31$ and for aged conditions it increased to $\nu = 0.39$. Although these values are average since the water levels are different in each region of the adhesive layer, there is still an increase of 25% with the ageing process, meaning that for the same amount of axial strain applied to the adhesive, the aged sample will have more capacity to deform in the transverse direction. This increase can be prejudicial in real life applications in high stress environments where the material integrity is essential.

A.4.3 Fracture test results

Fracture toughness is generally considered as the most suitable property for measuring the resistance to crack propagation of a polymer adhesive system [37]. When subjected to low cycle fatigue conditions, the crack propagation region is dominant; as a result, high values of fracture toughness will result in longer fatigue life on higher load levels. Based on the strength test results, a lower reduction in shear properties was found compared to the tensile loading conditions. However, as discussed before, the specimens (dogbone and TAST) doesn't experience the same ageing level/mechanisms. In the current section the same study was conducted on fracture specimens (DCB and ENF). Both samples are the same in terms of geometry and dimensions and also in its of ageing conditions. The influence of ageing on the fracture behaviour of adhesives was studied by performing mode I and mode II tests. The results, presented in Figures 15 and 16, showed the effect of moisture depends on the loading conditions: for mode I, a 9.9% decrease was calculated for G_{Ic} from 2.62 to 2.36 [N/mm]. On the other hand, G_{IIc} improved after ageing, increasing 10.1% from 10.9 to 12.0 [N/mm].

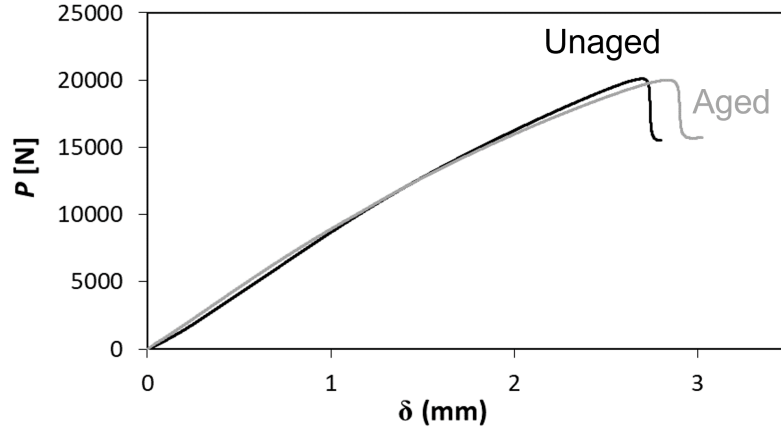


(a) P - δ curve.

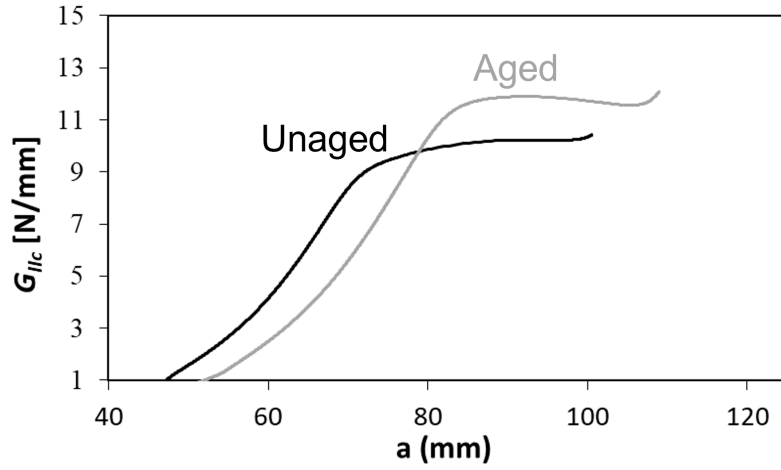


(b) R-curve.

Figure 15: DCB tests results for unaged and 15 days aged joints.



(a) P- δ curve.



(b) R-curve.

Figure 16: ENF tests results for unaged and 15 days aged joints.

A.4.4 Arcan results

Water content in adhesive layer

To investigate the water content in the adhesive layer of the Arcan joints after ageing, a numerical analysis was carried out. A relationship between heat transfer and fluid flow was explored in this simulation, where the nodal temperature of the elements are interpreted as the water uptake level in the adhesive.

The objective of this analysis is to understand the different humidity levels between the centre of the adhesive layer and the boundary. This way we can understand the variation of mechanical properties through the adhesive layer during a test. The results from the 1st stage of the experimental curve (see 9) were fitted by the Fick's law, as explained in Chapter A.4.1.

The results from this analysis (see Figure 17) showed that, for the boundary of the adhesive, the water level (wt%) is 42.2% higher than for the centre point, which

means the property degradation is considerably more severe at the edges than in the centre. Figure 18 shows the water uptake in the centre of the adhesive layer throughout the 15 days of ageing; the results show that it takes around 2 days (48 hours) for water to reach the centre of the adhesive layer.

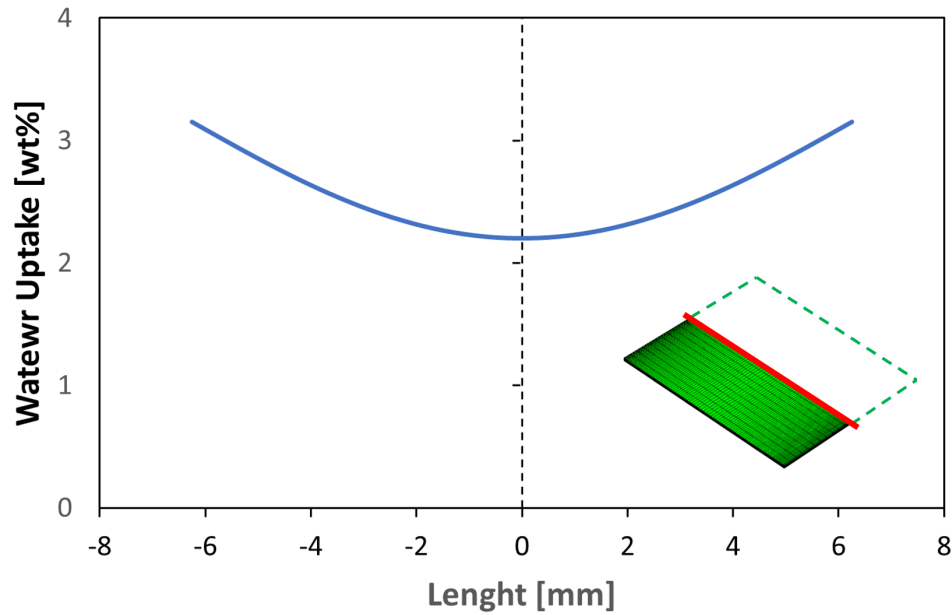


Figure 17: Numerical simulation of the water level [wt%] along the bondline after 15 days.

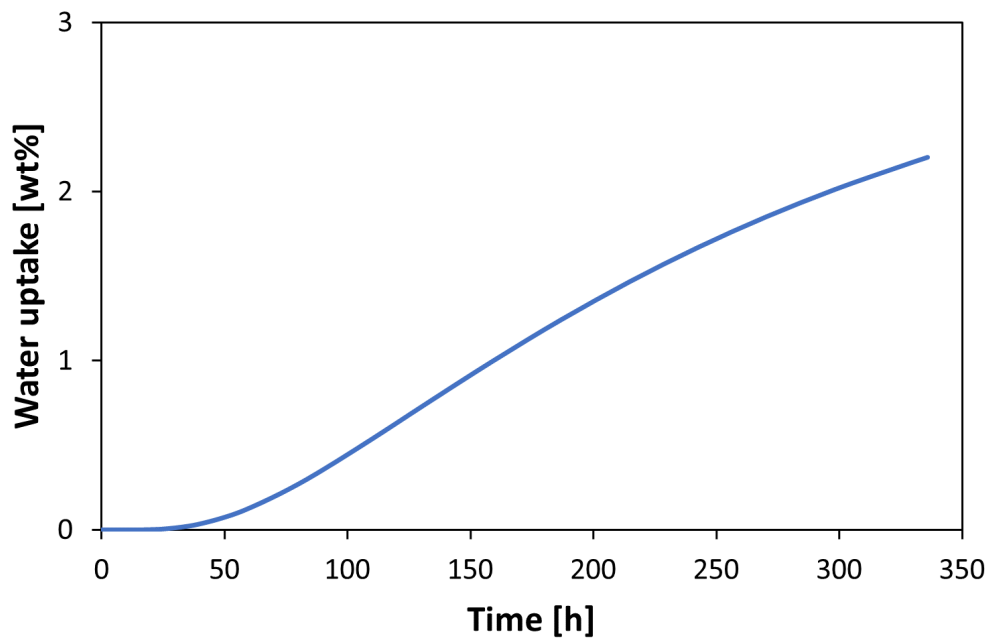


Figure 18: Numerical simulation of the water level [wt.%] in the center of the adhesive along the 15 days.

Static tests

Arcan static tests were performed in mode I, mode II, and mixed mode (45°) to evaluate if the rate of degradation due to ageing is similar if the same joint with the same water content is tested at different loading angles. The results were also useful to define the fatigue load levels. To understand the effect of the static tests, experiments were performed for both test conditions. Figures 19 and 20 show typical static load-displacement curves for different mode mixities and for the two conditions. Based on the results, although the considered mode mixity (45°) is mathematically at the middle of mode I and mode II, the experimental results showed that mixed mode strength is in fact closer to mode II than mode I. A possible cause for this are the failure mechanisms, possibly governed by the shear phenomena. By compiling all results from the static tests, a failure envelope was built for the two conditions (Figure 21); the effect of the ageing mechanism was significantly higher for mode I with a reduction on the maximum load on the aged samples of 43.7%, while for mode II this difference was 10.0% for mode II.

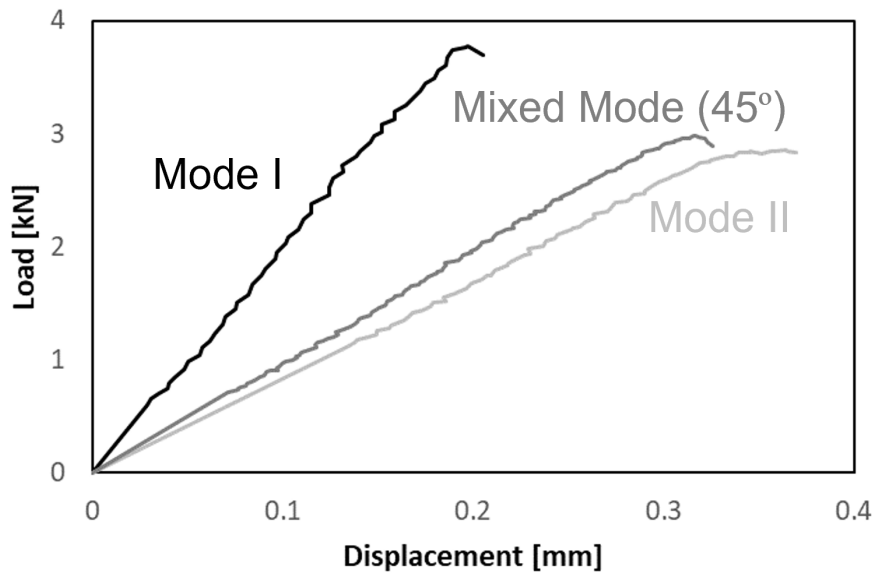


Figure 19: Load-displacement curves for Arcan tested under unaged conditions.

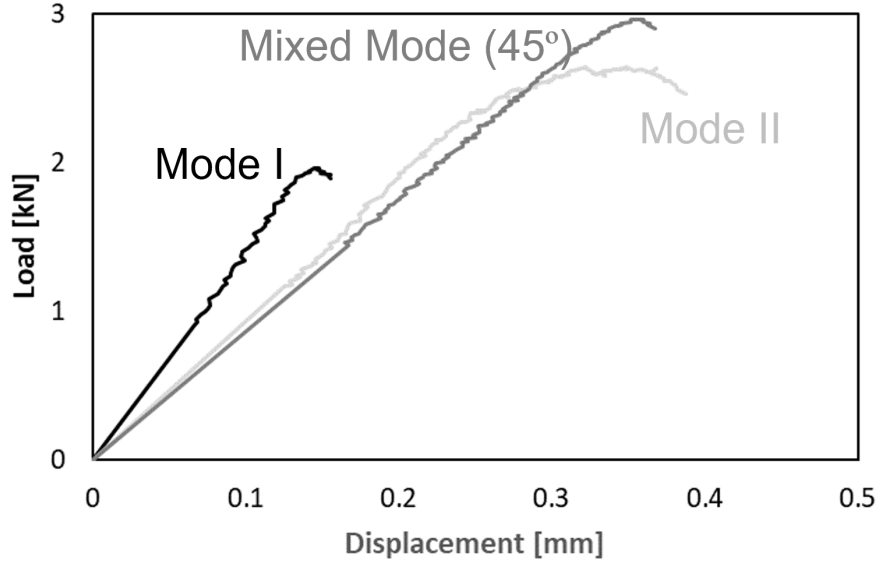


Figure 20: Load-displacement curves for Arcan tested under 15 days ageing.

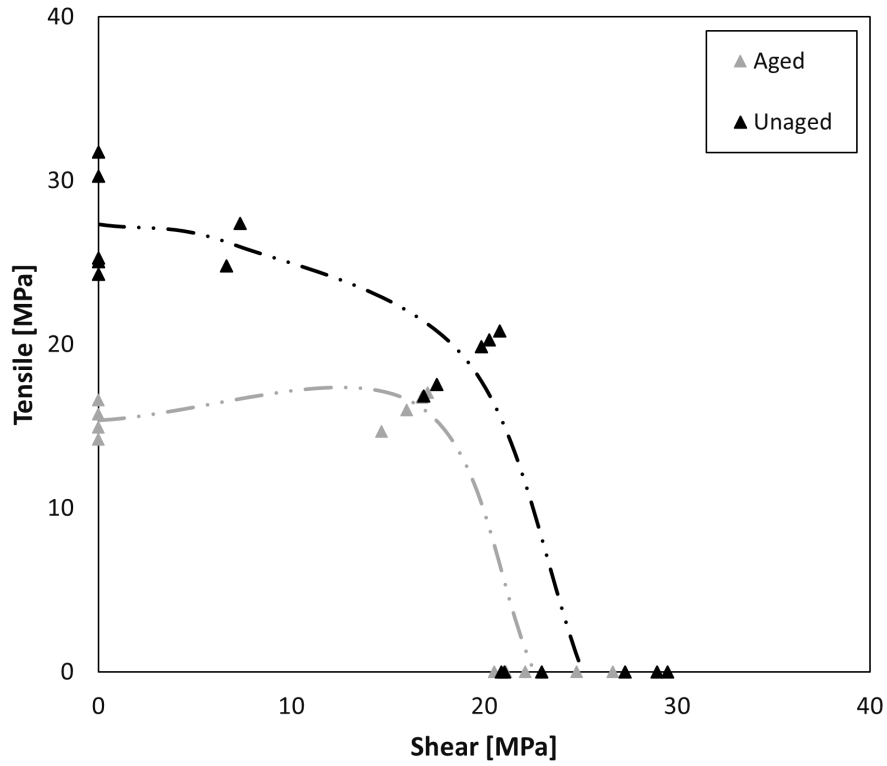


Figure 21: Failure envelope for Arcan static testing, unaged and 15 days ageing.

Fatigue tests

Fatigue tests were performed at different load levels and mode mixities. With the results from those tests, stress-life (S-N) curves were constructed – Figure 22.

As static tests showed, the fatigue life for mode I also had a larger decrease than for mode II and mixed mode loading conditions, for the same load level. The slope of the curves is also an important parameter, as a indicator of the sensitivity of the fatigue life of the joint to the variation of the applied fatigue load. Based on the results, in pure mode I, the curve is flatter, indicating that a small increment on the load level can drastically change the expected life of the joint. The lowered sensitivity of pure mode II and mixed mode to fatigue load conditions can be an important advantage on the life prediction of the joint, since any small changes in the applied load will not change the fatigue results significantly.

Fatigue results of aged samples showed that, for the same load ratio (fatigue load/static strength), the fatigue life is much higher when the specimens are loaded in mode II compared to mode I loading conditions.

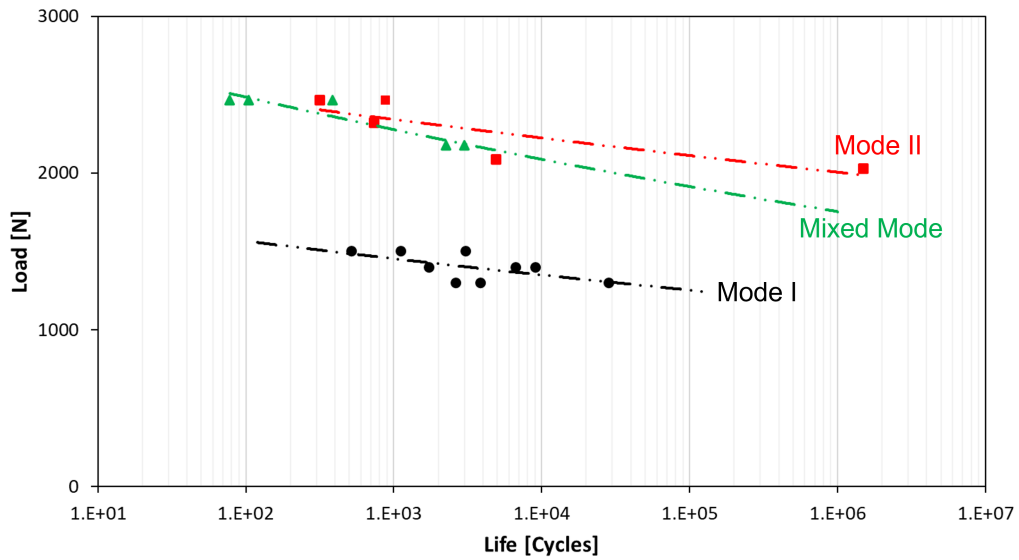
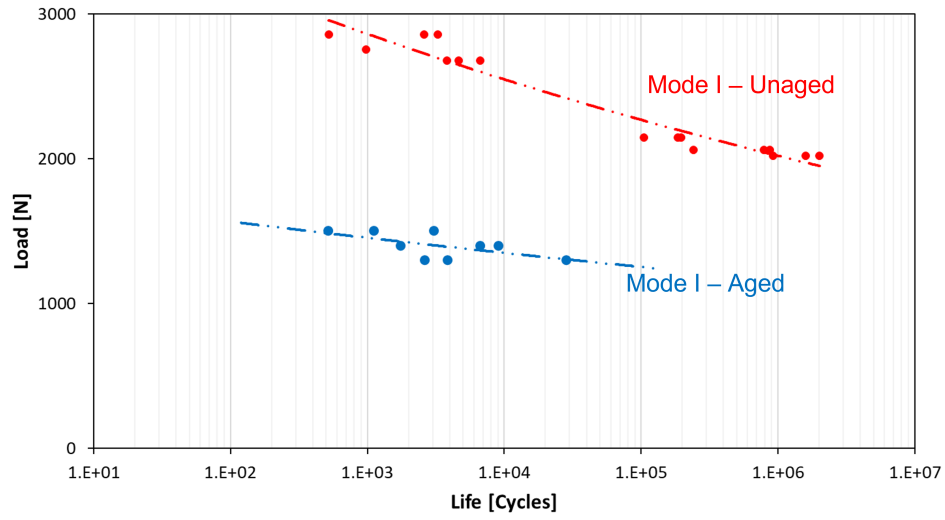
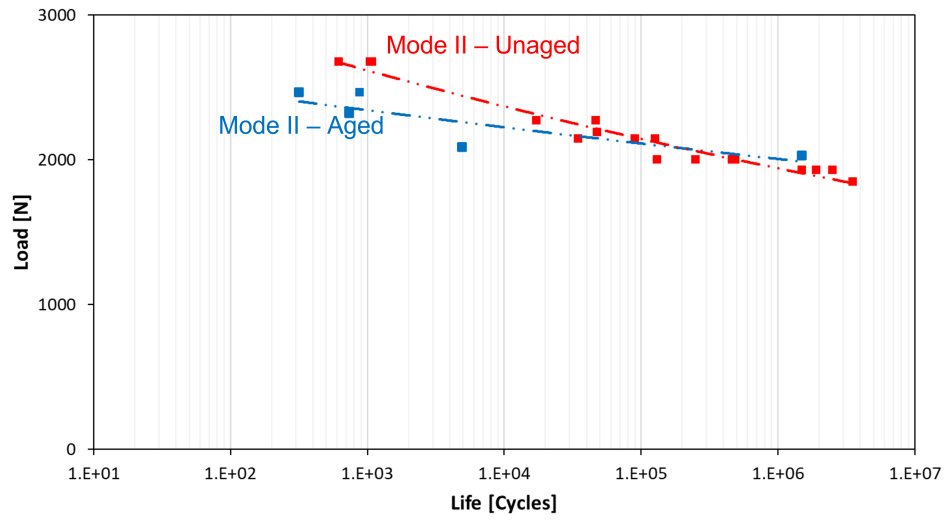


Figure 22: Load-displacement curves for Arcan specimens tested after 15 days ageing.

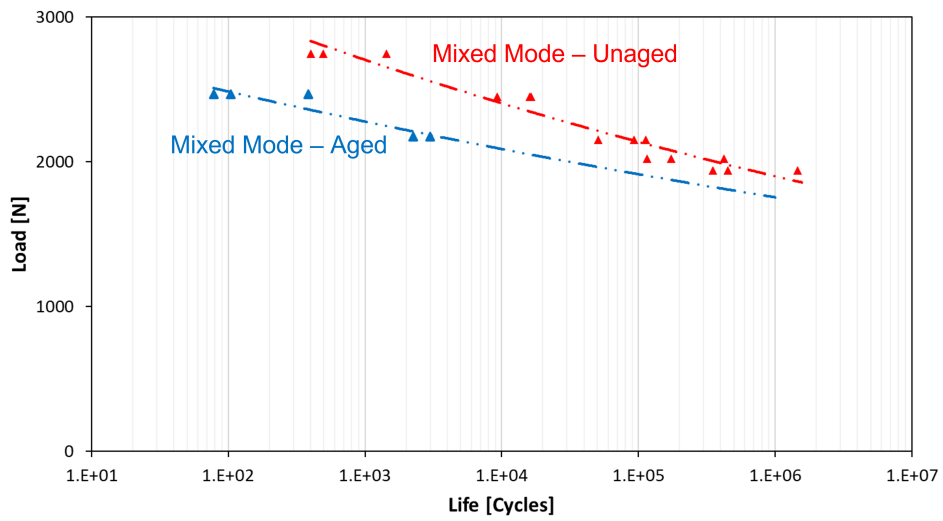
The results from the fatigue tests of aged Arcan specimens were compared to results of another study [38], where the same adhesive was tested using the same Arcan joints but for unaged conditions. However, a different curing cycle was considered, thus one must be cautious when directly comparing this data against the fatigue results. Figure 23 shows the fatigue results, being evident that the ageing mechanism degrades significantly the S-N behaviour of the bonded joints. For 15 days of ageing, the rate of change in life with fatigue load is significantly lower, indicating that the ageing mechanisms not only weakens the joint, but also increases its sensitivity to cyclic loading conditions; this effect is more severe for mode I than mode II.



(a) Mode I.



(b) Mode II.



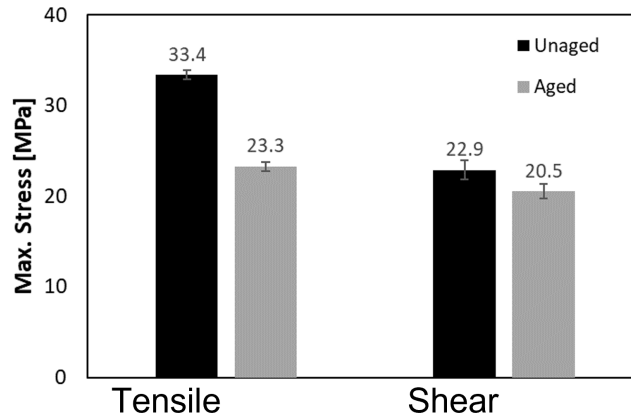
(c) Mixed mode (45°)

Figure 23: S-N curves for unaged [38] Vs. 15 days ageing °C.

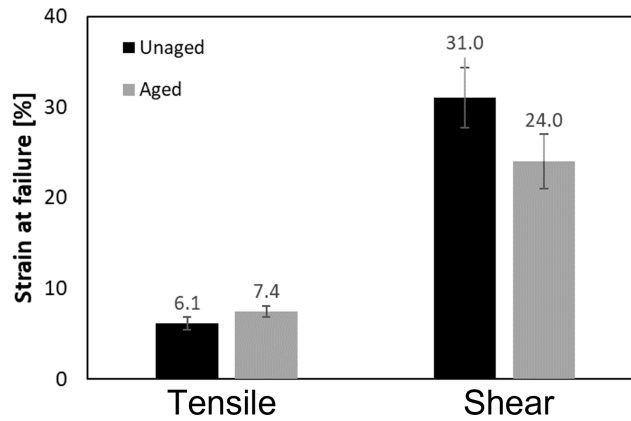
A.4.5 Effect of ageing on the rate of property degradation

According to the results presented in the previous sections, it was found that the rate of strength degradation due to ageing is a function of loading angle. Different tests were performed in this regard, including strength tests, fracture tests, Arcan static tests and also the Arcan fatigue tests. Both aged and unaged conditions were considered and for all testing techniques different loading angle/mode were experimentally analyzed. It was shown that the way that the ageing mechanism affect each property is different and should be analyzed individually.

The strength and Arcan tests (Figure 24 and 25) showed similar results for mode II, with about 10% of property degradation; for pure mode I there is a small difference in the results between the strength tests and the Arcan tests, which can be explained by the fact that strength test for mode I is a bulk test, which does not involve adherends and so the property degradation only happens in the adhesive layer while for Arcan there is also degradation in the interface.



(a) Yield Strength.



(b) Strain at failure.

Figure 24: Results and properties from strength test under unaged and aged conditions.

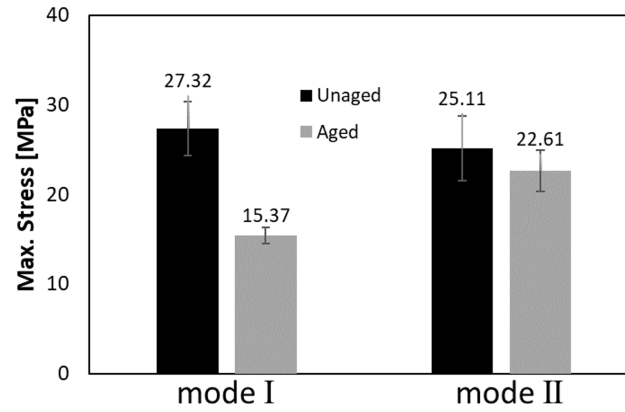
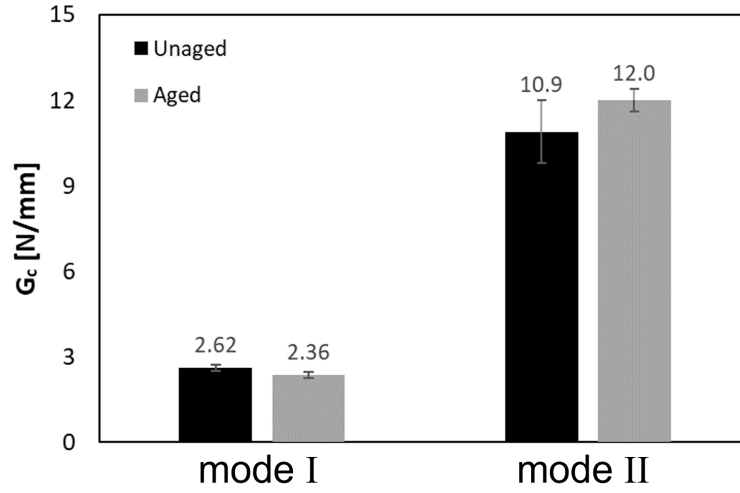
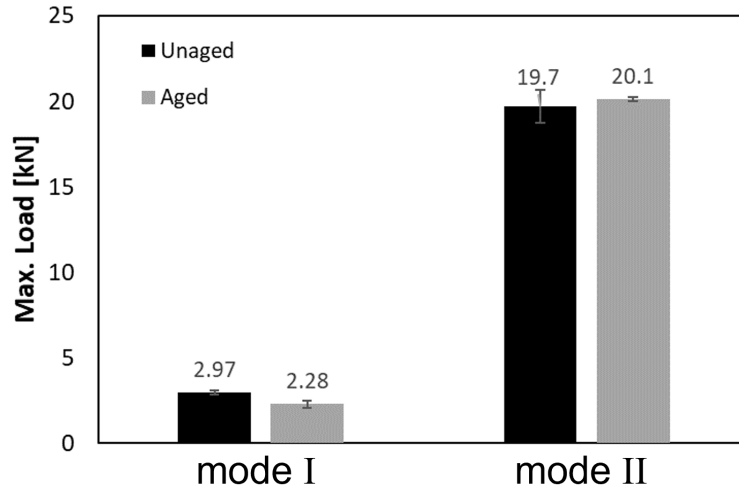


Figure 25: Effect of mode mixity and ageing on Arcan static strength.

Fracture tests showed a significant difference between mode I and mode II: for mode I the fracture toughness decreased 10% with ageing. On the other hand, mode II experienced a property improvement after ageing from 10.9 to 12.0 [N/mm]. The maximum load decreased in both mode mixities, being more severe in mode I; a possible cause for this, in addition to the moisture, could be the small differences in the initial crack length of the specimens, a_0 .



(a) Fracture toughness.



(b) Maximum load.

Figure 26: Results and properties from fracture tests under unaged and aged conditions.

Figure 27 shows the effect of moisture on the calculated properties of the different tests performed, by dividing the respective property at unaged conditions by its value after ageing. Values more close to zero indicate significant property degradation, while values higher than one mean that the adhesive's properties were improved by the effect of humidity. The results showed mode mixity is a crucial parameter when analysing the effect of ageing on adhesive; the higher sensitivity of pure mode I to moisture was verified for all test types. This conclusion is important in joint design, where pure mode conditions effects should be evaluated separately to optimize the joint's performance. The fracture tests were found to be less affected by the ageing mechanism, since its values are closer to 1. In fact, for the ENF test, the fracture toughness on mode II even increased when compared with the unaged conditions; a possible cause for this could be the increase of ductility of the adhesive due to

introduction of water in its polymer chain.

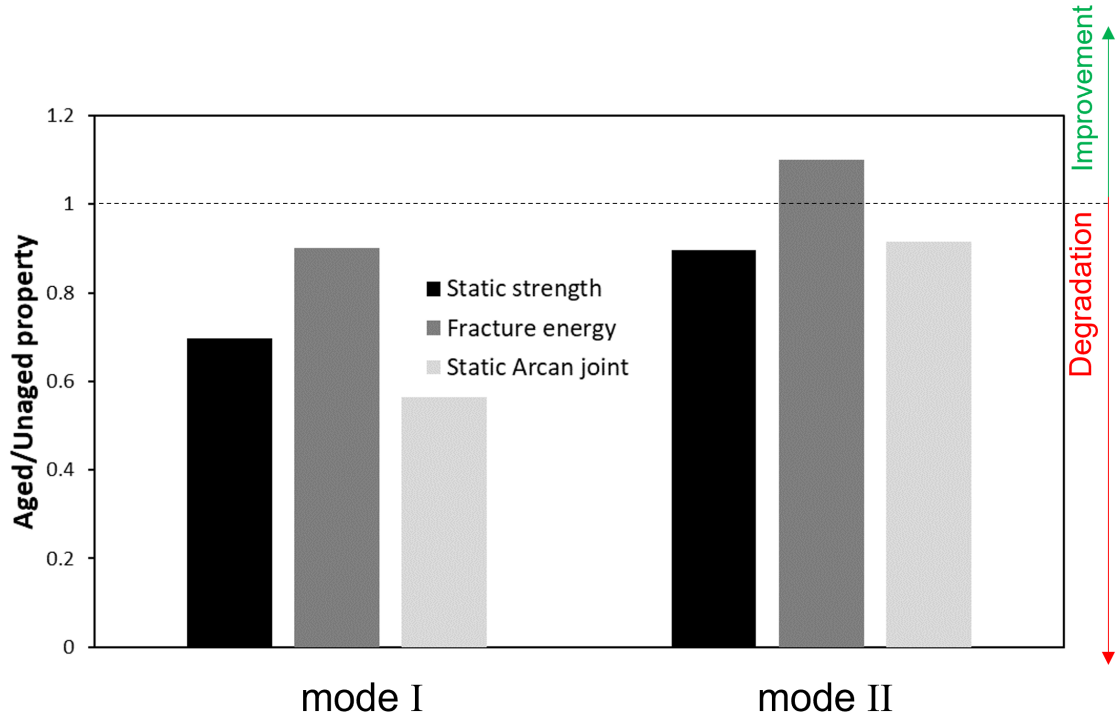


Figure 27: Influence of ageing on adhesive properties for different tests.

A.4.6 Fracture surface analysis of Arcan joints

This component of the study seeks to evaluate the effect of ageing on adhesive joints for different loading mode/type. The results showed that for mode II and mixed mode, where shear load phenomena are present, the effect of ageing appears to have been mitigated, while for pure mode I, the effect of ageing is noticeably more severe, with the joint performance decreasing drastically. To investigate possible causes for this, the joints' fracture surface were analysed.

Static fracture surfaces

The fracture surfaces of unaged Arcan joints showed a cohesive failure for all mode mixities (see Figure 28). However, in mode I the surface is smoother while for mode II and mixed mode, the surface is rougher due to the shear failure mechanisms. A lateral image from the joint, observable in Figure 29, shows the presence of shear hackles, which corroborates this theory. Shear hackles were present also in mixed mode (45°), indicating its presence does not require pure shear conditions, but only a partial mode II component. For aged samples, the different ageing levels throughout the adhesive layer, with more water in the borders is indicated by a darker surface, while in the centre the colour is more similar to the one found on

the unaged joints. The failure mechanism was not uniform and is similar for all mode mixities. All tested conditions showed both adhesive and cohesive failure in aged samples. However, mode I experienced less area with adhesive failure when compared with mode II and mixed mode (45°) where the adhesive failure region was predominant. Despite the theoretically higher rate of water diffusion through the interface [25], the lower rate of property degradation for the interface and also the short aging time that was not enough for interfacial saturation led to a mixed failure mode. Interfacial failure was more dominant for mode II and mixed mode loading conditions.

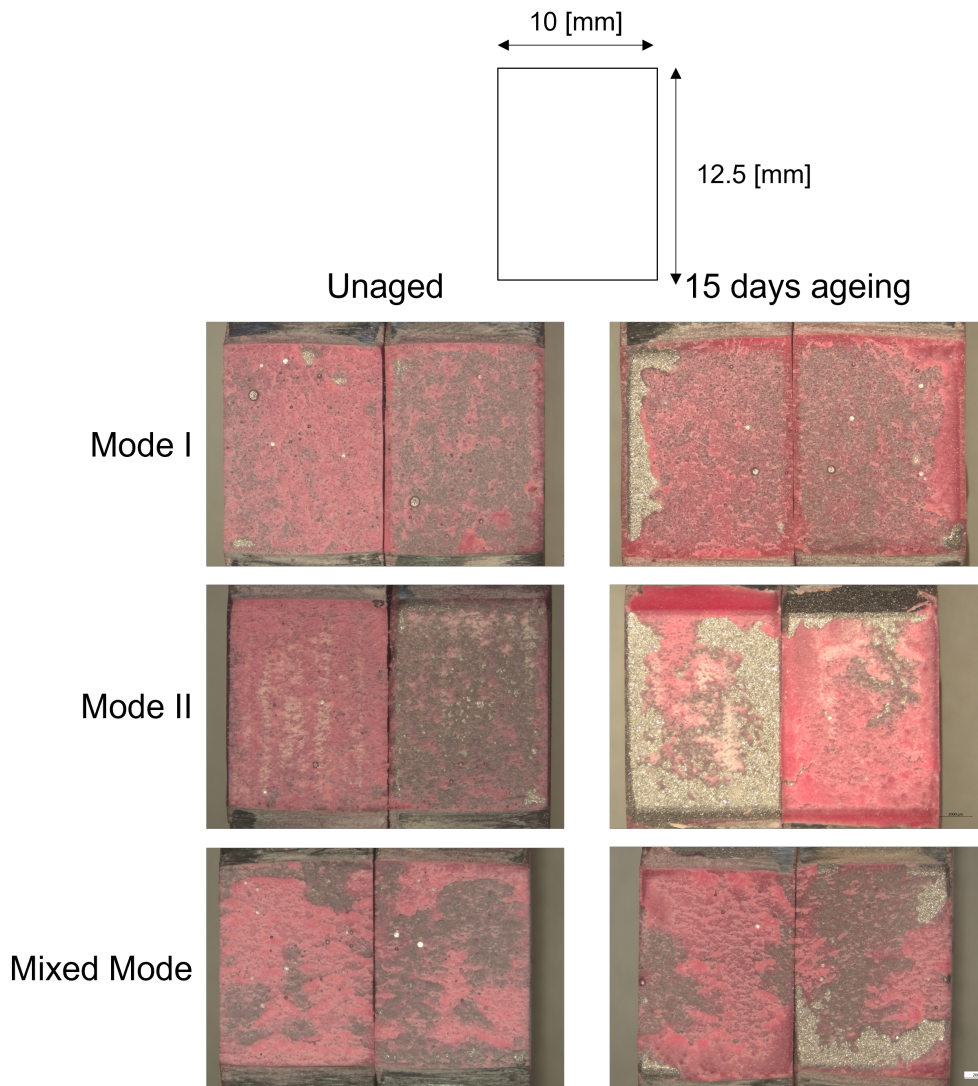


Figure 28: Failure surface of statically tested Arcan Joints under unaged and 15 days ageing for Mode I, Mode II and Mixed Mode (45°).

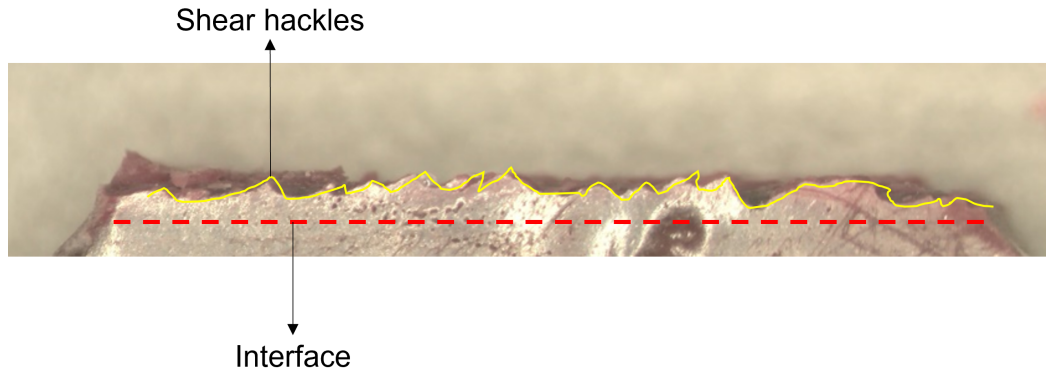


Figure 29: Shear hackles present in pure mode II Arcan joints.

Fatigue fracture surfaces

The failure surface of the joints subjected to cyclic loads showed similar results to the static ones in terms of failure mechanisms, again mode II loading with more adhesive failure mechanism than mixed mode, and also mixed mode failure mechanism showed more adhesive failure than pure mode I loading conditions as shown in Figure 30. However, for fatigue tested joints, the roughness is less significant than the respective results in static loading conditions. By analysing the crack paths for aged samples under fatigue loading conditions in mode I and mode II (see Figure 31), it can be observed that both cracks start from the interface region at the bonding edges. However, in mode I the crack shows an early deviation towards the adhesive layer, continuing to propagate through this layer until joint failure is reached, while for mode II the cracks propagate first at the interface with a late deviation towards the adhesive layer, leading to more adhesive failure in mode II.

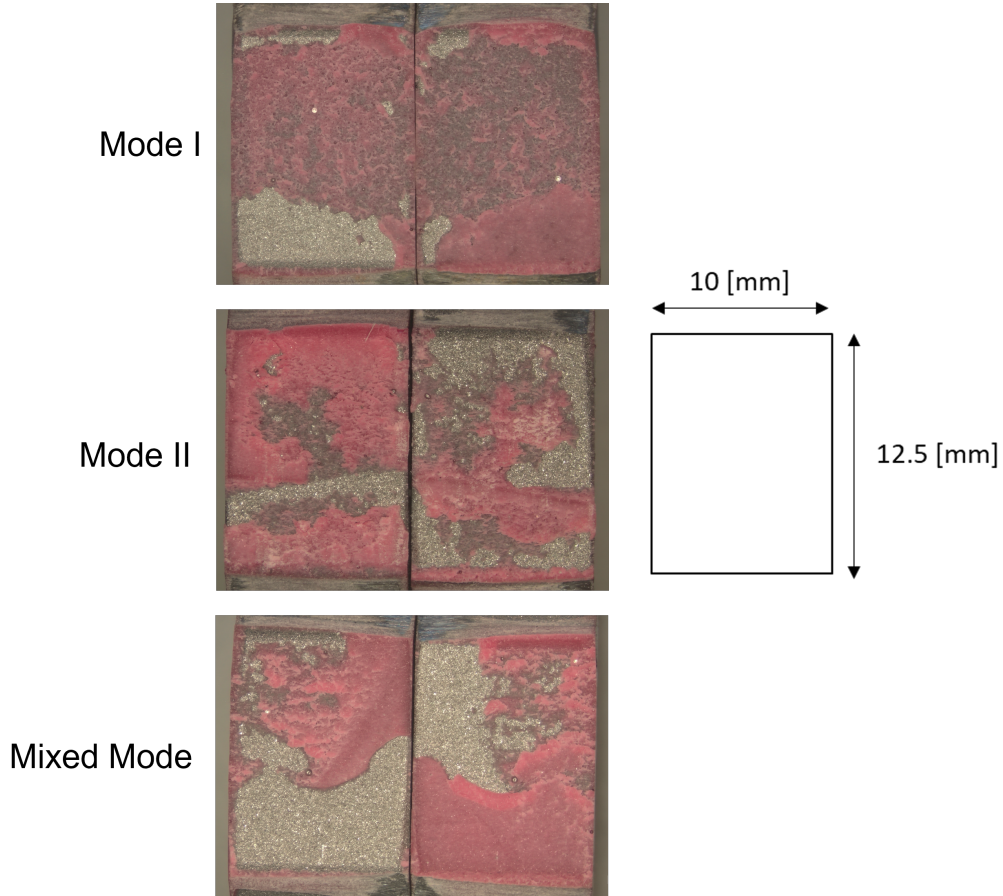


Figure 30: Failure surface of fatigue tested Arcan Joints under 15 days of ageing for Mode I, Mode II and Mixed Mode (45°).

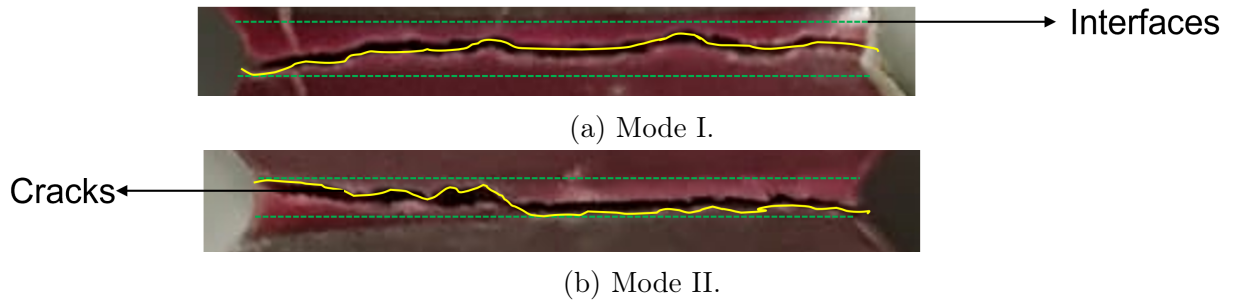


Figure 31: Crack path for aged Arcan joints under fatigue load conditions.

A.5 Conclusions

The effect of hygrothermal aging and mode mixity on the mechanical properties of an epoxy adhesive was studied. Multiple strength and fracture tests were carried out experimentally. Furthermore, Arcan joints were tested statically under pure mode I, pure mode II, and mixed mode (45°). Fatigue tests were conducted using the same Arcan joints. Fracture surface analysis of the Arcan joints helped to better

explain and understand the experimental data.

The presence of water on the adhesive was found to have degraded its mechanical properties. Results showed that 15 days of ageing in distilled water is enough time to significantly reduce the durability of the joints, especially in pure mode I conditions; in fact, it was determined that when shear forces are applied to the adhesive, its sensibility to humidity is much lower for both static and fatigue conditions. This trend was verified for all types of tests carried out during this work, including fracture tests, standard strength tests, Arcan static tests and Arcan fatigue tests.

Property degradation due to moisture was more severe for the Arcan tests and the strength tests, compared with fracture tests results. In fact, fracture toughness under pure mode II conditions increased after the 15 days ageing. It has also been found that for the same loading mode (mode I or mode II) applied to the Arcan joints, aging has a greater impact on fatigue strength than static strength.

The ageing phenomena were also found to affect the fracture surface, resulting in the appearance of some regions with adhesive failure. Results also showed that the load angles influences the failure mechanism: The fracture surface of Arcan joints showed more adhesive failure on mode II and mixed mode (45°) when compared with mode I, where cohesive failure is predominant.

These findings can be used to support the design of joints intended to operate in extreme environment applications, where the proper assessment of the adhesive's properties in pure mode conditions is fundamental to achieve a safe and durable design.

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B Paper B

The interaction of loading mode and temperature on the property degradation of an epoxy adhesive subjected to strength, fracture, and fatigue tests

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ABSTRACT

The type of loading and high temperatures have a significant impact on the mechanical behavior of adhesive joints. In real applications, joints are often under mixed stress states and usually the service temperature is different from room temperature. For these reasons, understanding how the mechanical properties of adhesives are affected by these factors is crucial in joint design. Although multiple studies have approached the effects of temperature on the behaviour of bonded joints, the role of loading mode and test type on the rate of degradation of adhesive/joint properties at high temperature has not been properly investigated.

Accordingly, the aim of the current study is to determine if adhesives experience the same rate of degradation at high temperature when they are subjected to different mode mixities and different types of tests. To do so, an epoxy adhesive was subjected to different loading conditions (strength tests, fracture tests, and fatigue tests, all for modes I and II) at different temperatures. The results show that there is a significant difference between the rate of degradation in mode I and mode II loading conditions and also that, for the same loading mode, the rate of degradation is mostly a function of the test type. For the fatigue experiments, the S-N response of the tested adhesive was obtained as a function of loading mode using Arcan joint testing. Based on the results, an S-N model was also developed to create a master S-N curve as a function of temperature.

B.1 Introduction

Adhesive bonding technology is assuming a more relevant role in industry in the last few decades, finding greater use in high-performance aerospace and automotive applications. Behind this growth are the very interesting advantages provided by this joining technique, especially when compared with traditional fastening methods such as welding and riveting. A more uniform stress distribution, combined with the flexibility to join many different types of materials are key advantages of adhesive bonding [1–4]. Despite its benefits, the use of adhesives has some important drawbacks, being one of the most important the high sensitivity to severe environmental conditions. Many authors have been studying these effects on the performance of adhesive joints [5–9]. The effect of the temperature on adhesive joints is a relevant research topic since service temperatures are in practice often below or above the standard room conditions.

Service temperature is a main factor affecting adhesive’s performance. One reason behind the significant dependency of adhesive properties on the temperature is the glass transition phenomenon. For temperatures above the glass transition temperature (T_g), the adhesive will experience more elongation, with lower strength and stiffness. Adams et al. [10] highlighted the effects of thermal stresses by testing single lap joints at different temperatures. Some authors [11, 12] identified a significant relationship between the T_g of epoxy adhesives and the curing temperature, with higher curing temperatures leading to higher T_g also. They also concluded that a post-cure after a low temperature cure also elevates the T_g of the adhesive. It was also determined that, for very high curing temperatures, T_g decreases; the authors believed this is due to thermal degradation of the adhesive itself. Others authors [13, 14] also studied this phenomenon, reaching the same conclusions.

The property degradation due to the exposure to different temperatures under quasi-static conditions has been also investigated by multiple authors. Na et al. [15] subjected dissimilar adhesive joints to different temperatures concluding the increase of temperature also brings a decrease into the joint’s strength, but a significant increase in its strain. Deb et al. [16] used double lap joints with a rubber-toughened epoxy-based adhesive to study the effect of different extension rates and temperatures; they concluded that for higher temperatures the adhesive becomes softer and its strength decreases; on the other hand, for temperatures below room conditions, they found an increase on the strength of the joint. However, some solutions have been proposed by authors. da Silva and Adams [17] studied the advantages of using dual adhesives along a single bondline to increase the range of working temperature of adhesive joints. The effect of temperature on pure tensile and shear properties of adhesives have been rarely considered; Banea et al. [18] reported lower effect of the temperature on shear lap strength compared with bulk tensile tests. On the other hand, da Silva et al. [19] reported similar levels of degradation for both

tensile and shear properties, when changing the temperature between -22 and 200 °C.

Fatigue loading is a common loading conditions that bonded joints may experience in service. Fatigue loading degrades the properties of an adhesive cycle-by-cycle and a combination of fatigue loading and high temperature can significantly reduce the durability of the bonded joints. When fatigue loads are combined with different temperatures, this can lead to premature and unexpected failure of the adhesive joint. Some research has already been carried out to determine on the influence of different temperatures on fatigue behavior and performance of adhesives. Some authors studied the effect of temperature on the failure mechanism on joints under fatigue conditions [20–22], concluding the failure mechanism does not significantly change as percentage of adhesive and cohesive failure. However, lower temperatures resulted in a flatter fracture surface while above a specific temperature cracks started to be evident on the surface. Schneider et al. [23] studied the fatigue damage behavior of an epoxy adhesive using scarf joints under three different temperatures including 10, 23 and 50 °C. They experienced a reduction of fatigue strength with increasing temperature for constant load conditions. Wei Tan et al. [20] studied the fatigue behavior of a polyurethane adhesive under different service temperatures, using thick adherend shear joints (TAST). They subjected the joints to five different temperatures, ranging from -40 to 80 °C. They concluded that the fatigue performance of the adhesive gradually decreases with the increase of temperature. Beber et al. [24] studied the influence of temperature on the fatigue behavior of an epoxy, using many joint geometries. From the experimental results, they concluded that fatigue lifetime decreases with temperature.

The effect of the mode mixity on the static and fatigue response of adhesive joints at room temperature has also been the target of multiple authors [25–28]. Sousa et al. [27] investigated the influence of different mode mixities on the fatigue life of an epoxy adhesive. They concluded that the fatigue life of the joint was reduced with the introduction of a shear-loading component.

Although experimental testing of common joint geometries, such as lap shear joints, provides an insight on the effects of temperature on the joint/adhesive behaviour, due to the complex mode mixity that these joints experience during the test, such results cannot be directly used for design purposes. On the other hand, pure mode conditions (mode I and mode II) are rarely considered in studies, especially for fatigue testing at different temperatures. Furthermore, the rate of degradation of the adhesive properties as a function of loading mode and test type has not yet been fully investigated and understood. Accordingly, the aim of the current study is to address this knowledge gap, doing so by investigating the combined interaction of temperature and mode mixity on the rate of property degradation of an epoxy adhesive subjected to static and fatigue loading conditions. To achieve this, stan-

standard strength tests, fracture tests, static Arcan joint tests and fatigue tests using Arcan assemblies were conducted, for both mode I and mode II and at room and high temperatures. Fatigue tests at different load levels were carried out to generate S-N curves. Finally, a fatigue parameter was developed to build master curves at different testing temperatures.

B.2 Experimental Procedures

In this study, an epoxy adhesive was tested under static and fatigue loading conditions at different mode mixities using Arcan joints. Strength and fracture tests were conducted for mode I and mode II loading conditions. All tests were performed at different temperatures.

B.2.1 Material

A two-component epoxy based structural adhesive was used to study the effect of temperature and load angle on the fatigue response of Arcan joints. The considered adhesive is suitable to join similar and dissimilar materials. Table 1 shows the mechanical properties of the adhesive. All values, apart from the density, were determined during this research work. The Arcan substrates used throughout this work were machined from an AISI 316 stainless-steel bar.

Table 1: Mechanical properties of the adhesive.

Properties	Adhesive
Maximum tensile strength, MPa	33.4 ± 0.5
Maximum tensile strain, %	6.1 ± 0.7
Maximum shear strength, MPa	22.9 ± 1.1
Maximum shear strain, %	31.0 ± 10.6
Young's modulus, E	1706 ± 42
Shear modulus, G	649 ± 9
Poisson's ratio, ν	0.32
Density, $g \cdot cm^{-3}$	1.07

B.2.2 Geometry and manufacturing procedure

Figure 1 shows the geometry of the Arcan specimens. Arcan joints were manufactured using a specific mould (see Figure 2) and the thickness of the adhesive layer was set at 0.2 mm using spacers. To ensure a good level of adhesion, all surfaces were sandblasted with alumina particles and then cleaned using acetone [29].

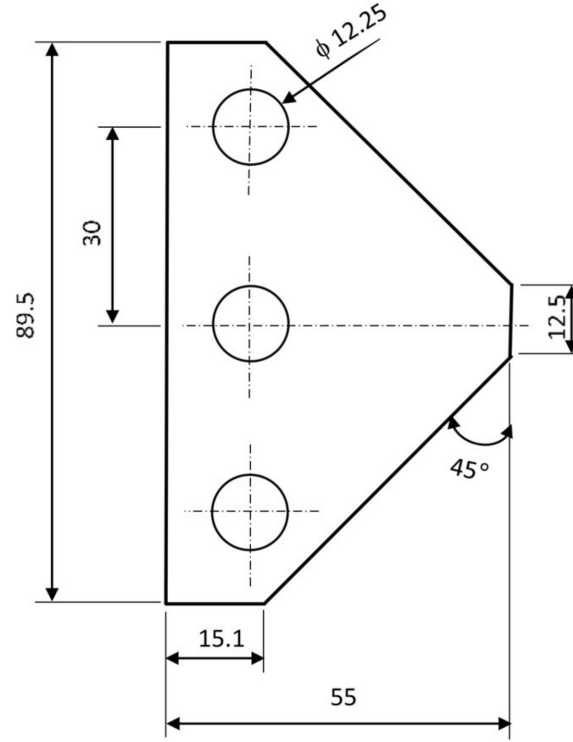


Figure 1: Arcan drawing, dimensions in mm.

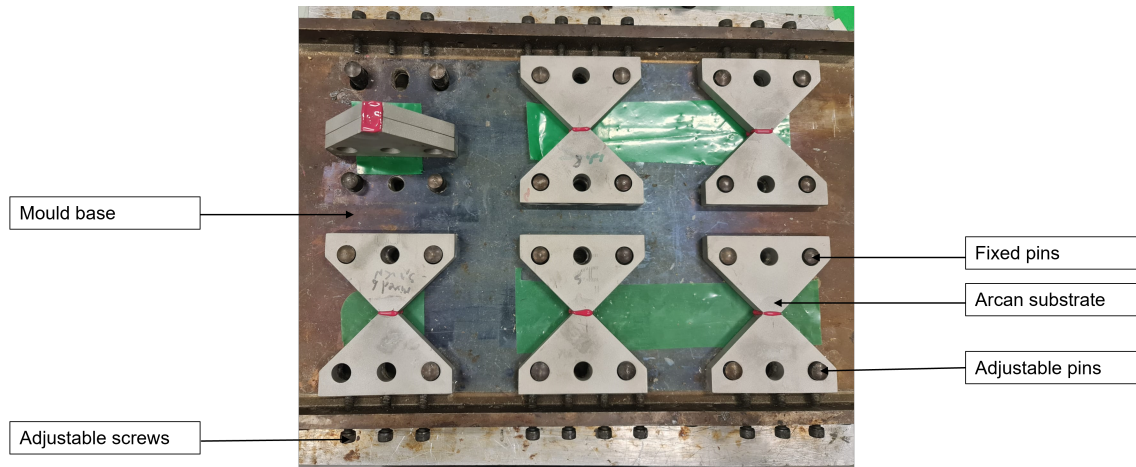


Figure 2: Mould for Arcan joints production.

To determine the adhesive strength degradation at different temperatures, a series of standard strength tests was conducted. Bulk tensile tests were performed according to the British standard BS2782. Accordingly, a specific mould was used to manufacture a 2 mm thick adhesive plate, which afterwards was cut to the final standard shape (see Figure 3) using a CNC machine. To evaluate the behavior of the adhesive under shear loading conditions, thick adherend shear tests (TAST) specimens were manufactured and tested. TAST joints were manufactured following the ISO 11003 2:2019 standard. To control the substrate alignment and the adhesive thickness during the curing process a specific mould, shown in Figure 4a, was

used. Figure 4b shows the specimen's dimensions. During testing, a digital image correlation (DIC) based technique was used to precisely measure the shear strain field. Accordingly, a speckled pattern was applied with paint, as shown in Figure 4c.

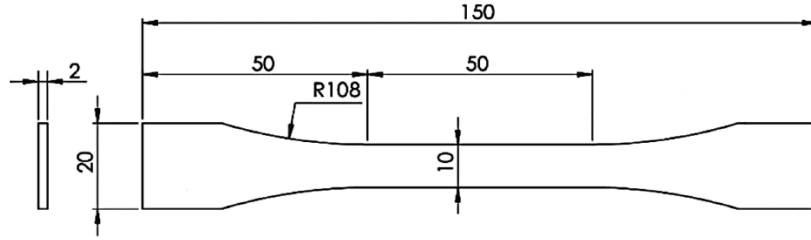
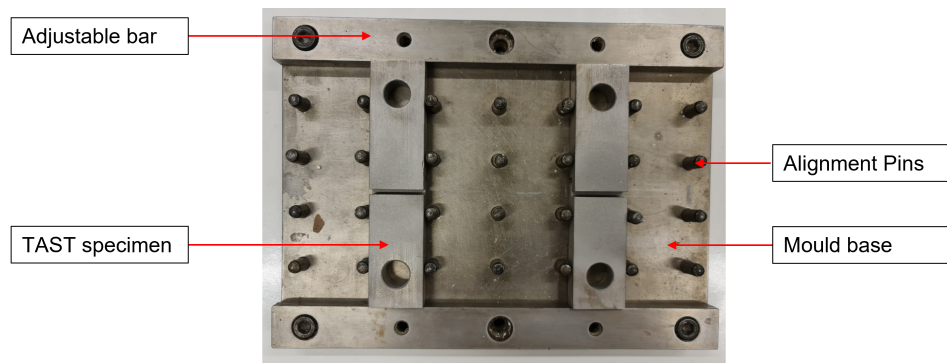
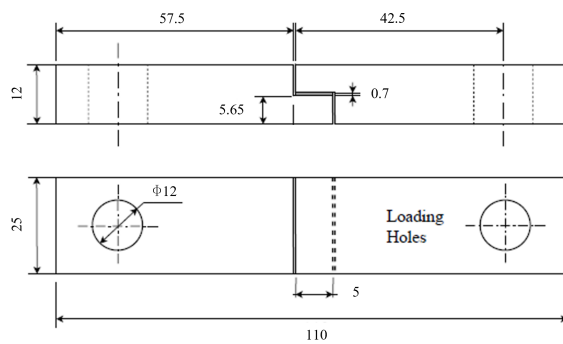


Figure 3: Tensile bulk specimen, dimensions in mm.

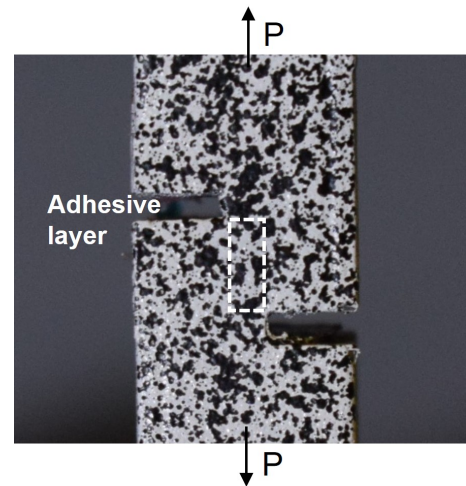
To evaluate the effect of temperature on the fracture energy of the adhesive for mode I and mode II, double cantilever beam (DCB) and end notched flexure (ENF) tests were performed, respectively. The specimens used for both tests are shown in Figure 5), where the only difference lies in the loading conditions, as the scheme in Figure 6 shows. The joints were produced in a specific mould to control the thickness and also to maintain substrate alignment during the test.



(a) mould used.



(b) joint scheme, dimensions in mm.



(c) speckle used for DIC.

Figure 4: TAST joint.

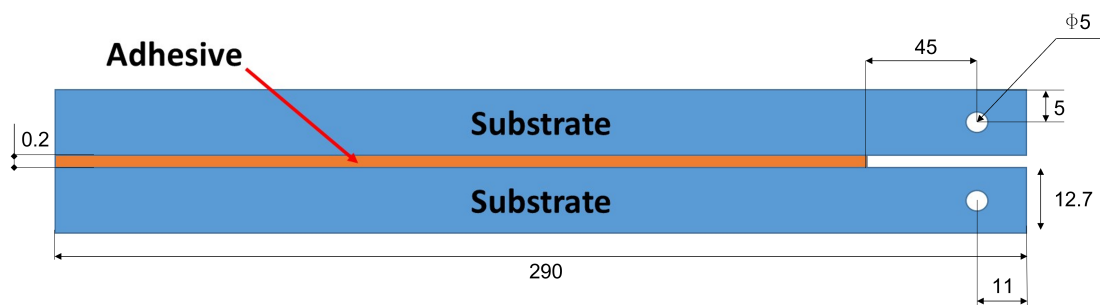


Figure 5: DCB/ENF specimen, dimensions in mm (not to scale).

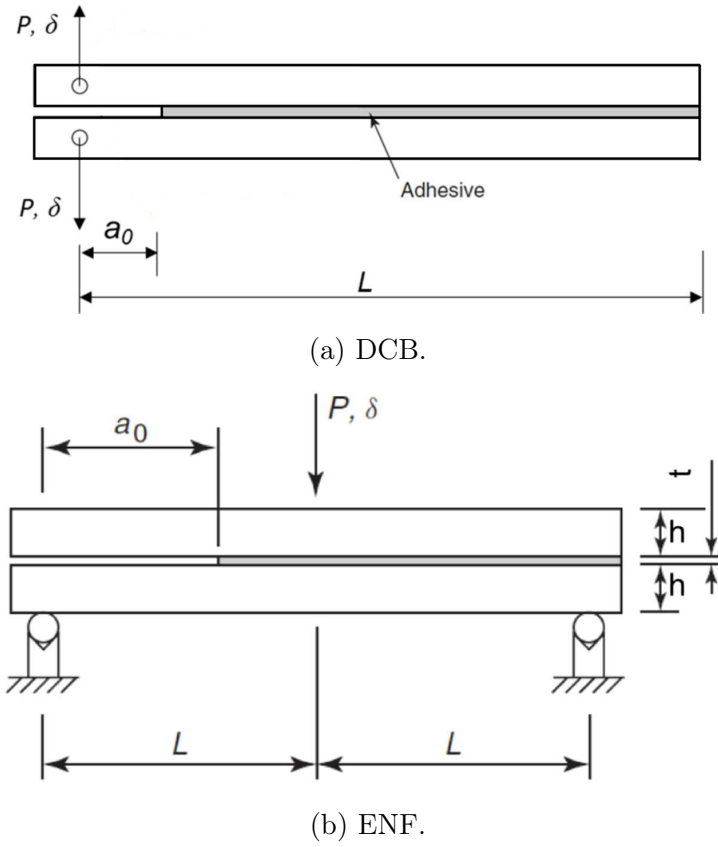


Figure 6: Schematic representation of fracture tests.

B.2.3 Curing cycle

The adhesive's curing cycle was studied as explained in Chapter B.4.1. The study aims to evaluate the effect of the temperature on the adhesive's behaviour, without the influence of the T_g . It is known that the curing cycle is the major parameter that influences the T_g , apart from the adhesive system itself. As a result, it was necessary to define a curing cycle that leads to a T_g well above the maximum testing temperature, which is 50 °C for this research. Three different curing cycles were studied, as shown in Figure 7. The first one (I), is the one proposed by the manufacturer in the data-sheet (14 days at room temperature), where no post-cure process is suggested. Then, a post-cure of 45 min at 80 °C was considered as the end of this curing cycle considered as Case II. However, since a 14 day long curing cycle is extremely time consuming, it was decided to reduce the curing time at room temperature to only 1 day (24 hrs), followed the posed curing procedure defined above. This final curing cycle was considered as Case III.

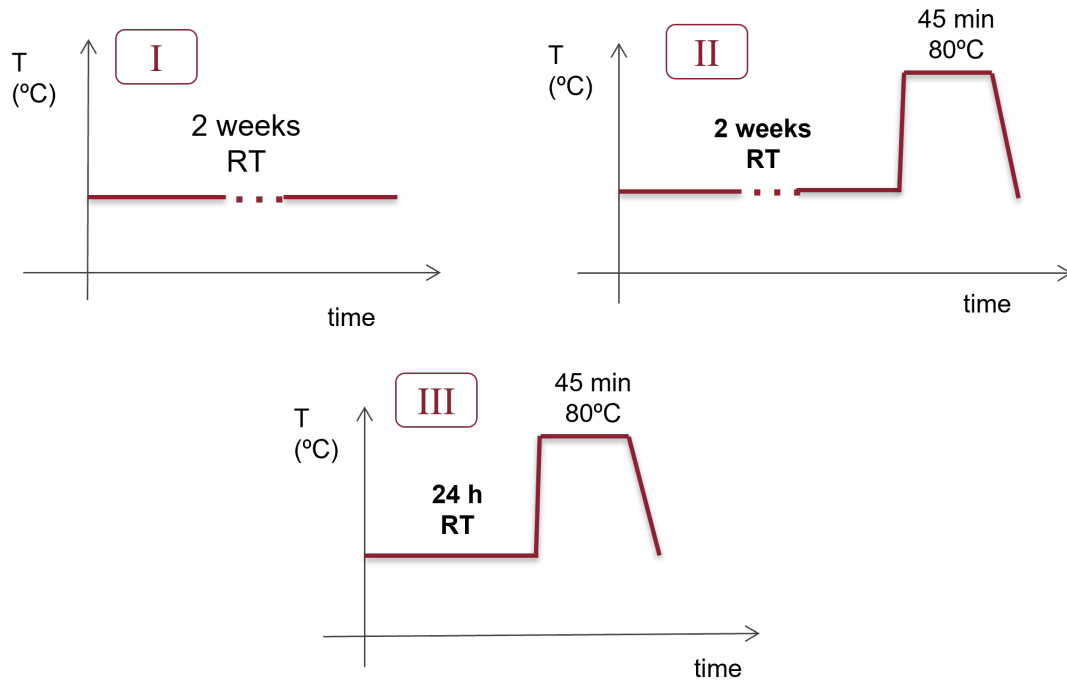


Figure 7: Different curing cycles tested.

Based on the results from this study, the curing cycle was defined as room temperature curing for 24 hours, followed by a post-curing process of 45 minutes at 80°C in an oven, which corresponds to the curing cycle number I from Figure 7.

B.2.4 Testing conditions

Strength and Arcan static tests were performed with a displacement rate of 1 mm/min, while for fracture tests the displacement rate was set to 0.2 mm/min. All joints were tested at room temperature and high temperature (50°C). For the fatigue tests, Arcan joints were tested using an Arcan assembly. The ratio between the maximum and minimum load, R ratio, was set to 0.1, and the load frequency fixed to 10 Hz. Tests were performed under different loading angles and several load levels. For the strength tests, a universal testing machine (INSTRON 3367) was used. All other joints were tested using a servo-hydraulic machine (INSTRON 8801). A thermal chamber was used to control and stabilize the temperature of the specimen during the high temperature tests. By using a thermocouple attached to the test specimens, test temperature was precisely monitored. Tables 2, 3 and 4 summarize the different test conditions considered in this study.

Table 2: Strength test conditions.

Test	Temperature
Tensile bulk test	Room temperature
	50 °C
TAST	Room temperature
	50 °C

Table 3: Fracture test conditions.

Test	Temperature
DCB, mode I	Room temperature
	50 °C
ENF, mode II	Room temperature
	50 °C

Table 4: Static and fatigue Arcan test conditions.

Static		Fatigue	
Mode mixity	Temperature	Mode mixity	Temperature
Mode I	Room temperature	Mode I	50 °C
	50 °C		
Mode II	Room temperature	Mode II	50 °C
	50 °C		

To analyze the shear and Young's moduli of the adhesives, the adhesive strain should be precisely measured during testing. Accordingly, an extensometer was used for tensile dogbone tests, while digital image correlation (DIC) was used for the TAST specimens. To process the images, a commercial software (GOM Correlate) was used. However, as shown in Figure 8 in Arcan tests the load is transferred through several connectors and components until it finally reaches the bondline. Each of these elements has its own stiffness and elongation and, accordingly, the displacement measured by the machine is not precise enough to be considered as the true displacement of the adhesive layer. Accordingly, only displacement values are reported for the Arcan tests.

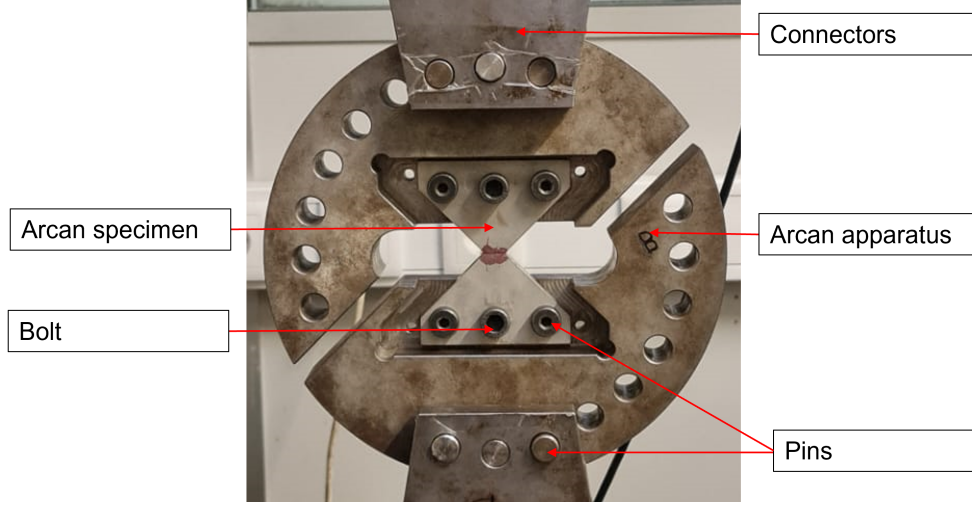


Figure 8: Arcan apparatus in pure mode I loading conditions.

B.3 Numerical Model

To analyse the stress distribution in bonded Arcan joints, a numerical analysis employing the finite element method (FEM) was conducted in Abaqus. The results are used to calculate the fatigue stress parameter considered in the master S-N curve. The proposed fatigue stress parameter is an extension of an already developed model [30]. The extended model can be used for fatigue life analysis of adhesive joints at high temperatures. This numerical analysis aims to obtain the maximum principal stress, the von Mises stress, and the hydrostatic stress distribution along the mid-plane of the bondline. A 2D linear elastic FEA was performed, which allows a short computation time. This type of analysis only requires two adhesive behaviour parameters: the Young's modulus, obtained from the bulk test and the Poisson's ratio obtained from the relation between the Young's modulus and the shear modulus. In the considered mesh configuration, two types of solid elements were used: 3-node (triangular shaped) plain strain elements for the substrates and 4-node plain strain elements for the bondline. Eight elements were used along the adhesive thickness and mesh refinement was carried out near the edges of the bondline. Figure 9 shows the mesh configuration considered in this analysis. The boundary conditions used aim to accurately describe the conditions found in the experimental setup. For the bottom holes, movement on both directions was restricted, while for the top holes the force is applied by associating reference points to the hole circumference.

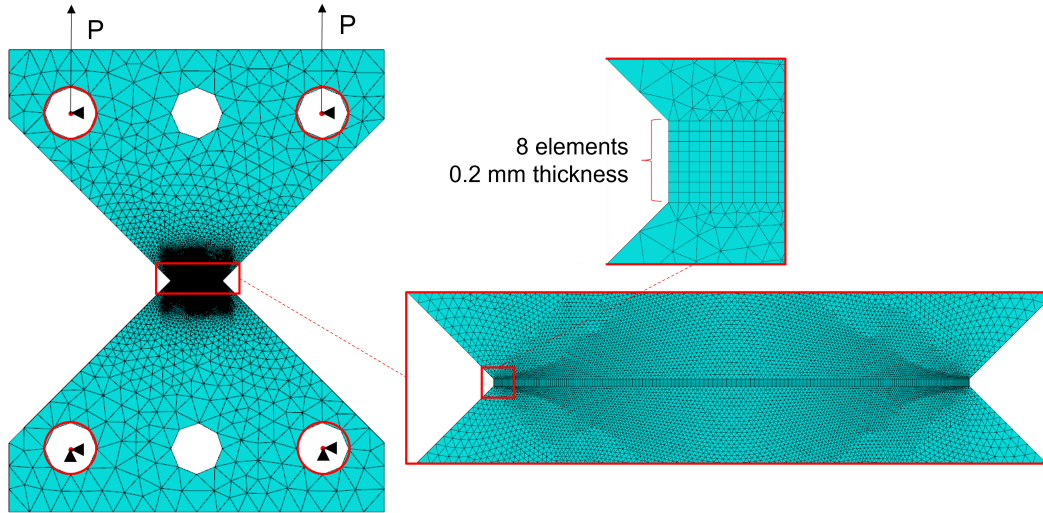


Figure 9: Arcan 2D FEA model loaded in pure mode I condition.

B.4 Results and Discussion

B.4.1 Effect of the curing cycle

The results showed that the T_g significantly changes by the curing cycle for this adhesive (see Figure 10). According to the results, the use of a post-cure is the main influencing parameter for the final T_g of the adhesive. In fact, by comparing Cases I and II, T_g was improved from 52 °C to 94 °C, representing 81% increase. However, by comparing cases II and III the results are very similar, indicating that the curing stage at room temperature does not play a significant role in the T_g of the tested adhesive. Based on these results, curing cycle I was discarded as it leads to a T_g very close to the testing temperature, 50 °C. On the other hand the T_g results for cases II and III are similar and consequently cases III can be the best curing cycle due to the very short cycle that the adhesive requires for cure in this case. However, except for the T_g the strength properties of the adhesive should be also analyzed as a function of the curing cycle. To analyze the effect of the curing cycle on the tensile behaviour, bulk tensile tests were performed for the 3 different cycles defined above. The results, presented in Figure 11, demonstrate that both stages of the curing cycle have a similar impact on the tensile strength of the adhesive. However, these results also show that the post cure process decreases the strain at failure of the adhesive, resulting in a lowered capacity for energy absorption. Based on these observations, the curing cycle selected to be used throughout the study was the third one (case III), since it was thought to represent a good compromise between the high T_g obtained and a short production time.

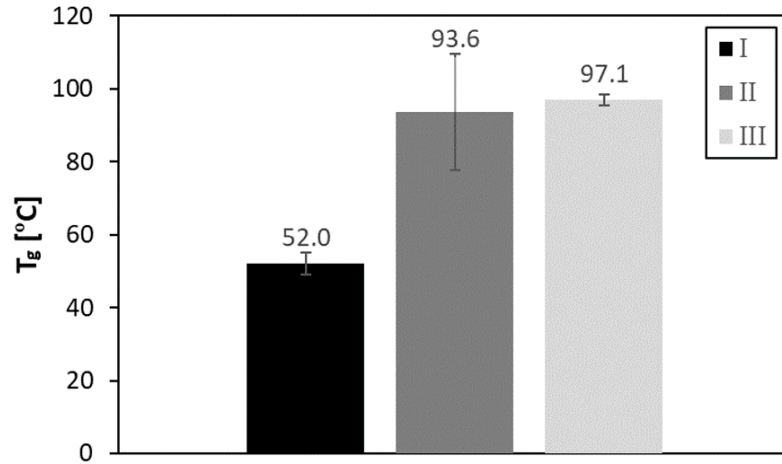


Figure 10: Effect of curing cycle on T_g .

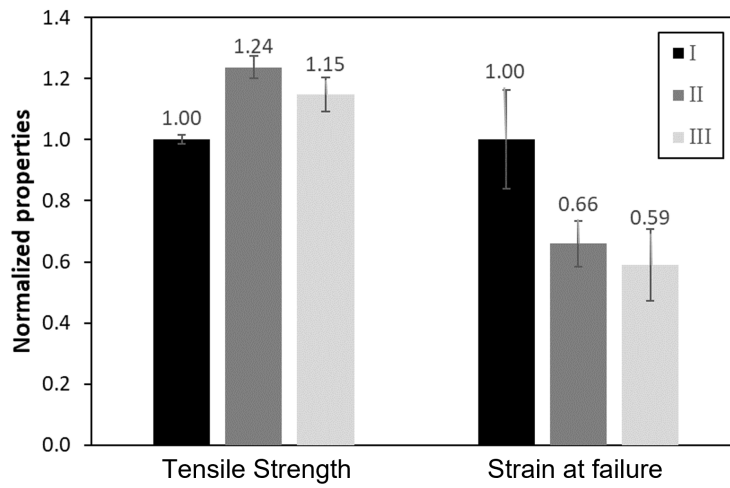


Figure 11: Effect of curing cycle on tensile strength and strain at failure.

B.4.2 Strength tests

Figure 12 shows typical stress-strain curves obtained for room temperature (23 °C) and 50 °C. The results showed that an increase of temperature by around 27 °C had a significant impact on the adhesive behaviour. The results (see Figure 13), showed a 37% decrease in the tensile strength, compensated with a 132% increase in elongation. When evaluating the stiffness, the degradation with temperature was 22%.

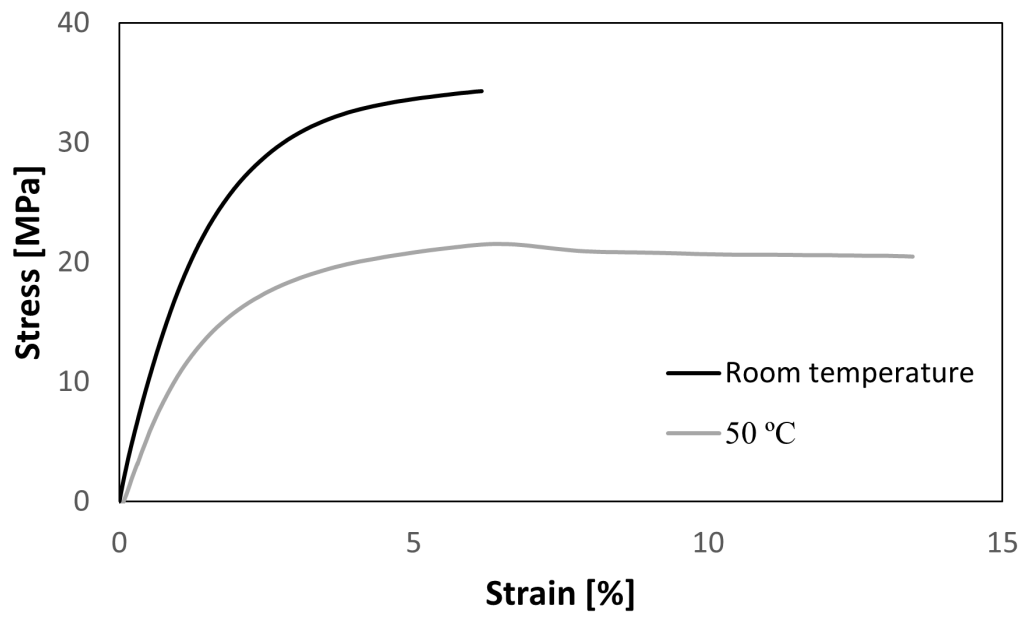
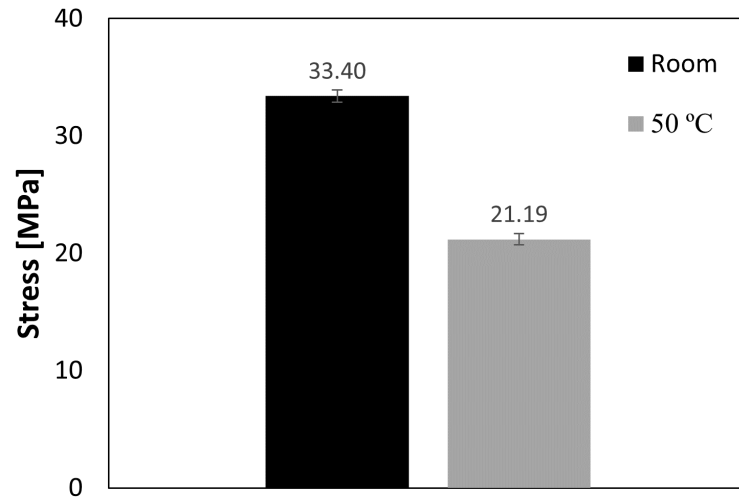
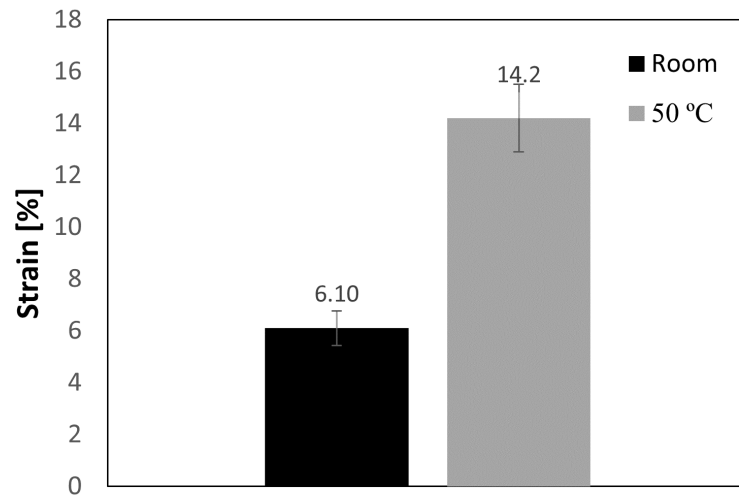


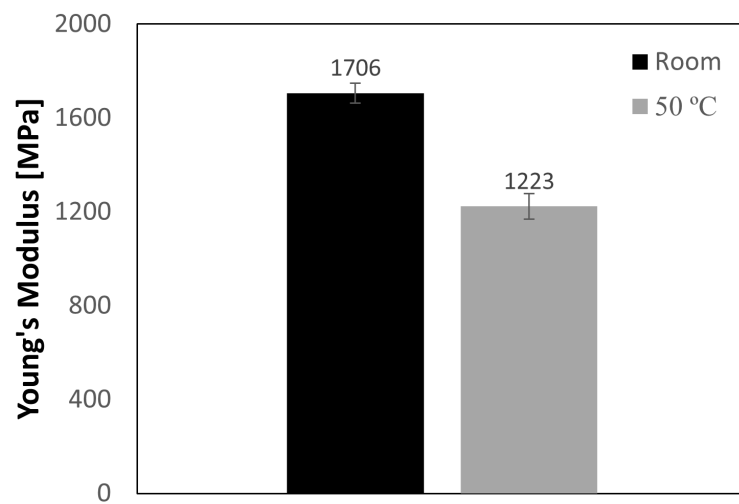
Figure 12: Stress-Strain curves for bulk tensile test for room temperature and 50 °C.



(a) Yield Strength.



(b) Strain at failure.



(c) Young's modulus, E .

Figure 13: Properties from bulk tensile tests for room temperature and 50 °C.

To analyze the rate of reduction in shear properties, TAST specimens were used. These results showed that the adhesive shear behaviour was not affected by the temperature as the tensile properties, as evidenced in Figure 14. In this analysis, the shear stress is obtained by dividing the applied load by the bonded area. To measure the strain, the relative displacement of both substrate obtained using DIC method was divided by the adhesive thickness. The results showed that the adhesive's strength and strain was not significantly affected by the testing temperature. On the other hand, the stiffness of the adhesive under pure shear conditions, as evaluated by the shear modulus, G , decreased by 31%. In what regards the failure mechanism, a fully cohesive failure was found.

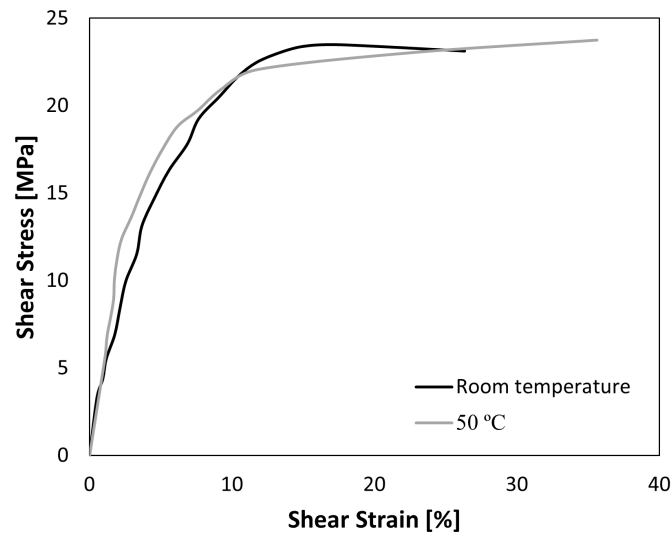
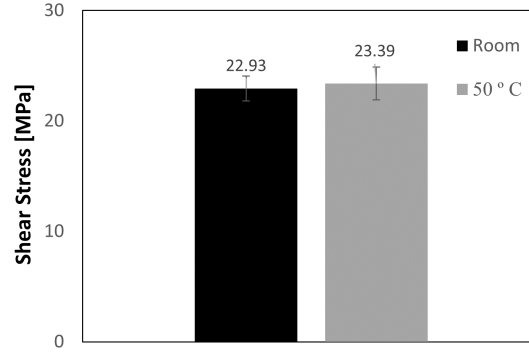
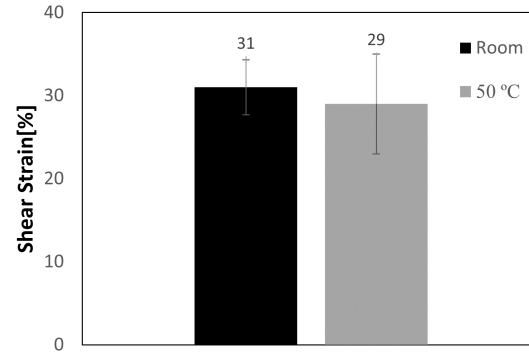


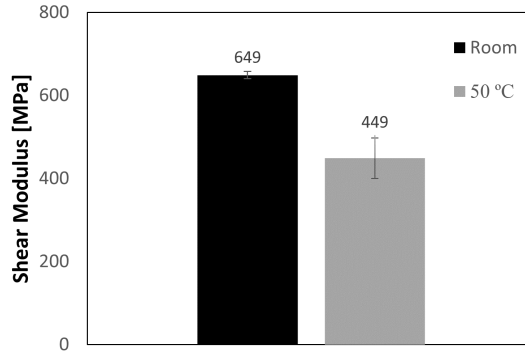
Figure 14: Stress-Strain curves for bulk tensile test for room temperature and 50 °C.



(a) Shear Strength.



(b) Shear strain at failure.



(c) Shear modulus, G .

Figure 15: Properties from bulk tensile tests for room temperature and 50 °C

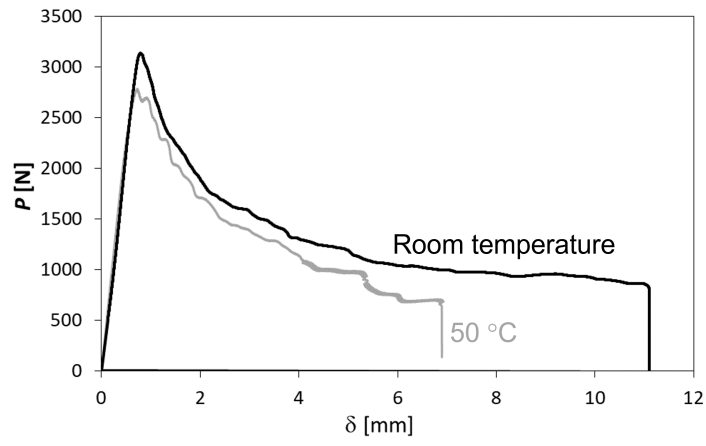
Strength tests showed that the adhesive is indeed significantly affected by the temperature, when loaded in a tensile condition, while for pure shear solicitation (TAST) its behaviour appears to be almost independent from the temperature. The Poisson's ratio's variation with the temperature was also analyzed by applying the values obtained from the strength tests on Equation 1. The results showed that the temperature increased this parameter by almost 20%. In an actual structure, this variation can have a significant influence on its integrity. A detailed discussion, regarding the different rate of degradation of adhesive properties as a function of loading mode is presented in Chapter B.4.5.

$$E = 2G(1 + \nu) \quad (1)$$

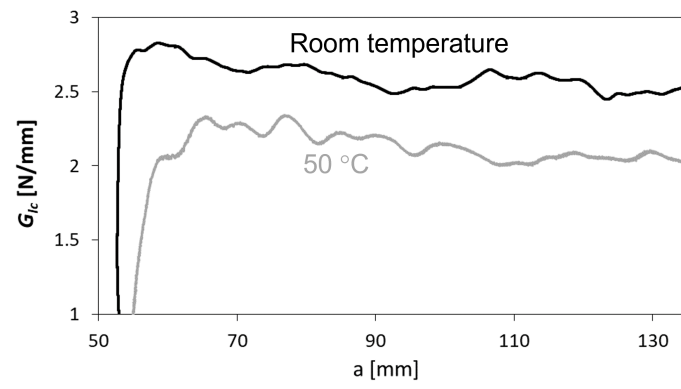
where E is Young's modulus, G shear modulus and ν Poisson's ratio.

B.4.3 Fracture tests

Fracture toughness describes the resistance of materials against the crack propagation. The influence of the temperature on the fracture behaviour of the adhesive was assessed by performing DCB and ENF tests. The results for mode I (see Figure 16) showed a 25% decrease in G_{Ic} , from 2.62 [N/mm] to 1.97 [N/mm]. Banea et al. [31] also already reported a decrease of mode I fracture energy with temperature, being this more severe when the temperature is close to T_g . Mode II tests, on the other hand, showed an increase of 27% of G_{IIc} with temperature (see Figure 17b). These results showed that loading mode significantly affects the response of adhesives to temperature, with mode I and mode II behaving differently in this case: pure mode I experienced fracture toughness degradation, while for mode II there was an improvement in the fracture energy. However, it should be noted that the fracture energy is a function of both displacement δ (or crack size) and load. DCB testing results showed a decrease in the maximum δ before failure at high temperature (see Figure 16a). Mode II R-curves (Figure 17b) showed that the stable crack propagation region, represented by the plateau, was delayed by increasing the temperature.

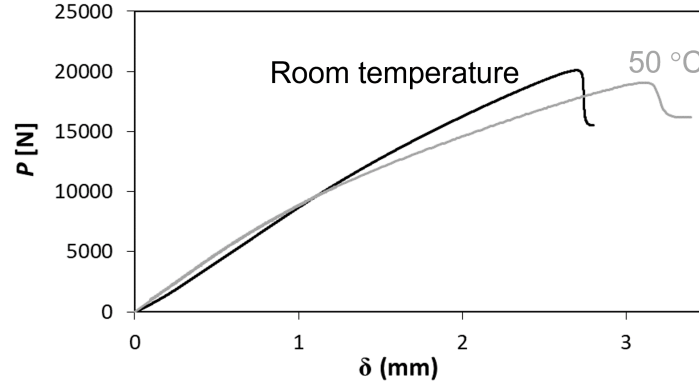


(a) P - δ curve.

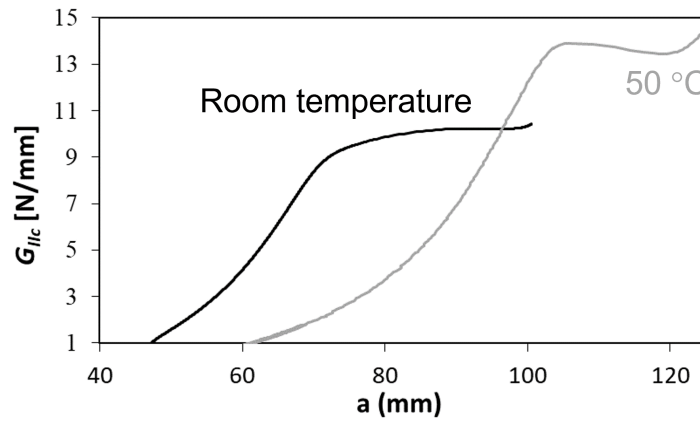


(b) R-curve.

Figure 16: DCB tests results for room temperature and 50°C



(a) P- δ curve.



(b) R-curve.

Figure 17: ENF tests results for room temperature and 50°C

B.4.4 Arcan tests

Static tests

Static Arcan tests were performed in pure mode I and mode II to investigate if temperature affects the rate of degradation at different loading angles. Results also helped defining the load levels for the fatigue tests. Those tests were repeated for both temperatures under study (room temperature and 50 °C). Load-displacement curves were generated directly from the data of the testing machine; Figures 18 and 19 show typical curves for the different temperatures. Results showed that the influence of temperature on joint strength is more severe when the joint is loaded in pure mode I. Based on these results, the influence of the temperature on the displacement at maximum load was found to be similar for both loading angles: for mode I, the increment of temperature resulted in a 13% increase on the displacement, while for mode II the increase was 15%.

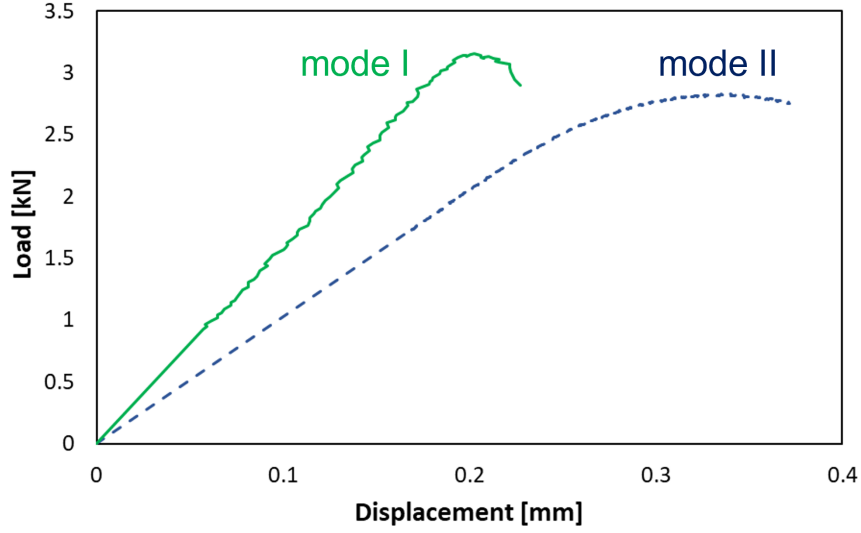


Figure 18: Load-displacement curves for Arcan tested at room temperature.

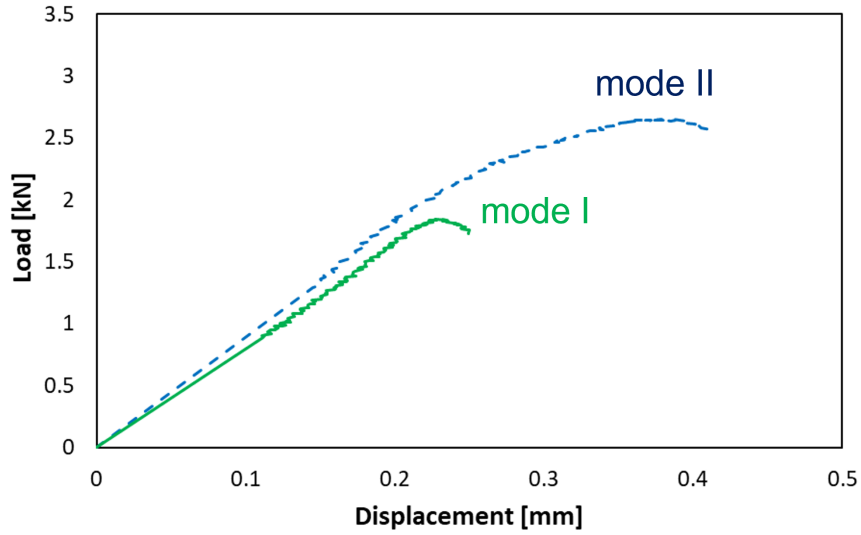


Figure 19: Load-displacement curves for Arcan tested at 50 °C.

Stress values were simply calculated by dividing the load by the bonded area. The results showed, in a manner consistent with the strength results in Chapter B.4.2, that in mode I the strength decreases significantly with the temperature, even though the temperature is still far from the T_g , with a reduction of 48%. On the other hand, for mode II, the reduction was only 2%, which can be considered as almost negligible.

The fracture surface analysis of the static tests (see Figure 20) showed similar failure modes for mode I for both testing temperatures. However, for mode II, where shear phenomena are present, the roughness of the joint tested at 50 °C is higher than the one tested at room temperature. The roughness in mode II is related to

the hackles phenomenon, a common failure mechanism when a material is subjected to shear loads.

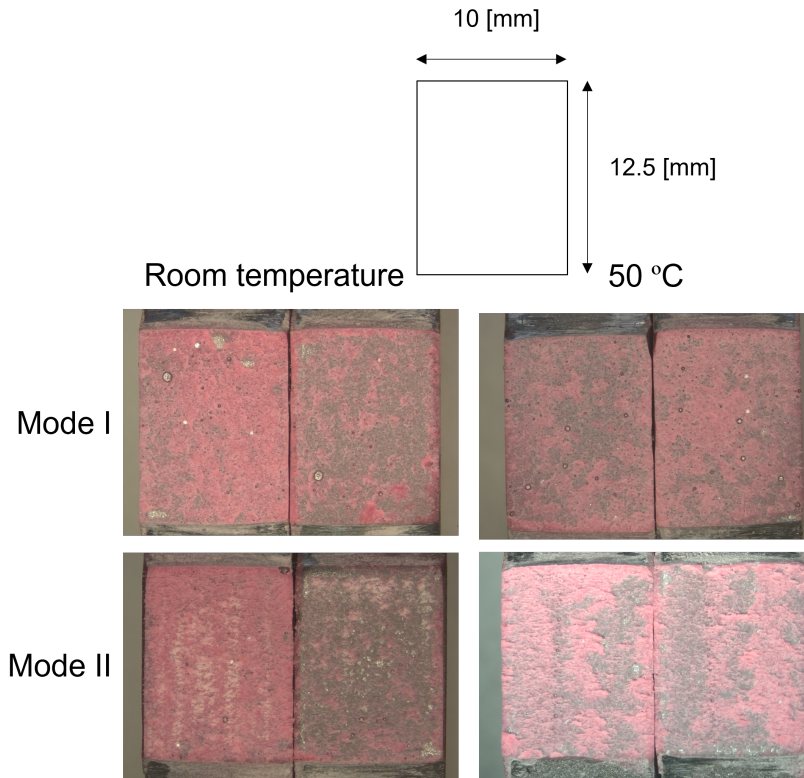


Figure 20: Fracture surfaces of Arcan joints tested statically.

Fatigue tests

Different load levels and mode mixities were considered for the fatigue tests. Stress-life (S-N) curves were constructed using the experimental results obtained (see Figure 21). Results showed that at 50 °C, mode II is expected to have higher fatigue strength; as an example, a specimen loaded with 2000 N is expected to fail after 1 cycle for mode I, while for mode II it should last for around 1000 cycles, which is three orders of magnitude above. Also the rate of change in life with fatigue load of the two curves is similar, indicating that the sensitivity to temperature is identical for both mode mixities when the loading condition is cyclic. In other words, that means the same percentage of load reduction leads to the same increase in joint life.

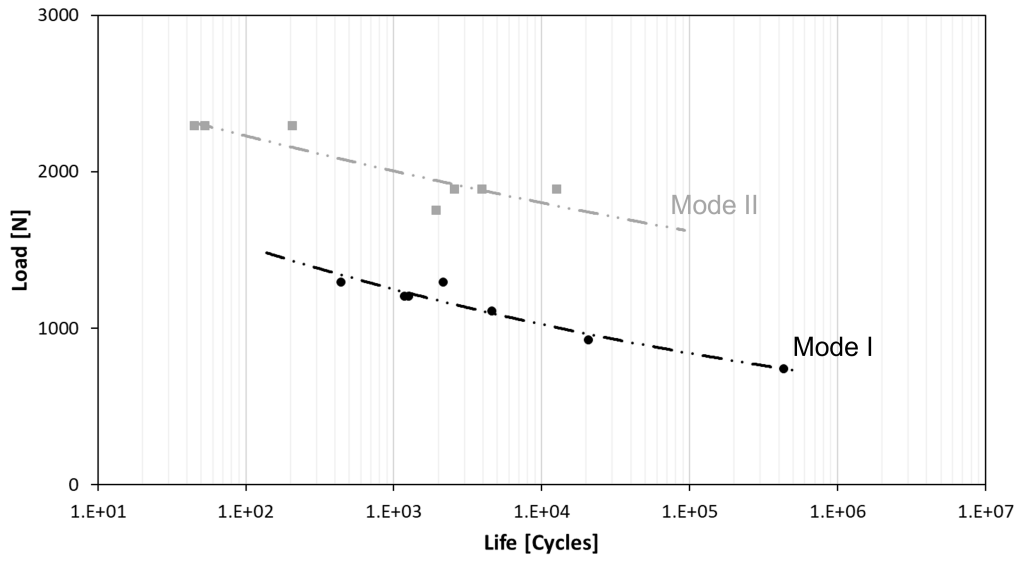
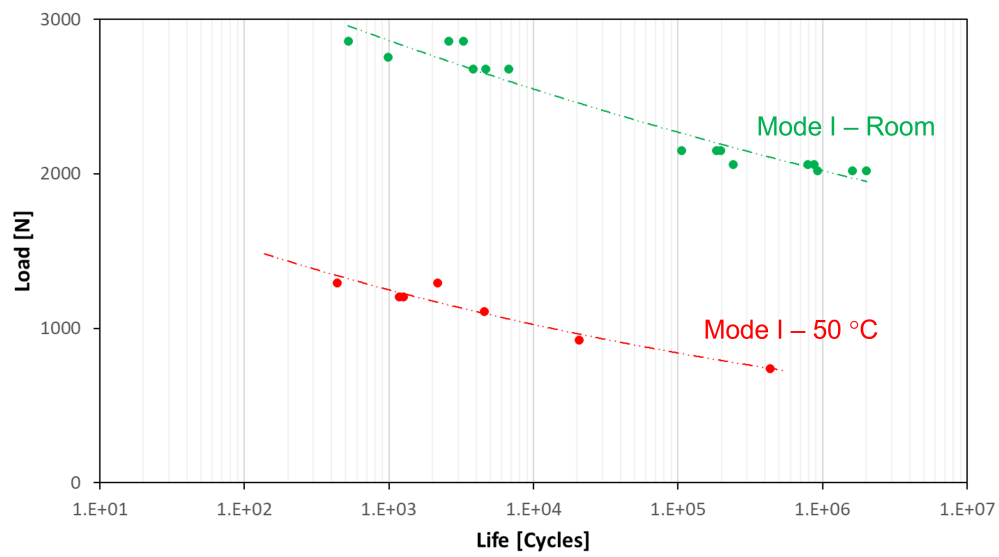
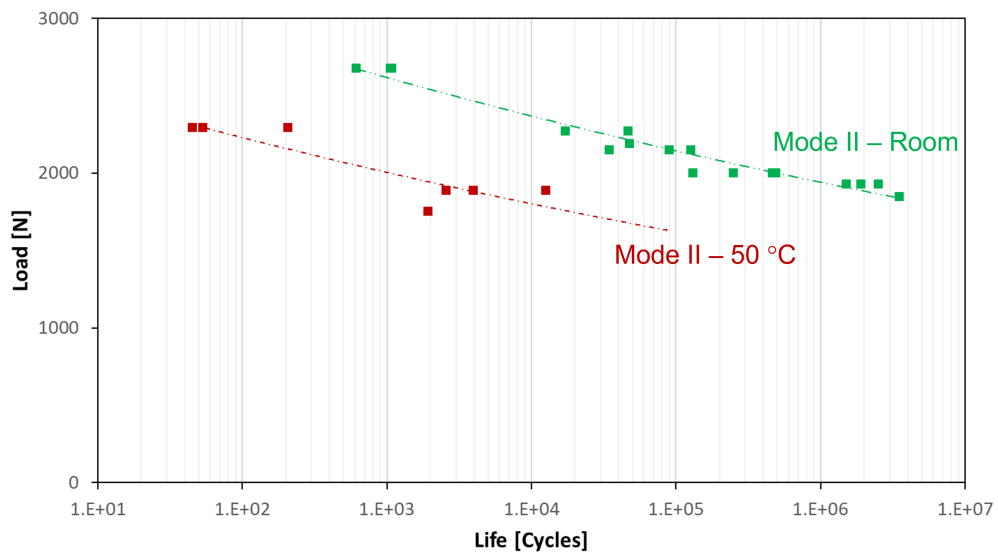


Figure 21: Stress-life (S-N) curves for Arcan tested at 50 °C.

In a previous study by Antelo et al. [32], where fatigue tests were performed in Arcan joints at room conditions, it was concluded that the loading angle does not play a significant role on the strength of this adhesive (see Chapter B.4.1). However it should be mentioned that the authors used curing cycle Case I (14 days curing at room temperature), even though no significant difference in joint strength was found as a function of the curing cycle. The results of Antelo et al. [32] for room temperature conditions were compared with high temperature fatigue results obtained in this study (see Figure [24]). According to these results, for mode I, the degradation in fatigue properties is much greater than for pure mode II. It is also important to note that although the static strength of Arcan joints for mode II is almost independent of the temperature, for fatigue loading conditions the specimens tested at high temperature do experience a lower fatigue life. These results suggest that higher temperatures lead to an increased sensitivity to cyclic loads, although the static results do not show such a significant dependency on the temperature. Figure 23 shows small zones of adhesive failure in pure mode II, while for mode I fatigue tests the failure mechanism exhibits a fully cohesive nature.



(a) Mode I.



(b) Mode II.

Figure 22: S-N curves for room temperature [32] vs. 50 °C.

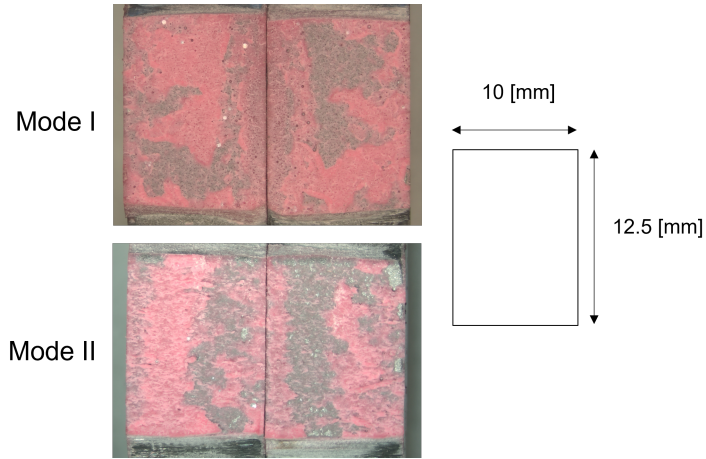
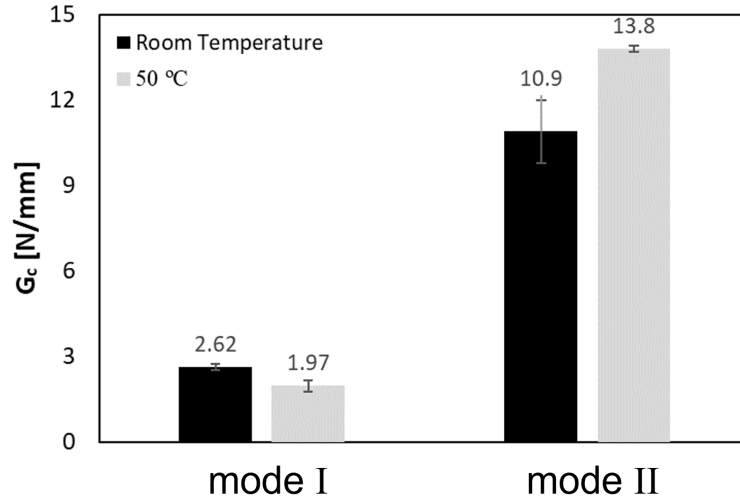


Figure 23: Fracture surfaces of Arcan joints tested under fatigue.

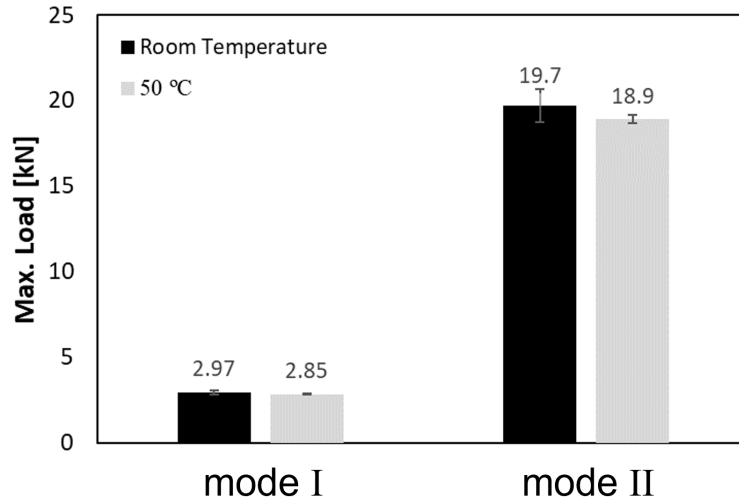
B.4.5 Effect of temperature on the rate of property degradation

According to the results presented in the previous sections, it can be concluded that mechanical property degradation is a function of three parameters, including temperature, loading type, and loading angle. Results from the strength tests, fracture tests, Arcan static tests and also the Arcan fatigue tests were all considered to study this effect. Both room temperature and 50 °C conditions were considered and for all testing techniques different loading modes were tested. The results showed that the way that temperature affects each property is different and should be individually analyzed.

Fracture tests highlighted the most significant difference between mode I and mode II: for mode I the property degradation was noticeable for high temperature, with fracture toughness decreasing by 25% and the maximum load being reduced by 13%. On the other hand, for mode II, the fracture toughness was not reduced and in fact it increased 27% with temperature. Thus, no significant reduction in load was found for both mode I (DCB) and mode II (ENF) tests. However the temperature mainly affect the maximum displacement, especially for DCB fracture tests.



(a) Fracture toughness.



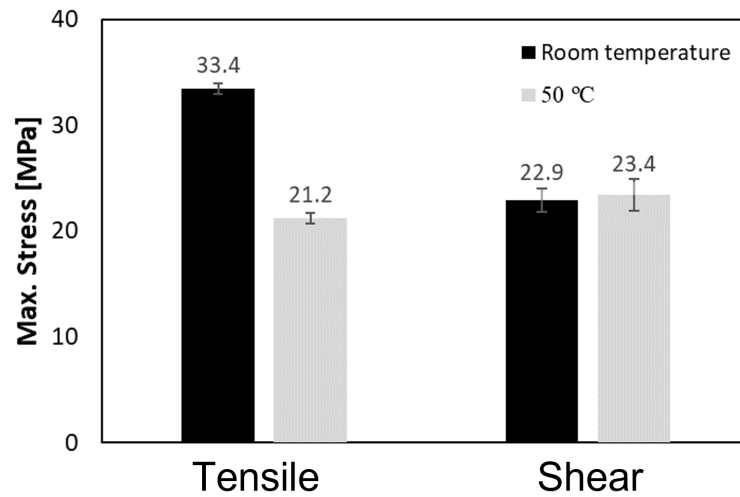
(b) Maximum load.

Figure 24: Results and properties from fracture tests at different temperatures.

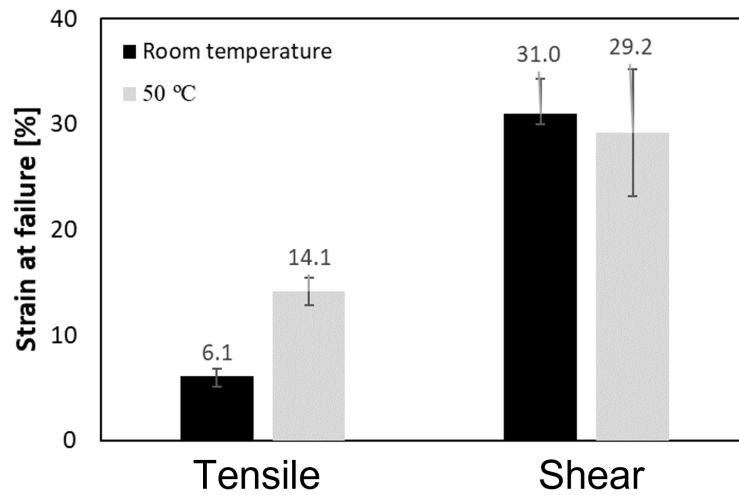
Testing Arcan joints in tensile and pure shear conditions, led to mechanical properties which are consistent with those obtained in dogbone and TAST tests, respectively. However, some important considerations must also be made here. Firstly, when comparing bulk tensile test and TAST, effects other than mode mixing must be considered. For example, bulk testing does not consider the effects of substrates, and as a result, at high temperatures, the role of substrates and residual stresses that may be induced in joints are not properly considered. However, Arcan joints allow to test the exact same joint under different pure mode conditions, and consequently this test can be a better evaluator of the effect of load angle at high temperature for an adhesive located within a joint.

Figure 25 summarizes effects of temperature as a function of temperature for strength tests. These results show that the strain at failure is significantly affected

by temperature, with a variation from 6.1% for room temperature to 14.1% at 50 °C; however, for TAST, the shear strength and strain were not significantly affected by the temperature, with both strength and strain presenting similar values (variations of less than 5%). The effect of mode mixity can be better described by Arcan tests. Results presented in Figure 26 showed that mode I is more sensitive to temperature variations than mode II, with a 45% strength reduction on mode I from 27.3 to 14.9 MPa compared with a 14% strength variation for mode II, 25.1 to 21.7 MPa.



(a) Yield Strength.



(b) Strain at failure.

Figure 25: Effect of temperature on the tensile and shear strength properties.

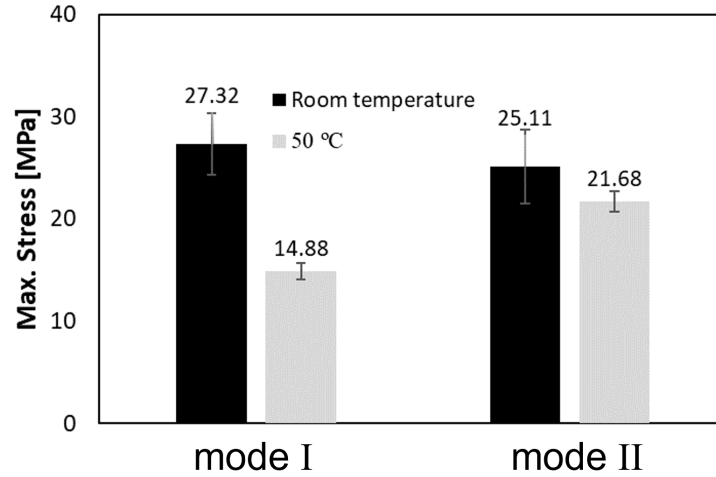


Figure 26: Effect of mode mixity and temperature on Arcan static strength.

Fatigue tests (Figure 21 and 22) showed that the temperature does not influence the rate of property degradation as a function of the fatigue life (cycles). In fact, even though the S-N curves for high temperature are below those defined at room temperature, this difference is exclusively related to the loss in the static strength of the joints. On the other hand, mode mixity affected the fatigue behaviour of Arcan joints: results showed fatigue strength as a function of the static strength is higher for mode II than for mode I for both temperatures tested.

As Figure 27 shows, mode I loading conditions lead to a severe property degradation when compared with tests carried out under mode II. In fact, mode II showed almost less sensitivity to the temperature increment, with 2 out of 3 test types even increasing adhesive's properties. By comparing the different tests over each mode mixity, the results obtained for mode I/tensile evidenced degradation that is higher in Arcan joint, with the increase of temperature almost decreasing the joint's strength to half, followed by the bulk tests, where temperature reduced almost 40% the tensile strength. Finally, DCB testing was less affected, with a reduction of the fracture toughness in mode I of just 23%. Under mode II, Arcan joints showed a 19 % degradation. The TAST strength was not affected by temperature and, finally, G_{IIc} increased over 20% with the increase in temperature, which can be related to the reduction of the stiffness of the adhesive at higher temperatures.

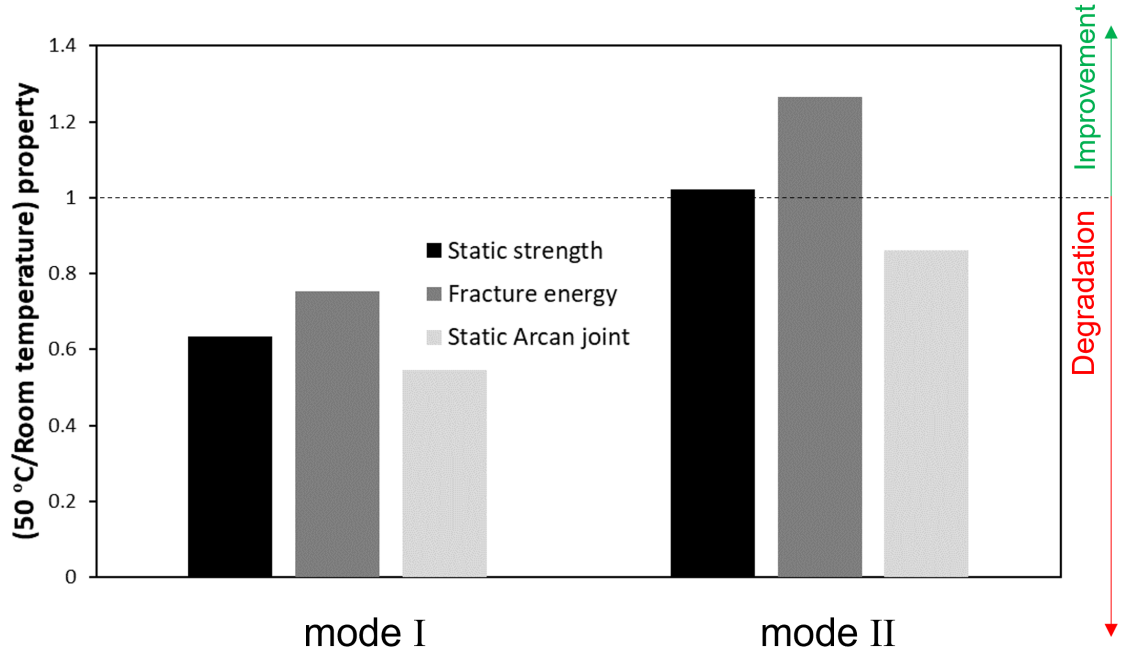


Figure 27: Influence of temperature on adhesive properties for different tests.

B.4.6 Master curve and fatigue life prediction

Fatigue life of an adhesive is usually divided into two parts: crack initiation and crack propagation. The first one, is dominant for joints under lower load levels where elastic deformations are dominant, resulting in a high cycle fatigue life which is the most typical fatigue condition in real life applications. In opposition, for joints where the crack propagation is dominant, the load levels are closer to the static strength. With that in mind, there are three main different approaches to predict the fatigue life of a joint. The first are known as total-life methods, referred as stress-life (S-N) approach. The second group of methods are the fracture-mechanics based, and they focus on the crack propagation phase. Finally, the third group follows damage mechanics approaches, considering both crack initiation and propagation, via the introduction of a damage parameter, D [1,33]. Since this is a highly crucial research topic, some authors have carried out reviews over the state of the art on fatigue in adhesive joints [33–35].

The objective of this component of the current work was to validate the model proposed by Sousa et al. [30] for fatigue lifetime prediction for a different adhesive system using a master curve, and mitigate a known flaw of that model which, by not considering the effect of temperature, only was shown to only work for room temperature. To do so, a new parameter, defined as corrected effective stress, σ_{eff} , was defined.

Sousa et al. [30] proposed a fatigue life prediction model based on a specific parameter, defined as "effective stress". The model proposed by the authors made

possible to eliminate the effect of loading angle, by combining 3 different mode mixities (mode I, mode II and mixed mode (45°)) in one single master-curve. In their model, the number of cycles was plotted as a function of " A new effective stress composed by parameters that play a key role in the fatigue behaviour of adhesive joints." (see Equation 2). The effective stress is considered at the location where maximum principal stress is maximum; values of effective stress were calculated numerically as explained in the study [30].

$$\sigma_{\text{eff}} = \sigma_{\text{VM}} + (\sigma_{\text{H}} \times S) \quad (2)$$

being S the stress triaxiality defined as the ratio between hydrostatic stress and von Mises stress ($\frac{\sigma_{\text{H}}}{\sigma_{\text{VM}}}$).

Corrected effective stress, σ'_{eff}

The fatigue life of a joint is highly dependent on its static strength. At the same time, its static strength depends on many factors, which include the loading angle and the temperature. Fatigue results showed that the main factor affecting fatigue life at different temperatures is the static strength itself.

As a result, a new corrected effective stress, σ'_{eff} , is proposed as Equation 3 where the effective stress suggested by Sousa et al. [30] is multiplied by the ratio between the strength of the joint at a considered temperature and mode mixity to the strength of the same joint tested at room temperature:

$$\sigma'_{\text{eff}} = \sigma_{\text{eff}} \times \frac{\sigma_{\text{M},T}}{\sigma_{\text{M},\text{Room}}} \quad (3)$$

M is mode mixity and T is the considered high temperature.

Master curve construction

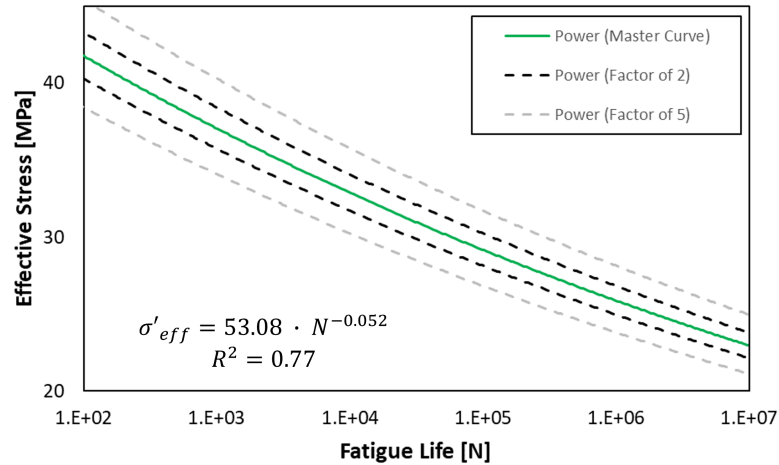
To predict the fatigue life of the joints subjected to mixed mode loading conditions, two analysis steps should be followed. The first step is to obtain a master curve. To achieve this, and based on the procedure defined by Sousa et al. [30], the effective stresses for three load levels for pure modes I and II conditions needs to be first determined. This procedure was carried out two times: first for room temperature results [32] and then, for 50 °C. To calculate the corrected effective stress in the numerical model, the corresponding moduli was considered based on the strength tests. Table 5 show the reference points used to build the master curves.

Table 5: Reference points used for the master curve and Basquin's constants.

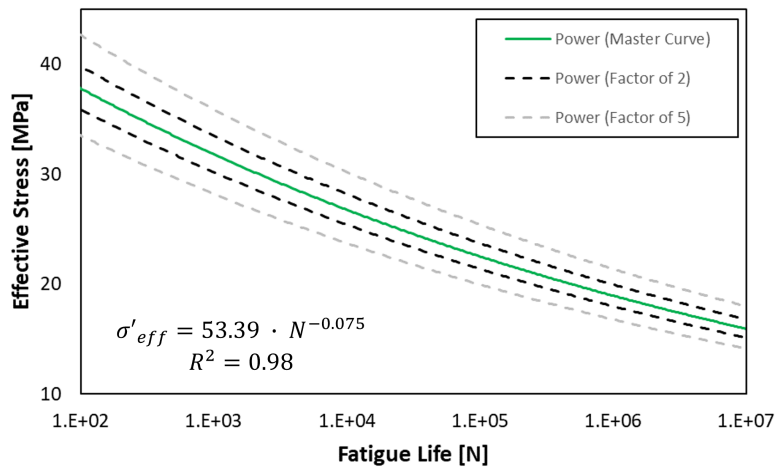
	Mode I		Mode II		Basquin's constants	
	Life (cycles)	σ'_{eff} (MPa)	Life (cycles)	σ'_{eff} (MPa)	α	β
Room temperature	2130	34.6	913	39.1	53.08	-0.052
	5068	31.9	31998	33.1	$R^2=0.77$	
	162695	25.6	333145	29.1		
50 °C	1221	32.8	4031	28.3	53.39	-0.075
	20675	25.2	101	37.1	$R^2=0.98$	
	433859	20.2				

Afterwards, a curve fitting is necessary and thus a Basquin relation was implemented (see Equation 4). Results presented in Figure 28 showed a good fit between the curve to the experimental results.

$$\sigma'_{eff} = \alpha \times N^{\beta} \quad (4)$$



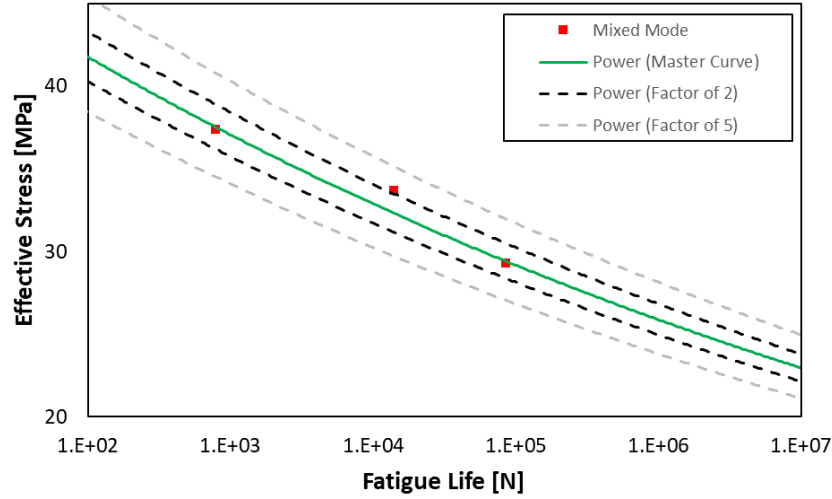
(a) Room temperature.



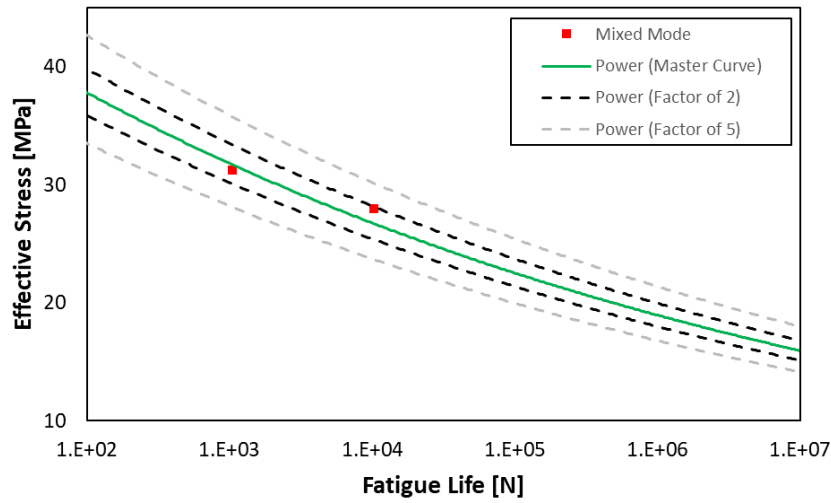
(b) 50 °C.

Figure 28: Fatigue life prediction curves for different temperatures.

Finally, the mixed mode points were plotted together with the respective master curves obtained. As Figure 29 shows, experimental results showed good agreement with the proposed curve, suggesting that such a model is suitable for use with different temperatures.



(a) Room temperature.



(b) 50 °C.

Figure 29: Comparing the predicted and the experimental experimental fatigue lives for mixed mode (45°).

Effect of temperature on master curve

Figure 30 shows, the curves start from similar points for one cycle and, for longer lives the curves tend to diverge. This results show that it might be possible to find a relation between the curves for different temperatures and. However, to investigate this, fatigue results for a wide range of temperatures are needed which is out of the scope of the current study. One important consideration is the fact that this model was only tested for temperatures safely below T_g and thus the presented model

might not be accurate outside of those conditions.

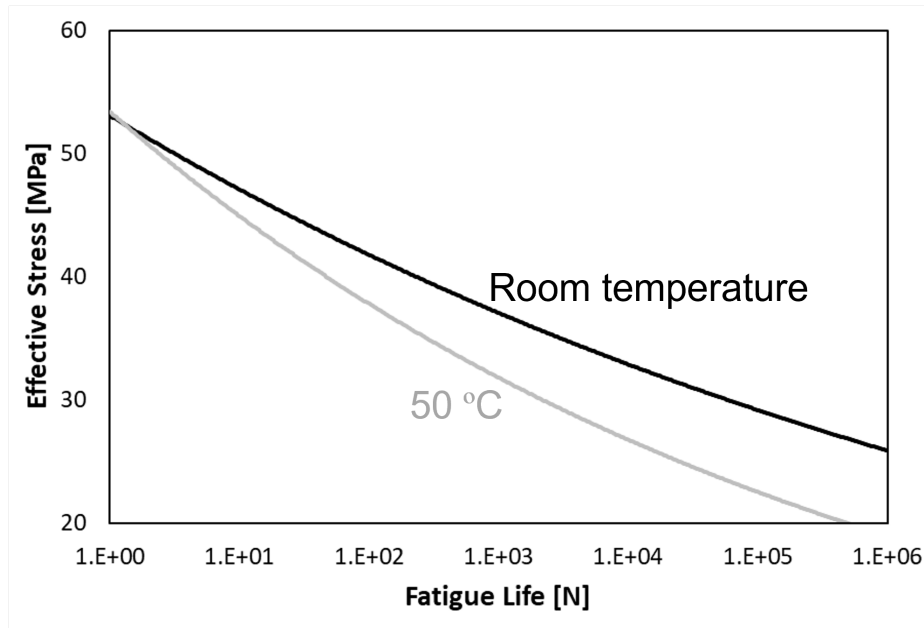


Figure 30: Comparison of the predictions curves for different temperatures.

B.5 Conclusions

The effects of service temperature and mode mixity on the degradation of the mechanical properties of an epoxy-based adhesive were studied. To do so, different tests were conducted, including strength and fracture tests. Arcan joints were also tested statically under pure modes I and II. Fatigue tests using the same Arcan joints enabled the construction of S-N curves.

Experimental results showed that the post-cure temperature is the most significant parameter that governs that glass transition temperature of the adhesive. On the other hand, the presence of a post-cure at high temperature leads to a decrease in the elongation of the material.

When subjected to high temperatures, the adhesive experienced important changes in its properties. Results showed that, under tensile/mode I load conditions, the effect of temperature is quite significant, while under pure shear/mode II the adhesive is practically insensitive to the different temperatures considered in this study. Static Arcan results showed that the increase of the temperature from room conditions (23 °C) to 50 °C resulted in a 48% decrease in mode I strength, while for mode II the reduction was only of 2%. Fracture tests showed the fracture toughness is not so affected by temperature. G_{Ic} decreased 25% while G_{IIc} improved by 27%.

Fatigue Arcan tests, for those carried out under static conditions, showed again that the adhesive works better under pure mode II conditions at high temperature. According to the S-N curves obtained, fatigue strength as a function of the static strength (%) is higher for mode II than mode I. Finally, the proposed fatigue model

was able to predict the fatigue life of Arcan joints subjected do different mode mixities and temperatures.

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